Thermodynamic Considerations in Quantum Information Processing

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"Thermodynamics is a funny subject. The first time you go through it, you don't understand it at all. The second time you go through it, you think you understand it, except for one or two points. The third time you go through it, you know you don't understand it, but by that time you are so used to that subject, it doesn't bother you anymore."

Arnold Sommerfeld [1, p. 1]

"Do you guys just put the word 'quantum' in front of everything?"

Scott Lang (Paul Rudd), *Ant-Man and the Wasp* (00:33:44), Peyton Reed (director), Walt Disney Studios Motion Pictures (distributor), 2018

"Eine Heizung funktionert so: zuerst nimmt man die Kälte, und schmeißt sie in den Mistkübel. Dann nimmt man Papier, und daraus wird dann Kohle, und die wird dann zu Feuer. Und dann kommt die Feuerwehr und löscht."

(Translation "A radiator works like this: first you take the cold and throw it in the the dustbin. Then one takes paper, which becomes coal, and that then becomes fire. Then the fire fighters come and extinguish it.")

Noah Friis (3 years old, private communication, 8 August 2021)

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Contents

Part I

From Thermodynamics to Quantum Information – and Back

I.1 Introduction

While the historical roots of modern thermodynamics go back to the nineteenth century and the foundational works of renowned figures such as *Sadi Carnot*, *Rudolf Clausius*, *William Thomson* (Lord Kelvin), *James Clerk Maxwell*, *Ludwig Boltzmann*, *Max Planck*, and *J. Willard Gibbs* (amongst many others, for an overview, see, e.g., the introductory textbook Ref. [2]), the origins of quantum information can only be traced back to the late twentieth century. Although some seminal works, in particular, by *Albert Einstein*, *Boris Podolsky*, and *Nathan Rosen* [3] as well as by *Erwin Schrödinger* [4], identified entanglement as a crucial feature of quantum theory already in 1935, the importance of these works was only widely recognized much later, most notably by *John Stewart Bell* in 1964 [5], whose famous inequality (or variants thereof, e.g., due to *Clauser*, *Horne*, *Shimony* and *Holt* [6]) was subsequently tested in 1972 by *Freedman and Clauser* [7] and throughout the 1980s by *Aspect*, *Grangier*, *Roger*, and *Dalibard* [8–10]. In parallel, first concerted efforts towards quantum communication protocols (by *Wiesner* [11], and *Bennett* and *Brassard* [12]), quantum computing (by *Bennett* [13], *Poplavskii* [14], *Benioff* [15], *Feynman* [16], *Toffoli* [17], *Deutsch* [18], *Igeta* and *Yamamoto* [19], and *Milburn* [20]), and the general information-theoretic understanding of quantum mechanics (e.g., by *Holevo* [21], *Ingarden* [22], *Park* [23], *Dieks* [24], *Wootters* and *Zurek* [25], and *Werner* [26]) were made throughout the 1960s, 1970s, and 1980s, before the field of quantum information and computation decisively built up momentum in the 1990s, with a first major textbook (which is still of high relevance today) was published by *Nielsen* and *Chuang* [27] in 2000. What is interesting to note here is that some of the earliest contributions to the theory of quantum computing, namely Poplavskii's article [14] entitled (in its English translation) '*Thermodynamic models of information processes*' as well as Bennett's paper [28] '*The thermodynamics of computation – a review*', explicitly reference thermodynamics as centrally important for considerations about quantum computers. Indeed, the relevance of thermodynamics is not limited to *quantum* information and computation but more broadly to *information theory* in general. This is witnessed, e.g., by the impact on modern quantum thermodynamics of even earlier work by Landauer [29], which connects thermodynamics to classical computing.

The common denominator of thermodynamics and quantum information (and quan-

tum computation) is information. Whereas it is obvious that quantum information theory is concerned with studying how information can be encoded, manipulated, and communicated using quantum systems (see, e.g., [27, 30]), it may not be as clear immediately how crucial the notion of information is in thermodynamics. Historically, thermodynamics is concerned with physical systems that have many degrees of freedom, typically too many to keep track of all of them. Instead, one often has direct access only to a few macroscopically observable parameters, such as energy, temperature, or pressure. Classical thermodynamics provides a quantitative description of these parameters via the laws of thermodynamics. Meanwhile, statistical mechanics underpins this framework by providing microscopic explanations for the macroscopically observed phenomena. At the heart of these explanations lies the information that is available about a given system. In other words, the starting point of statistical mechanics is the assignment of probabilities to the microstates of a system via statistical ensembles, given information about the system's energy, composition, and interaction with its environment (e.g., whether the system is in thermal equilibrium with a heat bath or a non-equilibrium system), as discussed, for instance, in [31]. While information about a few macroscopic parameters of a quantum system may thus be available, this typically means that there also is an enormous lack of information about the specific microstate of the system.

In particular, one can ask which macrostate should be assigned to a (quantum) system in thermal equilibrium with respect to an assumed external heat bath at fixed temperature when all that is known is its (average) energy. By Jayne's maximum entropy principle [31, 32], the unique answer is: a thermal state[1], i.e., a mixed state (represented by a density operator) that maximizes the (von Neumann) entropy for fixed average energy. This captures the idea that, when in doubt, it is prudent to assume the minimal amount of information (hence the maximal amount of entropy) about the system. Conversely, we can identify the thermal state as the state with minimal (average) energy given fixed entropy. Information-theoretic considerations hence enter thermodynamics at a fundamental level. One aim of this habilitation thesis is to draw attention to the fact that the converse is also true. In other words, here we take the point of view that thermodynamic considerations also enter the foundations of quantum information theory, as we shall elaborate on shortly.

The premise of many quantum information processing tasks is the assumed ability to prepare pure quantum states, to perform unitary operations, and to carry out ideal projective measurements, at least to within any desired margin of error. And, indeed, in modern quantum optics and quantum technologies, we have unprecedented control over individual, or small groups of quantum systems. For instance, current state-of-the-art setups for quantum computing are able to control around 20-50 qubits using trapped ions [33–35], with similar qubit numbers achieved in superconducting processors by companies such as

[1] Also called a Gibbs state. When the system is assumed to be of fixed composition but may be in states of varying energy, this corresponds to the canonical ensemble, which we will consider here unless stated otherwise.

Google [36], IBM [37], and Rigetti Computing [38], and up to 10 qubits can currently be fully controlled in solid state memories [39]. At the same time, gate fidelities of well above 99% have been reported for single- and two-qubit gates for physical qubits in different platforms [40, 41], and even logical entangling gates have recently been realized [42]. These and many other impressive developments notwithstanding, it is clear from a fundamental thermodynamic perspective that pure states can only ever be prepared approximately.

More specifically, the third law of thermodynamics dictates that no (quantum) system can be cooled to the ground state (a *pure* state for non-degenerate Hamiltonians) with finite resources [43–48]. Here, resources include the time and energy invested in the preparation, but also the complexity (qualitatively, the Hilbert space dimension that is assumed to be fully and exactly controllable via the application of arbitrary unitary operations) of the involved quantum systems. By extension, the third law stipulates that no pure state can be prepared exactly, even if perfectly unitary transformations could be applied to any controlled system. However, even in the most well-isolated vacuum chambers, residual particles remain and may interact with any controlled system therein, leading to decoherence and noise. Meanwhile, the application of unitaries themselves often involves precise timing of interactions, which in turn relies on perfect clocks. The latter, of course also have intrinsical statistical inaccuracies (see, e.g., [49] for a discussion) in any finite-resource scenario. Finally, as we have shown in [50], this implies that no ideal projective measurement can be carried out with finite resources either, unless one initially already has complete knowledge of the respective quantum systems and may hence assume the availability of (zero-temperature) pure states a priori. Conversely, we thus see that carrying out state preparation, (quantum) computation, and measurements in a precise and controlled way requires the investment of time and energy to move systems out of inevitable thermal equilibrium that reflects one's incomplete information about the system in question. Thus, it becomes evident that thermodynamic considerations naturally enter the domain of quantum information theory — and vice versa [51].

Over the past decade, this synergy between thermodynamics and (quantum) information theory has shaped and propelled the field of quantum thermodynamics, which, broadly speaking, is concerned with thermodynamics in the quantum regime, and includes (but is not limited to) such topics as equilibration and thermalization of quantum systems, quantum heat engines, storage of work in 'quantum batteries', work extraction from and refrigeration of quantum systems, fluctuations and measurements of work, irreversibility in out-of-equilibrium processes, thermodynamics of strongly coupled systems, and information-theoretic approaches to thermodynamics. Overviews over these, and a variety of other aspects of quantum thermodynamics can be found in the respective chapters of [52].

Here, we discuss some exemplary situations in quantum thermodynamics (covered in detail in the selected publications [50, 53–58] that are presented in full in Part II), which highlight the interplay between thermodynamics and quantum information theory, and

their respective resources. We consider situations where (i) we have incomplete information about the quantum systems in question and we hence assume that these systems are initially at thermal equilibrium with respect to their environment, and where (ii) work is either converted into resources valuable from an information-theoretic point of view [50, 53, 54, 57] (i.e., various types of correlations), or where the extraction [55], storage [56], and estimation [58] of work is restricted by information-theoretic limitations (e.g., the complexity of the allowed operations).

This habilitation thesis is organized as follows. To provide some context for the publications presented later on, the remaining sections of Part I provide an overview over pertinent concepts and definitions in quantum thermodynamics, quantum optics, and quantum information. In particular, Sec. I.2 examines the problem of defining and estimating work and its fluctuations. In Sec. I.3 we then briefly review recent approaches for understanding fundamental and practical limitations on work extraction and storage in quantum optical systems. In Sec. I.4, we then turn to conversion of work into correlations. For this purpose, we discuss different types of correlations and how they can be established at the expense of work. Section I.5 the concludes Part I with a brief summary and outlook on open problems. In Part II, we then present the mentioned selected publications [50, 53–58].

I.2 Work and Work Estimation

I.2.A Heat and Work

A conceptually fundamental distinction that is made in (classical) thermodynamics is that between *heat* and *work*. The first law of thermodynamics stipulates that the change in internal energy can be assigned to changes in either one of these two contributions. Whereas heat corresponds to changes of energy on the microscopic level, work is associated to macroscopic changes in energy. In other words, heat is energy attributed to unordered thermal motion of the microscopic constituents of a system, whereas work performed by a system is energy that is transferred from one system to another via the exertion of macroscopic mechanical forces that are measurable and can in turn be stored for later use as potential energy, e.g., by lifting a weight against the gravitational pull. In this sense, work is 'ordered' energy.

Heat and work can in principle be converted into each other, but the laws of thermodynamics enshrine the observation that there is a preferred direction to this conversion, i.e., work can in principle be turned into heat at unit efficiency, but the conversion of heat into work is limited in its efficiency. As argued, e.g., in [2, pp. 117], the first law of thermodynamics immediately provides the bound that a heat engine which uses an input $\delta Q \geq 0$ of heat and attempts to turn it into work δW must not violate the relation $\delta W \leq \delta Q$. Meanwhile, a heat engine that harnesses the flow of heat (according to the second law of thermodynamics) from a hot bath at temperature T_{H} to a cold bath at temperature T_{C} in order

to perform work on a working substance can never achieve a conversion of heat into work at an efficiency beyond the Carnot efficiency $\eta_{\max} = 1 - T_{\mathrm{C}}/T_{\mathrm{H}}$. This implies a notion of irreversibility in the conversion of work to heat.

A different way of expressing this irreversibility is via the second law phrased as a statement about entropy S. For an idealized closed system that is initially at thermal equilibrium with its environment at temperature T and which may exchange no matter but only energy with the latter, infinitesimal changes $\mathrm{d}S$ of the entropy due to an irreversible process are bounded from below by the ratio of δQ, the infinitesimal increment[2] of heat transferred to the system, and the environment temperature T, that is $\mathrm{d}S \geq \delta Q/T$, with equality when the process is reversible. Since $\delta Q \geq 0$, an implication of the above is the observation that the entropy of any closed system may never decrease: $\mathrm{d}S \geq 0$. In this sense, irreversibility is tied to an increase in entropy (or, from an information-theoretic point of view: loss of information), which is in turn associated (albeit not identified) with the transfer of heat. Conversely, a process in a closed system for which $\mathrm{d}S = 0$ also satisfies $\delta Q = 0$ (for any finite environment temperature), and all changes in internal energy can be attributed to work in such a case. Heat and work can thus both be considered to be thermodynamical resources, but there is a clear hierarchy between them: work is strictly more useful than heat.

This leaves us with the question of identifying the contributions of heat and work in general irreversible processes. Now, in classical macroscopical examples such as a weight that has been lifted to a particular height and is being held in place there, it is reasonable to say that the work has been stored in the system in the form of potential energy and work can hence be attributed to the system. But of course the premise for such a statement is that the weight is at a well-defined height, and hence has a well-defined potential energy. In practice this is generally not the case: measurements of the height will vary ever so slightly in their values, leading to fluctuations in the assigned work value. Nevertheless, in a macroscopic setting one may typically safely assume that the fluctuations around the mean are small, much smaller than the attributed work, and can thus be disregarded for most practical purposes. However, there is no reason to expect that this is the case in the quantum domain, and even providing a definition for work is no longer straightforward, as we will discuss in the next section.

I.2.B Work in Quantum Thermodynamics

Along with ever-increasing control over quantum systems and unprecedented access to technologies at the nanoscale which have been brought on by the second quantum revolution [59] come questions regarding their fundamental limitations. Currently, quantum technologies are considered to be operating in what is often referred to as the noisy intermediate-

[2]Note that the difference in notation between infinitesimal increments $\mathrm{d}S$ of entropy and δQ of heat highlights that the entropy is a function of the system state only, whereas heat is not.

scale quantum (NISQ) regime, see, e.g. [60], a name one might consider a portent in the sense that itf implies the hope that further technological advances will lead to noise-robust large-scale quantum devices in the future. In this endeavour, residual heat and noise are ever-present adversaries, and moving systems away from thermal equilibrium with their surroundings requires sufficient control, as well as the investment of time and energy. Indeed, from a fundamental thermodynamic perspective, pure states can only be prepared approximately since Nernst's unattainability principle [61, 62] — the third law of thermodynamics— demands that infinite resources are required to cool any system to its ground state. To accurately assess the resources required for a specific task, it must therefore in principle be assumed that the respective system is initially in a thermal state and that work must be invested to change this.

But what is a suitable definition of work in the quantum regime? What can be considered a suitable representation of the 'weight' that is often so evocatively associated with work in classical thermodynamics. In the context of *quantum* thermodynamics, the notion of work is ultimately tied to the quantum systems that one assumes may store or provide energy, and to the control that one assumes to be able to enact over these systems and their interactions with each other and with the environment. Consequently, a number of different approaches to this problem have emerged, and we will here take a glance at a few of them.

One possibility, argued for by Reeb and Wolf [63], is to consider a system S in contact with a thermal bath B, initially in some joint state ρ_{SB}. The closed joint system is then assumed to be controlled sufficiently well so that it can be manipulated by a global unitary transformation U_{SB}. Then, for a fixed joint Hamiltonian H_{SB}, the work W necessary to move the system from its initial state $\rho_{\mathrm{S}} = \mathrm{Tr}_{\mathrm{B}}\big(\rho_{\mathrm{SB}}\big)$ to the final state $\tilde{\rho}_{\mathrm{S}} = \mathrm{Tr}_{\mathrm{B}}\big(U_{\mathrm{SB}}\,\rho_{\mathrm{SB}}\,U_{\mathrm{SB}}^{\dagger}\big)$ is defined via the average energy change of the joint system, i.e.,

$$W = \mathrm{Tr}\Big(H_{\mathrm{SB}}\big[\rho_{\mathrm{SB}} - U_{\mathrm{SB}}\,\rho_{\mathrm{SB}}\,U_{\mathrm{SB}}^{\dagger}\big]\Big). \tag{I.1}$$

An alternative to this viewpoint was discussed by Alicki and members of the Horodecki family in Ref. [64], where the control over the system of interest is phrased in terms of time-dependent coupling (with otherwise fixed interaction Hamiltonians) to reservoirs and an external driving resulting in an explicitly time-dependent system Hamiltonian $H_S(t)$. The contributions of heat and work to the total system energy change $dE(t)$, where $E(t) = \mathrm{Tr}\big(\rho_{\mathrm{S}}(t)\,H_S(t)\big)$, are then identified with the changes associated with the state and Hamiltonian, respectively. In particular, work is defined as

$$W(t) = \int_0^t dt'\,\mathrm{Tr}\Big(\rho_{\mathrm{S}}(t')\,\frac{dH_S(t')}{dt'}\Big). \tag{I.2}$$

Both of these approaches follow certain intuitions about the qualities of heat and work, but ultimately do not provide an operational interpretation for the specific work values. In particular, no explicit system for work storage — no 'weight' — features in these models.

In contrast, exactly such an explicit representation of a work-storage system has been considered by Åberg [65], as well as by Brunner, Linden, Popescu and Skrzypczyk [66]. There, work and its statistics can be defined directly via the Hamiltonian H_{W} along with the reduced states $\rho_{\mathrm{W}}^{\mathrm{initial}}$ and $\rho_{\mathrm{W}}^{\mathrm{final}}$, before and after an interaction with the system S, respectively, i.e.,

$$W = \mathrm{Tr}\Big(H_{\mathrm{W}}\big[\rho_{\mathrm{W}}^{\mathrm{final}} - \rho_{\mathrm{W}}^{\mathrm{initial}}\big]\Big). \tag{I.3}$$

A drawback of this approach identified by Gallego, Eisert and Wilming in Ref. [67] is that a work value is assigned to a change of average energy that could be brought about by what would (classically) be considered a source of heat, e.g., thermalization of the work-storage system. The authors of [67] further put forward an axiomatic approach to the definition of work in quantum thermodynamics. That is, work is abstractly defined (non-uniquely) as a function $W(\rho^{\mathrm{initial}} \to \rho^{\mathrm{final}})$, assigning a real work value to each state transformation from an initial state ρ^{initial} to a final state ρ^{final}, such that certain axioms are fulfilled. For instance, it is argued that the work value should be expressible as a difference of functions of the initial and final state, i.e., $W(\rho^{\mathrm{initial}} \to \rho^{\mathrm{final}}) = g(\rho^{\mathrm{final}}) - g(\rho^{\mathrm{initial}})$ for some suitable function g. One candidate for the function $g(\rho)$ is the free energy $F(\rho)$ with respect to a heat bath at temperature T, given by

$$F(\rho) = E(\rho) - T\,S(\rho), \tag{I.4}$$

where $E(\rho) = \mathrm{Tr}(H\rho)$ is the average energy of the system and $S(\rho) = -\mathrm{Tr}\big(\rho \ln \rho\big)$ is its von Neumann entropy. Such an axiomatic approach is thus useful to narrow down the list of potential candidates for work quantifiers but, unless a particular one of them is uniquely determined by the axioms, there is still room for ambiguity in the interpretation of the work values.

The free energy $F(\rho)$ indeed has appealing properties that one would desire for a work quantifier. In particular, for given system Hamiltonian H_S and fixed heat-bath temperature T, it is minimal for a thermal state of the same temperature. Such a thermal state τ_S is of the form

$$\tau_S(\beta) = \frac{e^{-\beta H_S}}{\mathcal{Z}} = \frac{\sum_n e^{-\beta E_n} \mid E_n \rangle\!\langle E_n \mid}{\sum_m e^{-\beta E_m}}, \tag{I.5}$$

where $\beta = 1/T$ is the inverse temperature[3] of the heat bath, the system Hamiltonian has the spectral decomposition $H_S = \sum_n E_n \mid E_n \rangle\!\langle E_n \mid$, and $\mathcal{Z} = \mathrm{Tr}\big(e^{-\beta H_S}\big)$ is the partition function. We can then perform a quick calculation to determine the free energy of such a thermal state as

$$\begin{aligned} F(\tau_S) &= \mathrm{Tr}\big(\tau_S H_S\big) + T\,\mathrm{Tr}\big(\tau_S \ln\big[\tfrac{e^{-\beta H_S}}{\mathcal{Z}}\big]\big) = \mathrm{Tr}\big(\tau_S H_S\big) + T\,\mathrm{Tr}\big(\tau_S\,[-\beta H_S - \ln(\mathcal{Z})]\big) \\ &= \mathrm{Tr}\big(\tau_S H_S\big) - \beta T\,\mathrm{Tr}\big(\tau_S H_S\big) - T\,\ln(\mathcal{Z})\,\mathrm{Tr}\big(\tau_S\big) = -T\,\ln(\mathcal{Z}). \end{aligned} \tag{I.6}$$

[3] We use units where $k_{\mathrm{B}} = \hbar = 1$ throughout.

In addition, the thermal state maximizes the entropy S at fixed average energy E, and simultaneously minimizes the average energy at fixed entropy. In the following, we will discuss the relationship between the free energy and work in more detail.

I.2.C Work Estimation

A different point of view to that described in the previous section arises in stochastic thermodynamics (see, e.g., the reviews [68–70]), where work (and heat) take the roles of stochastic random variables. As such, work performed on (or extracted from) a system in a given process Λ is characterized by a probability distribution, whose average provides an expected work value $\langle W \rangle$. Such a work distribution also allows one to study higher statistical moments: work fluctuations, which have been a significant focus in quantum statistical mechanics (see, e.g., [71–74] for a selection of the large body of available literature on this topic). To define this work distribution, we will consider one of the most prominent approaches for estimating work in an out-of-equilibrium process, the so-called *two-point measurement* (TPM) scenario [75], which we shall describe now following the presentation in Ref. [58].

I.2.C.1 The Two-Point Measurement Scheme

The situation considered in the TPM scenario is that of a quantum system with initial Hamiltonian $H_S^{(0)} = \sum_n E_n^{(0)} \mid E_n^{(0)} \rangle\langle E_n^{(0)} \mid$ that is initially at thermal equilibrium with respect to its environment at ambient temperature T. In the beginning, the system is therefore described by a thermal state $\tau_S^{(0)} = \exp(-\beta H_S^{(0)})/\mathcal{Z}^{(0)}$ with partition function $\mathcal{Z}^{(0)} = \mathrm{Tr}\big(\exp(-\beta H_S^{(0)})\big)$. At time $t = t_0$, the system is driven out of equilibrium by a process Λ resulting in a time-dependent system Hamiltonian $H_S(\lambda_t)$, where λ_t is an externally controlled parameter such that $H_S(\lambda_{t_0}) = H_S^{(0)}$. At time $t = t_\mathrm{f}$, the process results in a final Hamiltonian

$$H_S(\lambda_{t_\mathrm{f}}) = H_S^{(\mathrm{f})} = \sum_n E_n^{(\mathrm{f})} \mid E_n^{(\mathrm{f})} \rangle\langle E_n^{(\mathrm{f})} \mid , \tag{I.7}$$

and it is assumed that $\lambda_t = \lambda_{t_\mathrm{f}}$ for all $t \geq t_\mathrm{f}$, i.e., the process concludes at $t = t_\mathrm{f}$. The time evolution of the system can be described by a unitary $U_\Lambda = \mathcal{T}_+ \exp\left(-i \int_{t_0}^{t_\mathrm{f}} H_S(\lambda_t)\right) dt$, where $\mathcal{T}_+$ denotes time-ordering, which leaves the system in the final state

$$\rho_S^{(\mathrm{f})} = U_\Lambda \tau_S^{(0)} U_\Lambda^\dagger . \tag{I.8}$$

Within this setting, the goal is then to estimate the work (distribution) of this out-of-equilibrium process. To this end, two projective measurements with respect to the eigenbases $\{\mid E_n^{(0)} \rangle\}_n$ and $\{\mid E_m^{(\mathrm{f})} \rangle\}_m$ of the initial and final system Hamiltonian, respectively, are carried out, one directly before and one directly after the dynamics represented by U_Λ [69, 75], as illustrated in Fig. I.1.

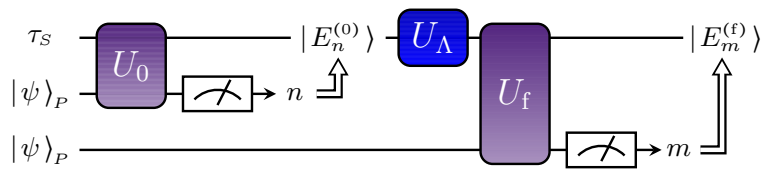

Figure I.1: Work estimation in the TPM scheme. To estimate the work done on or extracted from a system during a process Λ represented by a unitary U_Λ, two measurements are carried out before and after the process occurs, respectively. The outcomes of these two ideal measurements, labelled "n" and "m", allow one to conclude that the system is left in the states $|E_n^{(0)}\rangle$ and $|E_m^{(\mathrm{f})}\rangle$, respectively. Both of these ideal measurements can be modelled as a unitary interaction of the system of interest with (two copies of) a pointer system P initially prepared in a pure state $|\psi\rangle_P$. As discussed in Ref. [58], assuming that the pointers are initially not in pure states leads to *non-ideal* measurements, where the post-measurement system states are no longer pure, resulting in corrections to the estimated work.

The TPM scheme comprises two projective measurements with respect to the eigenbases of the Hamiltonians $H_S^{(0)}$ and $H_S^{(\mathrm{f})}$ before and after the process Λ, respectively. Here, a crucial assumption — which we will return to later — is that the measurements that are performed are ideal [50], in particular, that the post-measurement states of the system are pure states. In any given run of such an estimation procedure, two outcomes labelled by "n" and "m" are obtained in these measurements, and one thus concludes that, directly after each measurement, the system is left in the eigenstates $|E_n^{(0)}\rangle$ and $|E_m^{(\mathrm{f})}\rangle$, respectively. The probability $p_n^{(0)}$ for obtaining the first outcome n only depends on the initial state, $p_n^{(0)} = \langle E_n^{(0)}|\tau_S^{(0)}|E_n^{(0)}\rangle = \exp(-\beta E_n^{(0)})/\mathcal{Z}^{(0)}$, while the probability for a transition between the (pure) energy eigenstates during the process is $p_{n\to m} = |\langle E_m^{(\mathrm{f})}|U_\Lambda|E_n^{(0)}\rangle|^2$. The joint probability for obtaining the outcomes n and m in the first and second measurement, respectively, is thus given by

$$p(n,m) = p_n^{(0)}\, p_{n\to m} = \frac{\exp(-\beta E_n^{(0)})}{\mathcal{Z}^{(0)}}\, |\langle E_m^{(\mathrm{f})}|U_\Lambda|E_n^{(0)}\rangle|^2. \tag{I.9}$$

To each such transition, one may assign an unambiguous work value $W_{n\to m} = E_m^{(\mathrm{f})} - E_n^{(0)}$, and we can then define the work distribution $P(W)$ via

$$P(W) = \sum_{m,n} p(n,m)\, \delta\left(W_{n\to m} - W\right). \tag{I.10}$$

The average work performed during the protocol can then be obtained by integration, that is,

$$\langle W\rangle = \int P(W)\, W\, dW = \sum_{m,n} p(n,m)\left(E_m^{(\mathrm{f})} - E_n^{(0)}\right). \tag{I.11}$$

One may easily check that the average work defined in this way matches the average energy change of the system, i.e.,

$$\langle W\rangle = \mathrm{Tr}(H_S^{(\mathrm{f})}\rho_S^{(\mathrm{f})}) - \mathrm{Tr}(H_S^{(0)}\tau_S^{(0)}) = \Delta E. \tag{I.12}$$

In this way, an estimate of $\langle W \rangle$ can be obtained by performing pairs of ideal measurements and collecting the corresponding outcome statistics. However, the latter also allow one to evaluate higher statistical moments of the work distribution. For instance, one may consider the mean squared deviation of the work values from the average and define the quantity

$$(\Delta W)^2 = \int P(W)\,(W - \langle W \rangle)^2\, dW = \sum_{m,n} p(n,m)\big(W_{n\to m} - \langle W \rangle\big)^2, \tag{I.13}$$

which, following the terminology used in Ref. [56], we refer to as *the work fluctuations* for simplicity. However, we note that work fluctuations are often associated with a different quantity in quantum statistical mechanics, in particular, in the context of the celebrated fluctuation relations of Jarzynski [76] and Crooks [77], see, e.g., [69, 78] for an overview.

I.2.C.2 The Jarzynski Relation

Let us now briefly focus on the Jarzynski fluctuation relation, which follows as a corollary from the more general theorem by Crooks. For Jarzynski's relation, the quantity of interest is the average of the *exponentiated* work value. That is, it can be shown in the context of the TPM scheme that there is an *exact* relation between the average of $\exp(-\beta W)$ and the exponentiated equilibrium free-energy difference $\exp(-\beta\Delta\tilde{F})$, i.e.,

$$\langle e^{-\beta W} \rangle = e^{-\beta\Delta\tilde{F}}. \tag{I.14}$$

Here, $\Delta\tilde{F}$ is given by

$$\Delta\tilde{F} := F(\tau_S^{(\mathrm{f})}) - F(\tau_S^{(0)}) = T\,\ln\Big(\frac{\mathcal{Z}^{(0)}}{\mathcal{Z}^{(\mathrm{f})}}\Big), \tag{I.15}$$

where $\tau_S^{(\mathrm{f})} = \exp(-\beta H_S^{(\mathrm{f})})/\mathcal{Z}^{(\mathrm{f})}$ is the thermal state of the system with respect to the final Hamiltonian $H_S^{(\mathrm{f})}$, with the partition function $\mathcal{Z}^{(\mathrm{f})} = \mathrm{Tr}\big[\exp(-\beta H_S^{(\mathrm{f})})\big]$. What is particularly noteworthy in the relation in Eq. (I.14) is the fact that, while the left-hand side represents (a statistical average derived from work values in) an *out-of-equilibrium* process, the right-hand side is determined solely by the *equilibrium* quantity $\Delta\tilde{F}$, and we have used the tilde here to signify the discrepancy with respect to the non-equilibrium free-energy difference $\Delta F = F(\rho_S^{(\mathrm{f})}) - F(\tau_S^{(0)})$. In particular, we note that, since the system dynamics we consider here is unitary, such that $S(\rho_S^{(\mathrm{f})}) = S(\tau_S^{(0)})$, and since $\langle W \rangle = \Delta E$ from Eq. (I.12), we also have $\langle W \rangle = \Delta F$, whereas a straightforward application of Jensen's inequality $\langle e^X \rangle \geq e^{\langle X \rangle}$ for the convex function e^X leads to the second-law-like inequality

$$\langle W \rangle \geq \Delta\tilde{F}. \tag{I.16}$$

The notion of work as a random variable that is to be estimated thus brings about some interesting observations and generally follows the idea that '*work is not an observable*', as

argued by Talkner, Lutz, and Hänggi in Ref. [75]. In particular, the problem of work estimation leads to a number of fundamental questions regarding the specifics of the estimation process and the nature of the measurements necessary to do so (see, e.g., [71–74]), as well as regarding the control over the system, e.g., via feedback, and its role in fluctuation relations [79]. In addition, the possibilities for more general approaches than the TPM scheme have been investigated, e.g., by Perarnau-Llobet et al. in Ref. [80] and by Lostaglio in Ref. [81]. For details on many of these aspects, we refer the interested reader to a series of book chapters [82–88] on this subject.

An aspect of this estimation problem of particular interest to this thesis is the ideal character of the measurements in question. As we explain in detail in Sec. II.6, where Ref. [50] is presented, ideal measurements require pure input states for the pointer systems to which the measured system is coupled. However, preparing such pure states is only possible if infinite time, infinite energy, or infinitely complex interactions are available, a point that is often attributed to Nernst's unattainability principle [61, 62], but whose precise formulation — in particular, with regards to the notion of complexity — is still a subject of ongoing debate [89]. In the presence of pointer states at inevitable nonzero temperatures, the resulting measurements are non-ideal, which leads to mixed conditional post-measurement states for the system. The effects of such non-ideal measurements on the estimation of work, and on the formulation (as well as, on the ability to satisfy) the relations by Jarzynski and Crooks, is discussed in detail in another publication [58], that we present in full in Sec. II.7.

Finally, let us briefly draw some more attention to Ref. [90], where the point is made that, although work may not be representable as an observable on the system S itself, it is indeed possible to define work via an *external* observable. An astute observation made by Beyer, Luoma, and Strunz in Ref. [90] in this context is that the estimation of work in the TPM scheme requires knowledge of the system Hamiltonian, before and after the interaction, whereas in a classical setting, work may be estimated via an externally applied force. The authors of [90] therefore propose a scheme based on a collision model by which work is defined as an external observable also in the quantum regime. While this approach does not require knowledge of the particular system Hamiltonian, it makes very specific assumptions about the ability to control the external systems with which the driven system interacts, in particular, assuming the ability to precisely prepare an ensemble of pure states and to measure these with respect to carefully chosen measurement bases (depending on the particular ensemble state). A particular insight of Ref. [58] that we wish to emphasize in this context is that the precision of the work estimate ultimately depends on the amount of resources and control one is able, or willing, to invest into the estimation procedure. We thus come to the conclusion that every definition of work in the quantum regime comes with advantages and drawbacks, and is tailored to a set of specific assumptions about the control one is able to enact over the system of study and its interactions with the environment. Moreover, it appears that definitions of work must be chosen in accordance with the

particular task that one has in mind, e.g., for a heat engine [91]. This now leads us from general considerations of heat and work to particular tasks in quantum thermodynamics, specifically, to the extraction and storage of work, and the role that the control over the respective quantum systems plays in these procedures.

I.3 Extraction and Storage of Work using Quantum Systems

I.3.A Passivity and Ergotropy

Although different definitions of work are available in general, in particular when it comes to driving a system out of thermal equilibrium, the situation appears to be more clear-cut for the problem of work extraction. There, the main questions is: *How much work can be extracted from a given quantum system?* Here, it is understood that by 'quantum system' we mean a particular quantum state ρ in a fixed Hilbert space and a corresponding fixed Hamiltonian H. In order to make the question more precise, however, we need to specify the particular quantifier for work that we consider, which entails a restriction to certain processes by which this work is to be extracted, as we shall discuss.

If we wish to talk about extractable work as a property of the system specified by ρ and H, then our quantifier for extractable work should not depend on the specifics of any external devices that may be used to extract work. Suitable candidates for such quantifiers would hence be, for instance, the achievable differences in average energy, ΔE, or free energy ΔF. However, we also have to restrict the transformations that we consider for such a task.

First, it is clear that we have to demand that the work-extraction process modifies the system state, i.e., ρ is mapped to another state $\tilde{\rho}$, but returns the system Hamiltonian to its original form H when the process is complete. Otherwise, we could (more or less) arbitrarily change the average energy of the system simply by changing the energy gaps of the Hamiltonian. Second, it is reasonable to restrict to operations that can be represented as unitary operations of the system's density matrix only. If that was not the case, we could naively consider, e.g., a global unitary transformation on the system and some apparatus not initially correlated to the system. By maximizing the achievable work extraction from the system over all choices of apparatuses, we would obtain an expression for the extractable work that depends only on the system itself, as desired. At the same time, however, this expression would trivially be just the average energy of the system, since we could always consider the apparatus to be the same type of quantum system (same dimension and Hamiltonian) but initially in the corresponding ground state. A simple swap of the two systems would then lower the system average and free energies by $\Delta E = E(\rho)$ and $\Delta F = F(\rho)$, respectively, even while conserving the overall energy. While mathematically sound, such a definition of extractable work based on global (even energy-conserving) operations would hence effectively lead to equating work and heat.

The paradigmatic scenario [92] for quantifying the work extractable from a quantum system instead considers the case where the transformations that extract work from the system are realized by cyclic Hamiltonian dynamics. That is, the system's time evolution is governed by a time-dependent Hamiltonian $H(t)$ with cycles of duration τ, at the end of which the system is returned to its initial Hamiltonian, $H(n\tau) = H(0)$ for any $n \in \mathbb{N}$. Up to perturbations due to the interactions with the environment, this results in (local) unitary dynamics,

$$\rho \longmapsto \tilde{\rho} = U\rho U^\dagger, \qquad \text{and} \qquad H \longmapsto H, \tag{I.17}$$

described by unitary transformations U with $U^\dagger U = UU^\dagger = \mathbb{1}$ on the system. The unitary orbits of the input states thus determine the fundamental limits of operation of cyclic machines. Unitary work extraction has therefore been a focus of attention in quantum thermodynamics, see, e.g., [93–98]. In this scenario, we have $\Delta E = \Delta F$, and we can hence associate the extractable work with either quantity, and define the *ergotropy*

$$\mathcal{E}(\rho, H) := \max_U \operatorname{Tr}\big(H[\rho - U\rho U^\dagger]\big), \tag{I.18}$$

i.e., the maximal energy difference achievable within the unitary orbit of ρ. From this definition of extractable work, it follows that work extraction is only possible for states whose average energy can be lowered by unitary transformation, whereas all states for which this is not possible are called *passive*. The latter can be characterized in a rather simple way: all passive states ρ_{pass} are diagonal in the eigenbasis $\{|\, n \,\rangle\}$ of the Hamiltonian H, with $H\,|\, n \,\rangle = E_n\,|\, n \,\rangle$ (for simplicity we assume a discrete spectrum here), and the magnitudes of their diagonal entries are decreasing (not necessarily strictly) with increasing energy. In other words, any passive state ρ_{pass} can be written as

$$\rho_{\text{pass}} = \sum_n p_n \,|\, n \,\rangle\!\langle\, n \,|\,, \tag{I.19}$$

with $0 \leq p_n \leq 1$ and $\sum_n p_n = 1$, as usually, but additionally satisfying the constraints

$$p_n \leq p_m \qquad \text{when} \qquad E_n \geq E_m. \tag{I.20}$$

With this result at hand, the ergotropy can easily be rewritten as

$$\mathcal{E}(\rho, H) := \max_U \operatorname{Tr}\big(H[\rho - U\rho U^\dagger]\big) = \operatorname{Tr}\big(H[\rho - \rho_{\text{pass}}]\big), \tag{I.21}$$

where ρ_{pass} is the unique (up to unitaries in degenerate subspaces of H) passive state reachable from ρ.

The elegance and simplicity of this approach to work extraction notwithstanding, one may be led to ask how realistically achievable this figure of merit is in the light of imperfect control over complex quantum systems with (possibly infinitely) many dimensions. For instance, one may wonder how easily reachable the passive state is in practice for continuous-variable systems where full information about the initial state may not be available, or, conversely, which states can be reached by practically implementable transformations. As a

first step towards understanding such restrictions arising from limited control, Ref. [55], which is presented in detail in Sec. II.3, introduced the notion of *Gaussian passivity*, i.e., the class of continuous-variable states whose average energy cannot be reduced by Gaussian unitary transformations, which are typically considered as being practically easily implementable, and provided a full classification of these states via their first and second statistical moments.

As we shall see next, work extraction is not the only thermodynamically relevant task where such considerations regarding the practically realizable level of control over quantum systems come into play, and we thus turn to the problem of storing work in quantum systems.

I.3.B Quantum-Optical Systems as Quantum Batteries

As we have discussed, quantum thermodynamics has established a variety of different scenarios for the quantification of work and for modelling the associated state transformations. In this thesis, we have previously concluded that quantifiers for work must take into account the context of the task for which this work is to be used, and the task that we want to focus on now is that of supplying work to a particular quantum system. Within the broad spectrum of available scenarios for delivering such a work input, two distinct paradigms can be identified as the conceptual polar opposites (see, e.g., Refs. [46, 47, 89]): work can be supplied to a target system

(i) via a heat flow generated by a temperature gradient between two thermal baths, or

(ii) via a direct supply from a coherent work source.

Scenario (i) can be understood as the operation of a heat engine [99–104], where the heat flow supplies work incoherently to a working substance and the dynamics are globally energy-conserving. On the one hand, this paradigm is appealing from a thermodynamic point of view, since the system is overall closed and external control can be minimal in the sense that an external agent operating the machine is only needed to switch on (and off) interactions between the target and the heat baths. On the other hand, only a restricted class of state transformations is achievable within this paradigm [89] and practical laboratory situations in which quantum technologies are employed are not typically operated using heat engines.

An all-encompassing understanding of possible state transformations and their resource costs must therefore include coherent work sources as in (ii). Although the specific realizations of these work sources are often not included explicitly in modelling state transformations, doing exactly this will ultimately be necessary to truly obtain fine-grained descriptions that will lead to a better understanding of quantum systems beyond thermal equilibrium. Such descriptions can be envisioned to provide insights, e.g., regarding the effects of finite-time transformations, finite-size reservoirs, and fluctuations of relevant quantifiers.

A starting point for such a more general approach lies in modelling the work sources — commonly dubbed *quantum batteries* [94, 105] — on their own, i.e., independently of the systems that they eventually supply work to. In other words, quantum batteries are considered as quantum systems in which work can be temporarily deposited and from which it can subsequently be extracted. This approach has recently received a lot of attention, with main foci on the charging speed or power [106, 107], including different models for interaction between batteries and charging systems [108–115], the stability of the charged battery [116, 117], charging assisted by strong interactions and thermalization [118], and methods for describing fluctuations of the stored work [119–123].

Here, we focus on the approach of Ref. [56], which is presented in detail in Sec. II.4. There, battery charging is realized via cyclic Hamiltonian processes, mirroring the approach to work extraction discussed in Sec. I.3.A. In this case, the system Hamiltonian returns to its original form at the end of each cycle and the battery-system state, initially assumed to be thermal $\tau(\beta)$ as we have reasoned above, can be modelled to lie within the unitary orbit of the initial state [94]. The work that is transferred to the system in this way is thus just the difference in average energy, such that the transformation that is considered is

$$\tau \longmapsto \rho = U\tau(\beta)\,U^\dagger \qquad \text{with} \qquad \langle W \rangle = \Delta E = \mathrm{Tr}\big(H[U\tau(\beta)\,U^\dagger - \tau]\big). \tag{I.22}$$

This has the advantage that it allows us to consider the charging process independently of the specifics of other potentially involved auxiliary systems (e.g., charger systems as in [109, 112], or external classical power sources). We can thus focus on the properties of the charging process and of the charged battery, and study fundamental bounds on the chosen figures of merit. Here, we centre our attention on two particular quantities: the *charging precision*, quantified by the variance of the final battery charge

$$V(\rho) = \mathrm{Tr}\big[H^2\,\rho\big] - \big(\mathrm{Tr}\big[H\,\rho\big]\big)^2, \tag{I.23}$$

and the *work fluctuations* arising during the charging process

$$(\Delta W)^2 = \sum_{m,n} p_{m\to n}\big(E_n - E_m - \Delta E\big)^2, \tag{I.24}$$

where $H\,|\,n\,\rangle = E_n\,|\,n\,\rangle$, $p_{m\to n} = p_m\,|\,\langle\,n\,|\,U\,|\,m\,\rangle\,|^2$ is probability for a transition from energy level m to n, and $p_m = |\,m\,\rangle\,\tau\,|\,m\,\rangle$ is the probability to find the initial state in the energy eigenstate $|\,m\,\rangle$. While the charging precision captures a property of the final state, and the work stored in it, the fluctuations capture a property of the process that leads to this final state, and although it would of course be desirable to minimize both $V(\rho)$ and $(\Delta W)^2$ for any given work input ΔE, this is not possible in general [56] for arbitrary (in particular, thermal) initial states. Moreover, the optimal protocols for minimizing either one of the two quantities can generally be complicated, in particular for continuous-variable systems. However, as shown in Ref. [56], already simple Gaussian battery-charging protocols can provide sub-linear scaling of both the variance and work fluctuations with the input energy ΔE, even if neither can come close to the respective optimal (non-Gaussian) protocols.

Techniques from Gaussian quantum optics thus provide a useful toolbox, both from a theoretical and from a practical point of view, for quantum thermodynamical tasks, including work extraction and storage, showcasing the role of incomplete control (represented by restrictions to the available or realizable operations) and incomplete information (represented by thermal initial states). Moreover, as we will see next, thermodynamic considerations not only influence the extraction and storage of the thermodynamic resource work, but also have an impact on its conversion into information-theoretic resources: correlations.

I.4 Conversion between Work and Correlations

I.4.A Entanglement and Work

The interplay between thermodynamics and quantum information is enriched by another facet when considering quantum systems that consist of two or more subsystems. In such a setting, a control restriction typically considered in the context of quantum information, in particular, in entanglement theory (see, e.g., the reviews [124–129]), is that of *local operations*. That is, it is generally assumed that joint (global) operations on multiple subsystems are technically more challenging to execute or outright practically impossible to perform if the subsystems are sufficiently distant from each other. In quantum information this elevates the genuine quantum correlations called *entanglement* to the level of a resource, since entanglement cannot be created via local operations assisted by classical communication (LOCC) [130]. Meanwhile, the entanglement of pure states $|\,\psi\,\rangle_{\mathrm{AB}}$ can be quantified via the entropy of the subsystems, i.e., in terms of the *entropy of entanglement* given by

$$S_{\mathrm{E}}(|\,\psi\,\rangle_{\mathrm{AB}}) = S(\rho_{\mathrm{A}}) = S(\rho_{\mathrm{B}}), \tag{I.25}$$

where $\rho_{\mathrm{A/B}} = \mathrm{Tr}_{\mathrm{B/A}}(|\,\psi\,\rangle\langle\,\psi\,|_{\mathrm{AB}})$ are the reduced states of subsystems A/B and $S(\rho) = -\mathrm{Tr}\big(\rho \ln \rho\big)$ is the von Neumann entropy. Thermodynamically, this implies that any work invested in globally preparing an entangled pure state manifests locally as heat, meaning that work cannot be extracted locally from such an entangled state. For instance, consider the two-mode squeezed state

$$|\,\psi_{\mathrm{TMS}}\,\rangle = U_{\mathrm{TMS}}(r)\,|\,0\,\rangle = \frac{1}{\cosh(r)} \sum_{n=0}^{\infty} \tanh^n(r)\,|\,n\,\rangle_{\mathrm{A}}\,|\,n\,\rangle_{\mathrm{B}}\,. \tag{I.26}$$

of two harmonic oscillators labelled A and B with annihilation and creation operators a and $a^\dagger$, and b and $b^\dagger$, respectively. The state is obtained by applying the unitary $U_{\mathrm{TMS}}(r) = e^{r(a^\dagger b^\dagger - ab)}$ to the vacuum state $|\,0\,\rangle$, see, e.g., [131, Sec. 3]. The reduced states of both modes are thermal states with respect to the Hamiltonians $H_{\mathrm{A}} = \hbar\,\omega_{\mathrm{A}}\,a^\dagger a$ and $H_{\mathrm{B}} = \hbar\,\omega_{\mathrm{B}}\,b^\dagger b$, respectively, and their inverse temperatures are given by $\beta_{\mathrm{A/B}} = -\frac{1}{\hbar\,\omega_{\mathrm{A/B}}} \ln\big(\tanh^2(r)\big)$. Consequently, their temperatures $T_{\mathrm{A/B}}$, and thus the entropy of the subsystems and the entanglement of $|\,\psi_{\mathrm{TMS}}\,\rangle$ are monotonically increasing functions of the squeezing parameter $|r|$.

At the same time, also the average energy $\langle H_{\mathrm{A}} + H_{\mathrm{B}} \rangle$ is unitarily increased. Consequently, the work input required for applying $U_{\mathrm{TMS}}(r)$ can here be directly related to the creation of entanglement between the subsystems.

In this example, we can thus make two observations, first, the presence of correlations in this (globally pure) system implies that work can be extracted globally, but since the marginals are thermal, no work is extractable locally. This observation has been made on a much broader basis in Ref. [95], where it was concluded that, indeed, all correlations in locally thermal systems imply extractable work. Second, we note that the converse is also true, all correlations that are to be established in initially locally thermal systems are associated to a work cost. In this sense, the resources of quantum thermodynamics, i.e., work, and of quantum information, i.e., correlations and entanglement, can be converted into each other. However, the specifics of this conversion depend on the type of employed protocol and the initial temperatures of the subsystems. In particular, when attempting to create correlations entirely via cyclic Hamiltonian dynamics, i.e., within the unitary orbit of the initial state, potentially entangling unitaries have associated temperature thresholds above which no entanglement can be created [132]. For example, for the Gaussian transformation $U_{\mathrm{TMS}}(r)$ above to create entanglement when applied to two modes of the same frequency $\omega_{\mathrm{A}} = \omega_{\mathrm{B}} = \omega$ via a cyclic Hamiltonian process, the initial temperature of the modes may not exceed twice the value of the temperature defined by $\beta = -\frac{1}{\hbar\omega} \ln\big(\tanh^2(r)\big)$ [133]. In contrast, there exist non-Gaussian unitary transformations which may create entanglement at any initial temperature [53].

However, the creation of correlations between two initially thermal subsystems need not be limited to cyclic Hamiltonian (unitary) dynamics. For instance, in scenarios where the initial temperature exceeds the threshold temperature for unitary entanglement creation, an option is to invest work into first lowering the system temperature, before creating correlations unitarily. The naturally arising question in this context is then to identify the optimal conversion of work into entanglement, as quantified by a suitable operationally well-defined entanglement measure such as the entanglement of formation [134, 135], which can be defined via a convex-roof construction, i.e.,

$$E_{\mathrm{oF}}(\rho_{\mathrm{AB}}) := \inf_{\mathcal{D}(\rho_{\mathrm{AB}})} \sum_i p_i S_{\mathrm{E}}(|\,\psi_i\,\rangle_{\mathrm{AB}}), \tag{I.27}$$

where the infimum is defined over all pure-state decompositions, that is, $\mathcal{D}(\rho)$ is the set of all sets $\{(p_i, |\,\psi_i\,\rangle_{\mathrm{AB}})\}_i$ for which $\rho_{\mathrm{AB}} = \sum_i p_i \,|\,\psi_i\,\rangle\langle\,\psi_i\,|_{\mathrm{AB}}$, such that $\sum_i p_i = 1$ and $0 \leq p_i \leq 1$. The problem of optimizing $E_{\mathrm{oF}}(\rho_{\mathrm{AB}})$ for given work input was considered in Ref. [53], which we will discuss in more detail in Sec. II.1. However, even for locally thermal bipartite systems, answering this question has so far been limited to pairs of subsystems with identical Hamiltonians that consist of qubits [132], fermionic modes [53], or bosonic modes when restricting to Gaussian operations [53]. The bottleneck for answering this question more broadly lies in the computational intractability of calculating the relevant entanglement measures (here, the entanglement of formation) in general situations.

I.4.B Work Cost of Correlations

Nevertheless, it is possible to provide optimal protocols for converting work into correlations as quantified by the *mutual information*, which is given by

$$\mathcal{I}(\rho_{\mathrm{AB}}) = S(\rho_{\mathrm{A}}) + S(\rho_{\mathrm{B}}) - S(\rho_{\mathrm{AB}}). \tag{I.28}$$

For pure states, the mutual information trivially reduces to the entropy of entanglement above, but for mixed states a nonzero value of $\mathcal{I}(\rho_{\mathrm{AB}})$ may be due to classical or quantum correlations. For the mutual information, the optimal protocols for converting work to correlations when starting from two locally thermal identical subsystems at inverse temperature β were derived in [53] and consist of two steps: In the first step, the temperature of both subsystems is symmetrically lowered such that the subsystems both are left in thermal states at the same lower temperature, i.e., the first step maps the initial state $\tau_{\mathrm{A}}(\beta) \otimes \tau_{\mathrm{B}}(\beta)$ to a state $\tau_{\mathrm{A}}(\beta_{\mathrm{I}}) \otimes \tau_{\mathrm{B}}(\beta_{\mathrm{I}})$ with $\beta_{\mathrm{I}} \geq \beta$. The associated work cost is bounded from below by the non-equilibrium free-energy difference ΔF. In the second step, it is assumed that the subsystems are unitarily (i.e., via cyclic Hamiltonian dynamics) correlated such that both subsystems stay locally thermal but end up at a higher local temperature compared to the final state of the refrigeration step. In other words, the state $\tau_{\mathrm{A}}(\beta_{\mathrm{I}}) \otimes \tau_{\mathrm{B}}(\beta_{\mathrm{I}})$ is mapped to $\tilde{\rho}_{\mathrm{AB}} = U\, \tau_{\mathrm{A}}(\beta_{\mathrm{I}}) \otimes \tau_{\mathrm{B}}(\beta_{\mathrm{I}})\, U^{\dagger}$ such that $\mathrm{Tr}_{\mathrm{B/A}}(\tilde{\rho}_{\mathrm{AB}}) = \tau_{\mathrm{A/B}}(\beta')$ for $\beta' \leq \beta_{\mathrm{I}}$. For a given overall work input W, the correlations established in this way are bounded by

$$\mathcal{I}(\rho_{\mathrm{AB}}) \leq \beta\, W. \tag{I.29}$$

Whether or not this bound can be attained, i.e., if one can satisfy $\mathcal{I}(\rho_{\mathrm{AB}}) = \beta\, W$, then hinges on several details. First, equality can only be achieved if the cooling cost is indeed given exactly by the free-energy difference, which in turn depends on the particular cooling paradigm, see, e.g., [46, 47]. Second, there is a dependence on the work input itself, i.e., in the regime where the work input is small enough so that the optimal refrigeration step reaches a nonzero temperature, the protocol may operate in the linear regime where $\mathcal{I}(\rho_{\mathrm{AB}}) = \beta\, W$, but for larger W, the cooling step is naturally restricted by reaching $T = 0$, meaning an overall sublinear conversion of work into correlations.

Third, the second step of the optimal protocols operate based on the assumption of the existence of so-called *symmetrically thermalizing unitaries* (STUs) [57, 136], i.e., unitary transformations U that map the input pair of thermal states $\tau_{\mathrm{A}}(\beta_{\mathrm{I}}) \otimes \tau_{\mathrm{B}}(\beta_{\mathrm{I}})$ to a final state $\tilde{\rho}_{\mathrm{AB}}$ such that the marginals are both thermal at the same higher temperature, $\mathrm{Tr}_{\mathrm{B/A}}(\tilde{\rho}_{\mathrm{AB}}) = \tau_{\mathrm{A/B}}(\beta')$ for all β_{I} and all $\beta' \leq \beta_{\mathrm{I}}$, and for all Hamiltonians $H_{\mathrm{A}} = H_{\mathrm{B}}$. This question is discussed in more detail in Ref. [57] which we present in Sec. II.5. In addition to the previously known existence results for STUs for Hamiltonians with equally spaced energy levels [132, 137], Ref. [57] provided a number of proof techniques based on majorisation theory that provided existence proofs for STUs in local dimensions $d = 3$ and $d = 4$, but for higher local dimensions the question remains unsolved to date.

When the Hamiltonians of the two subsystems are not identical, STUs do not exist in general, but in that case STUs are also generally not the solutions to the problem of optimizing the mutual information at fixed energy input [57]. Besides this local but asymmetric situation, one may also wonder how interacting Hamiltonians influence the work cost of creating correlations. We have investigated this problem in Ref. [54], which is presented in full in Sec. II.2. As we show there, the presence of an interaction term in the Hamiltonian, and the resulting correlations (and even entanglement) in the initial thermal states can lead to a reduced work cost of newly created mutual information $\Delta\mathcal{I}$ in the sense that $\Delta\mathcal{I} > \beta W$.

I.5 Preliminary Conclusion and Outlook

As we have seen, ideas from thermodynamics and information theory can be fruitfully combined with techniques from quantum theory, especially quantum information and quantum optics, to gain a better understanding of the fundamental limits of achievable transformations of quantum systems in the face of limited information and restricted control. In particular, we have taken a closer look at the main resources of quantum thermodynamics and quantum information, work and correlations, respectively, and we have discussed the premise for meaningful definitions of work, its estimation, extraction, storage, and conversion to correlations. In all of these tasks, an initial lack of complete information about the systems under inspection and the apparatuses to which they are coupled leads to initial thermality. The latter leads to a loss in quality or performance in terms of the respective figures of merit for each task. But at the same time, this opens up an interesting field of study for determining optimal strategies for each task. The selection of works [50, 53–58], only briefly discussed so far but included in full in Part II, represent some of our contributions to this field, but research in this direction is far from concluded.

Indeed, many open questions remain in the overlap of quantum thermodynamics and quantum information. Even when limiting to the range of questions discussed here, one is met with a variety of further open problems, including (but not limited to) optimal battery-charging protocols for precision and fluctuations for systems (in particular, finite-dimensional) with arbitrary Hamiltonians, potential trade-offs between precision, fluctuations, and power in battery charging, as well as the influence of control restrictions (e.g., in terms of local versus global operations) on the latter. As far as non-ideal measurements (e.g., for work estimation) are concerned, much is yet to be learned about the influence on practical protocols such as the stabilization of open-system batteries (cf. [117]) or the specifics of realizing unbiased measurements in macroscopic detectors. As far as the question of the optimal conversion of work to correlations is concerned, an outstanding issue is the conjectured existence of STUs for local dimensions larger than $d = 4$, and, the more general (albeit significantly more complicated, if not to say almost unassailable) question of the optimal conversion of work into entanglement for arbitrary dimensions. In the area beyond the specific tasks discussed above, there is of course also a host of further open questions,

foremost, the identification of a satisfactory characterization of complexity in the context of Nernst's unattainability principle [89], and the detailed study of ensuing trade-offs between the energy costs, time requirements and complexity of operations for (ground-state) cooling, or even tasks that require additional structure such as the operation of autonomous quantum clocks [49], to name but a few.

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Part II

Selected Publications

In this second part, we present a selection of our publications that investigate questions at the intersection of quantum thermodynamics, quantum information and quantum optics. The publications are included here in chronological order, and each publication is introduced with a short overview of the content and a contribution statement.

II.1 The Thermodynamics of Creating Correlations: Limitations and Optimal Protocols

Publication:

David E. Bruschi*, Martí Perarnau-Llobet*, Nicolai Friis*, Karen V. Hovhannisyan, and Marcus Huber (*These authors contributed equally to this work.)

The thermodynamics of creating correlations: Limitations and optimal protocols

Phys. Rev. E **91**, 032118 (2015)

Publisher: American Physical Society

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Preprint: arXiv:1409.4647 [quant-ph]

Overview: In this publication, we consider the problem of maximally correlating pairs of quantum systems at fixed work cost. The systems are considered to be symmetric in the sense of having the same Hamiltonians and are assumed to be in thermal states initially. The paper presents results for the optimal conversion of work (bounded by the free-energy difference) into entanglement (as measured by the entanglement of formation) for systems of two fermionic or two bosonic modes, where the bosonic protocol is restricted to Gaussian operations but an analysis of the potential for non-Gaussian operations is included. As a main result, the paper then presents a general protocol valid for arbitrary symmetric systems for optimizing the conversion of work into general correlations (as measured by the mutual information).

Contribution: As shared first author, I was involved in deriving and formalizing the optimal protocol for creating mutual information (Sec. III of the paper), I derived the results for entanglement generation in bosonic and fermionic modes (Sec. IV of the paper), and I had a lead role in writing the paper.

The thermodynamics of creating correlations: Limitations and optimal protocols

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We establish a rigorous connection between fundamental resource theories at the quantum scale. Correlations and entanglement constitute indispensable resources for numerous quantum information tasks. However, their establishment comes at the cost of energy, the resource of thermodynamics, and is limited by the initial entropy. Here, the optimal conversion of energy into correlations is investigated. Assuming the presence of a thermal bath, we establish general bounds for arbitrary systems and construct a protocol saturating them. The amount of correlations, quantified by the mutual information, can increase at most linearly with the available energy, and we determine where the linear regime breaks down. We further consider the generation of genuine quantum correlations, focusing on the fundamental constituents of our universe: fermions and bosons. For fermionic modes, we find the optimal entangling protocol. For bosonic modes, we show that while Gaussian operations can be outperformed in creating entanglement, their performance is optimal for high energies.

I. INTRODUCTION

Correlations constitute fundamental resources for various tasks in quantum information processing [1]. In order to create the paradigmatic resource —entanglement —global operations are required. These operations come at a price: They require access to all of the subsystems of the target system and precise control over their interactions. This motivates the formulation of quantum information theory as a resource theory with respect to the limitations imposed by local operations and classical communication (LOCC) [2–5].

However, there is another price to be paid for correlating quantum systems. As any amount of correlation implies extractable work [6–10], it follows that energy is required to establish correlations. The required energy depends on the inevitable initial entropy of the system. This establishes a link to another resource theory —(quantum) thermodynamics, where the purity of the system, as well as the available free energy constitute fundamental resources due to the restrictions of the first and second laws of thermodynamics.

Recent interest in thermodynamics in the quantum domain (see, *e.g.*, [11–14]) is, in part, fueled by this interesting connection to (quantum) information and its implications for the very foundations of thermodynamic laws [15–17]. Combining the limitations of both theories shows that the resources of one theory are of great significance to the other as well. Examples range from an inevitable energy cost of measurements [18], and the role of entanglement (and other quantum effects) in thermal machines [19–25], to scenarios [26] in which thermodynamic resources play a role in the formation of entanglement and other types of shared information.

This naturally leads us to ask two fundamental questions about the physical limitations of quantum information processing: *What is the maximal amount of correlation and entanglement that can be generated for a given energy cost? How does the inevitable mixedness due to finite temperatures influence these costs, or, in other words, what is the role of the purity as a resource?* For closed systems, these questions were addressed in Ref. [26]. Here, we extend these results by (i) considering the presence of an auxiliary thermal bath, (ii) deriving fundamental bounds and optimal protocols for the creation of total correlations, and (iii) analyzing the minimal energy cost for creating genuine quantum correlations, *i.e.*, entanglement, in fermionic and bosonic systems.

First, assuming unlimited control over the system and an arbitrarily large thermal bath (see Fig. 1), we derive the ultimate limitations for any protocol to generate correlations as quantified by the mutual information. This top–down approach provides absolute bounds which cannot be outperformed, and we present a protocol for which these bounds can be saturated.

To complement these results, we then present a bottom–up approach for the generation of entanglement between fundamental physical systems—field modes with fermionic or bosonic statistics. Taking into account limitations such as superselection rules for fermions, and using experimentally feasible and widely available techniques for bosonic modes, we provide protocols for the creation of entanglement. While we find the fermionic protocols to be optimal, we show that the practical bosonic protocols become optimal only in the limit of large input energies. Surprisingly, we find that for both

* D. E. Bruschi, M. Perarnau-Llobet, and N. Friis have contributed equally to this work.

the total and genuine quantum correlations, operations involving the bath may be restricted to simple thermalization processes.

II. FRAMEWORK

Let us start by defining some of the basic notions of quantum thermodynamics. The energy E of any quantum system S is given by the expectation value of the corresponding Hamiltonian H_S in the system state ρ, that is, $E(\rho) = \mathrm{Tr}(H_S\rho)$. A crucial quantity, which we will refer to throughout this work, is the free energy F, *i.e.*,

$$F(\rho) = E(\rho) - TS(\rho)\,, \tag{1}$$

where $S(\rho) = -\mathrm{Tr}\big(\rho\ln(\rho)\big)$ is the von Neumann entropy. The free energy defines the amount of work that is extractable from a system when given access to a thermal bath at temperature T. For thermal states $\tau(\beta)$ of the form

$$\tau(\beta) = \frac{e^{-\beta H_S}}{\mathcal{Z}(\beta)}\,, \tag{2}$$

the free energy takes on its minimal value $F\big(\tau(\beta)\big) = -T\,\ln(\mathcal{Z})$, where $\mathcal{Z}$ is the partition function, $\beta = 1/T$, and we work in units where $\hbar = k_{\mathrm{B}} = 1$. For arbitrary states, $F(\rho)$ may be referred to as the nonequilibrium free energy. In the following, we consider the initial state of the system S to be thermal, $\rho_S = \tau_S(\beta)$.

We further assume that a heat bath B, that is, an arbitrarily large ancillary system in thermal equilibrium, is available. The total Hamiltonian is $H = H_S + H_B$, and the initial state can be written as $\tau_{SB}(\beta) = \tau_S(\beta)\otimes\tau_B(\beta)$. The Hilbert space $\mathcal{H}_S = \mathcal{H}_{S_1}\otimes\mathcal{H}_{S_2}$ of S is divided into two subsystems, S_1 and S_2 , which we assume to be noninteracting, such that $H_S = H_{S_1} + H_{S_2}$ and, consequently, $\tau_S(\beta) = \tau_{S_1}(\beta)\otimes\tau_{S_2}(\beta)$. These initially uncorrelated subsystems are to be correlated via a global unitary operation U_{SB} on the total Hilbert space $\mathcal{H} = \mathcal{H}_S\otimes\mathcal{H}_B$. The unitary U_{SB} is the most general operation available, assuming that S and B are isolated. Any such unitary can be thought of as a single cycle of a quantum machine. The associated energy cost W is defined as the average overall energy change,

$$W = \mathrm{Tr}\Big(H\big[U_{SB}\tau_{SB}(\beta)U_{SB}^{\dagger} - \tau_{SB}(\beta)\big]\Big) = \Delta E_S + \Delta E_B\,, \tag{3}$$

and it corresponds to the total work that needs to be performed to correlate S. Since U_{SB} leaves the total entropy of τ_{SB} invariant, W can be identified with the total change in free energy, which is minimal for the initial thermal state. Note that any initial state different from a thermal state at the temperature of the bath would provide extractable work that could be used to create correlations. To avoid this dependence on the initial state, and to properly account for the work invested in the system, we chose an initial thermal state at temperature T,

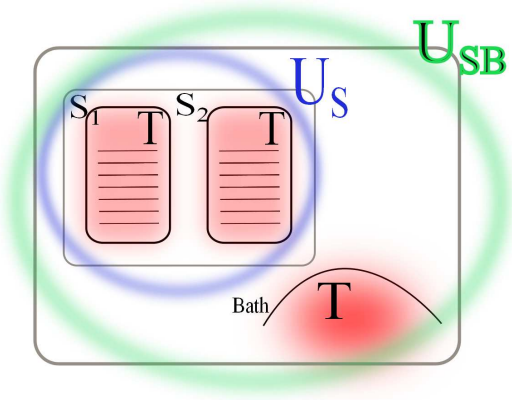

FIG. 1. **Illustration of the general setup:** Two quantum systems, S_1 and S_2, at thermal equilibrium with a bath at temperature T are acted upon either by a unitary U_S on the bipartite system or by a more general unitary U_{SB} that also involves the bath. The application of these unitaries, which correlate the system, requires a supply of external energy. In this general setting, we determine the optimal amount of correlations and entanglement that can be generated in the system for any given amount of energy.

corresponding to the temperature of the heat bath. It follows that $W \geq 0$, and hence any operation U_{SB} requires some energy. The aim of this paper is to determine how this energy may be used most efficiently to correlate the systems S_1 and S_2 .

We distinguish two kinds of correlations: total correlations, and genuine quantum correlations (entanglement). We quantify the former by the mutual information

$$\mathcal{I}_{S_1S_2}(\rho_S) = S(\rho_{S_1}) + S(\rho_{S_2}) - S(\rho_S)\,, \tag{4}$$

which measures the amount of global information shared among the systems S_1 and S_2, *i.e.*, the information encoded within the state ρ_S that is not accessible through its subsystems alone. Pure quantum states for which the mutual information is nonzero are entangled, but this is not necessarily the case for mixed states. To quantify genuine quantum correlations between S_1 and S_2, we employ the entanglement of formation (see, *e.g.*, Ref. [27] for a review of available entanglement measures), which can be defined as the minimal average mutual information across all decompositions of the mixed quantum state into pure state ensembles, *i.e.*,

$$E_{oF}(\rho_S) := \frac{1}{2}\inf_{\mathcal{D}(\rho_S)}\sum_i p_i\,\mathcal{I}_{S_1S_2}\big(|\psi_i\rangle\langle\psi_i|\big)\,, \tag{5}$$

where $\mathcal{D}(\rho_S) = \{p_i, |\psi_i\rangle \,|\, \sum_i p_i\,|\psi_i\rangle\langle\psi_i| = \rho_S\}$. In a finite-dimensional system, the entanglement of formation represents the number of maximally entangled states per copy that are needed asymptotically to create the state via LOCC.

III. CORRELATING QUANTUM SYSTEMS: ENERGY COST AND OPTIMAL PROTOCOLS

We now present our main results. We start with the top–down approach, where we determine the ultimate limitations of creating correlations, as quantified by the mutual information. Using the facts that the initial thermal state is completely uncorrelated, $S(\tau_S) = S(\tau_{S_1}) + S(\tau_{S_2})$, and that the global unitary leaves the overall entropy invariant, $S(U_{SB}\tau_{SB}U_{SB}^\dagger) = S(\tau_{SB})$, we combine Eqs. (1) and (3) to express the energy cost W in terms of the free energy difference as

$$W = \Delta F_S + \Delta F_B + T\,\mathcal{I}_{SB}\,, \tag{6}$$

obtaining a similar expression to those discussed, *e.g.*, in Refs. [6, 28–30] in related contexts. A detailed derivation of Eq. (6) can be found in Appendix B. In complete analogy to (6), we may split ΔF_S into the free energy differences of its subsystems, and their correlation as

$$\Delta F_S = \Delta F_{S_1} + \Delta F_{S_2} + T\,\mathcal{I}_{S_1S_2}\,, \tag{7}$$

for which a proof is also given in Appendix B. For any thermal state τ, the free energy difference to another (non-equilibrium) state ρ may be expressed through the relative entropy $S(\rho\|\tau) = -S(\rho) - \mathrm{Tr}(\rho\ln\tau)$ as $\Delta F = T\,S\big(\rho\|\tau(\beta)\big)$. This, in turn, allows us to write W in the form

$$\begin{aligned}\beta\,W = {} & S(\rho_{S_1}\|\tau_{S_1}) + S(\rho_{S_2}\|\tau_{S_2}) + S(\rho_B\|\tau_B)\\ & + \mathcal{I}_{S_1S_2} + \mathcal{I}_{SB}\,,\end{aligned} \tag{8}$$

where ρ_{S_1}, ρ_{S_2}, and ρ_B denote the final reduced states for the subsystems, S_1 and S_2, and the bath B, respectively. In other words, work can be invested to shift the thermal marginals away from equilibrium or to create correlations. Since all quantities on the right-hand side of Eq. (8) are non-negative, it can be immediately inferred that the following ultimate bound holds for the amount of correlation that can be generated between the subsystems for a given energy cost W and temperature $T = 1/\beta$:

$$\mathcal{I}_{S_1S_2} \leq \beta\,W\,. \tag{9}$$

Remarkably, it is possible to saturate this bound using a simple set of operations: unitary operations on S and interactions with the bath to thermalize the system. These operations are enough to obtain $W = \Delta F_S$ in (6) in the limit of an arbitrarily large bath that is complex enough to thermalize the system each time they come in contact (see Ref. [31] for a proof, and Ref. [32] for a description in terms of unitary operations). We are now ready to present the protocol achieving $W = T\mathcal{I}_{S_1S_2}$, which can be divided into two steps (see Fig. 2).

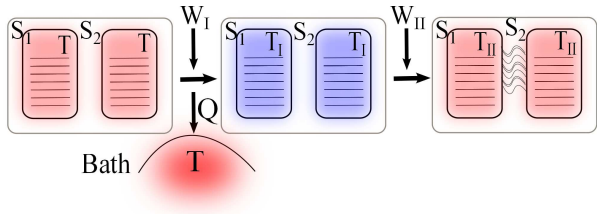

FIG. 2. **Illustration of the protocol:** In the first step the system is cooled down by a controlled interaction with the bath, and the heat Q is transferred to the bath. The associated work cost is W_{I}. In the second step, the system is isolated from the bath before it is correlated though a unitary operation, which effectively heats up the subsystems. The energy cost of the second step is W_{II}.

(I) **Cooling**: First, the temperature of S is lowered from T to $T_{\mathrm{I}} \leq T$, reducing the global entropy of the system. The (minimal) energy cost for this thermalization process is $W_{\mathrm{I}} = \Delta F_S$, *i.e.*,

$$W_{\mathrm{I}} = F\big(\tau_S(\beta_{\mathrm{I}})\big) - F\big(\tau_S(\beta)\big)\,, \tag{10}$$

where $\beta_{\mathrm{I}} = 1/T_{\mathrm{I}}$.

(II) **Correlating**: In the second step, the system is isolated from the bath and it is correlated via a unitary operation U_{corr}. Following Ref. [26], the unitary is chosen such that S_1 and S_2 are locally thermal at temperature $T_{\mathrm{II}} = 1/\beta_{\mathrm{II}} \geq T_{\mathrm{I}}$, *i.e.*,

$$\mathrm{Tr}_{S_1(S_2)}\big(U_{\mathrm{corr}}\tau_S(\beta_{\mathrm{I}})U_{\mathrm{corr}}^\dagger\big) = \tau_{S_2(S_1)}(\beta_{\mathrm{II}})\,. \tag{11}$$

This choice ensures that the systems are correlated at minimal energy cost W_{II}, see [26].

There is thus a tradeoff between the amount of work W_{I}, invested to cool down the system, which allows one to potentially obtain larger correlations, and the work W_{II}, invested to actually correlate it. As we show in detail in Appendix C, both contributions add up to

$$W = W_{\mathrm{I}} + W_{\mathrm{II}} = T\,\mathcal{I}_{S_1S_2} + T\,S\big(\tau_S(\beta_{\mathrm{II}})\|\tau_S(\beta)\big)\,. \tag{12}$$

Therefore, optimality is achieved when the local temperature of the final state marginals is identical to the initial temperature, $T_{\mathrm{II}} = T$, such that $W = T\mathcal{I}_{S_1S_2}$.

However, it may occur that this condition would require more energy to be used in the first step than is needed to reach the ground state. In such a case, the excess energy can be put to better use further correlating the final state, raising the local temperatures of the subsystems beyond $T_{\mathrm{II}} = T$. These considerations yield a more precise bound (see Appendix C), given by

$$\mathcal{I}_{S_1S_2} \leq \begin{cases}\beta W & \text{if } \beta W \leq S\big(\tau_S(\beta)\big)\,,\\ S(\tau_S(\beta_{\mathrm{II}})) & \text{if } \beta W > S\big(\tau_S(\beta)\big)\,,\end{cases} \tag{13}$$

where $\beta_{\rm II}$ is given by the implicit relation $E\big(\tau_S(\beta_{\rm II})\big) = W + F\big(\tau_S(\beta)\big)$. There are hence two distinct regimes. When an energy smaller than $TS\big(\tau_S(\beta)\big)$ is supplied, the correlations scale linearly with the work input. As more energy is provided, additional work needs to be invested to move the states further out of local equilibrium, leading to noticeably different behavior. For instance, for two bosonic modes, the correlations scale logarithmically with the work input for $\beta W \gg S\big(\tau_S(\beta)\big)$, as we show in Appendix D.

Finally, it is worth mentioning that our protocol is extendible to nonequilibrium initial states. One then needs to first extract the work content of the state, which leaves it in a thermal state at the temperature of the bath. Our protocol can then be readily applied using the extracted work in addition to any externally supplied energy to correlate the system.

IV. ENERGY COST OF ENTANGLEMENT GENERATION

Having provided general bounds on the energy cost of correlating two arbitrary systems, we now turn to the case of genuine quantum correlations, *i.e.*, entanglement. Here the situation is much more complex. Even determining whether a given quantum state is separable or not is generally NP hard. Therefore, obtaining a general solution for arbitrary systems is a daunting task that seems intractable. We therefore complement the previous top-down approach for general correlations by pursuing a bottom-up strategy to investigate the energy cost for generating entanglement. We focus our attention on two physically relevant cases, namely, systems of two fermionic or bosonic modes. For the low-dimensional fermionic problem and the case of bosonic Gaussian states, computing the entanglement of formation in Eq. (5) becomes feasible.

Besides making the problem more tractable, the very interesting features of bosonic and fermionic systems further motivate our choice. On one hand, modes of quantum fields play a fundamental role in the description of nature in the context of (relativistic) quantum theory. Hence, they provide a more general framework for our analysis than systems with a fixed number of particles, which appear as secondary quantities, *i.e.*, as excitations of the modes in question. On the other hand, this approach allows us to analyze the role of fermionic and bosonic particle statistics, and the corresponding finite and infinite-dimensional Hilbert spaces for two modes. In addition, the formulation in terms of individual mode operators naturally lends itself to the Hamiltonian structure, giving a clear interpretation to the involved energy costs.

In this section, we consider protocols along the same lines as previously, *i.e.*, first varying the temperature of the systems (not necessarily symmetrically) and then correlating them via unitary operations. This choice is well justified because any other operation that would either create correlations between the system and the bath or significantly change the state of the bath would have a higher energy cost, as can be seen from Eq. (6).

A. Fermionic systems

We now consider a finite-dimensional system, two modes of (equal) frequency ω of an uncharged, noninteracting fermionic field. On one hand, the simplicity of this system allows us to determine the amount of entanglement that may be generated for any given amount of energy. On the other hand, several conceptually interesting features arise from the fermionic algebra, that is, the mode operators b_1, $b_1^\dagger$, b_2, and $b_2^\dagger$ satisfy the anticommutation relations $\{\, b_m \,, b_n^\dagger \,\} = \delta_{mn}$ and $\{\, b_m \,, b_n \,\} = 0$, where $m, n = 1, 2$. The Hamiltonian of the system is (up to a constant) given by $H_S = H_{S_1} + H_{S_2} = \omega\big(b_1^\dagger b_1 + b_2^\dagger b_2\big)$. To distinguish the fermionic and bosonic case, we denote the fermionic Fock states by double-lined kets, *e.g.*, the vacuum state is written as $\|0\rangle\!\rangle$. The single-particle states are obtained by the action of the creation operators, *i.e.*, $\|1_m\rangle\!\rangle = b_m^\dagger \|0\rangle\!\rangle$. We define the two-particle state via $\|1_{S_1}\rangle\!\rangle\,\|1_{S_2}\rangle\!\rangle = b_1^\dagger b_2^\dagger \|0\rangle\!\rangle$, where we have omitted the symbol for the antisymmetrized tensor product on the left-hand side (see Refs. [33] or [34, pp. 37] for more details on the notation used here and the fermionic Fock space). The system we investigate here obeys Fermi-Dirac statistics, and the partition function is hence $\mathcal{Z}_{\rm FD}(\beta) = \big(1 + e^{-\beta}\big)$, and we specify temperatures in units of ω [recall, that ($\hbar = k_{\rm B} = 1$] from now on. The average initial particle numbers are given by $N_{S_1(S_2)} = {\rm Tr}\big(b_{1(2)}^\dagger b_{1(2)} \tau_S\big)$. The fermionic two-mode thermal state may then be expressed as

$$\tau_S = \frac{e^{-\beta}}{\mathcal{Z}_{\rm FD}^2}\Big(e^{\beta}\,\|0\rangle\!\rangle\langle\!\langle 0\| + \|1_{S_1}\rangle\!\rangle\langle\!\langle 1_{S_1}\| + \|1_{S_2}\rangle\!\rangle\langle\!\langle 1_{S_2}\| + e^{-\beta}\,\|1_{S_1}\rangle\!\rangle\|1_{S_2}\rangle\!\rangle\langle\!\langle 1_{S_2}\|\langle\!\langle 1_{S_1}\|\Big)\,. \tag{14}$$

With these preliminaries at hand, we consider protocols along the lines of that presented in Section III to create entanglement. In the first step of such a procedure, using the interaction with the bath, the temperature of the two modes is lowered as before, which manifests in altered particle numbers $N_{S_1}^{\rm I}$ and $N_{S_2}^{\rm I}$. The energy cost $W_{\rm I}$ for this step is given by the free energy difference to the transformed state.

In the second step of the protocol, unitaries on the two-mode space S are applied to correlate the system. In the case of fermionic modes, these operations are further restricted by superselection rules. Since the state of any single fermion acquires a phase of π upon a rotation around 2π, rotational symmetry prohibits coherent superpositions of even and odd numbers of fermions. Moreover, the superselection rules modify the definition of the entanglement of formation of Eq. (5) in the sense

that the minimization is carried out only over pure state ensembles that respect superselection [35]. We hence take as a measure of entanglement the minimum number, per copy, of maximally entangled states of the two fermionic modes, which are needed to assemble a given two-mode state. As is shown in Appendix E, this well-defined measure of entanglement can be expressed by the energy cost W_{II} of the correlating step as

$$E_{oF} = \ln(2)\sqrt{\frac{W_{\mathrm{II}}}{\omega}}\sqrt{2\frac{e^{\beta_{\mathrm{I}}}-1}{e^{\beta_{\mathrm{I}}}+1}-\frac{W_{\mathrm{II}}}{\omega}}\,. \tag{15}$$

Similar to the previous section, we determine the optimal splitting of W into W_{I} and W_{II}, and we express it in terms of the optimal final temperature T_{II}. The results of this numerical optimization are presented in Fig. 3. Although the protocol is very similar to that for the generation of mutual information, optimality is not achieved for $T_{\mathrm{II}} = T$, but rather when $T_{\mathrm{II}} \geq T$, see Fig. 3 (b).

One can further improve upon these results by taking advantage of the peculiar properties of fermionic entanglement, in particular the existence of mixed, maximally entangled states [36]. These particularities may occur because the subspaces of even and odd fermion numbers decouple. Consequently, no unitaries may introduce correlations between these subspaces. The optimally correlating unitary U_{corr} can therefore be decomposed into two independent rotations. Furthermore, we find that altering the temperatures of the subsystems asymmetrically, *i.e.*, cooling one mode while heating the other, can be beneficial. Allowing for such asymmetric temperatures, we numerically optimize the fermionic entanglement of formation generated at a fixed energy cost. The results are discussed in detail in Appendix E.

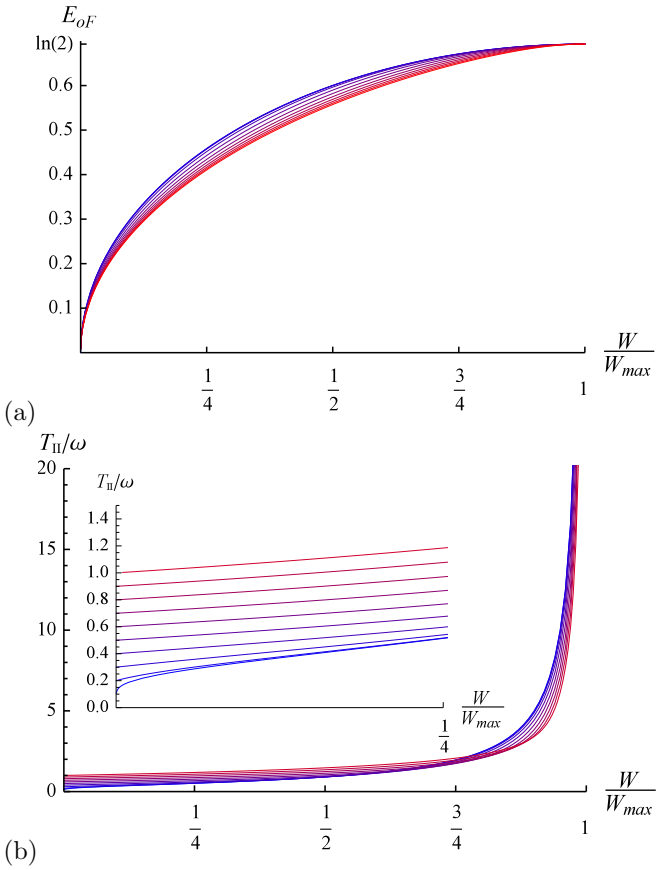

FIG. 3. **Fermionic entanglement cost**: The solid curves in Fig. 3 (a) show the amount of entanglement (of formation) that can maximally be generated in the even subspace of two fermionic modes that are initially in a thermal state, for a given energy cost W. The curves are plotted for initial temperatures varying from $T = 0$ to $T = 1$ in steps of 0.1 (top to bottom) in units $\hbar\omega/k_{\mathrm{B}}$. The horizontal axis shows the relative energy cost, *i.e.*, the fraction of W and the minimal energy cost $W_{\max} = \left[2T\ln\left(e^{\beta}+1\right)-\omega\right]$ to generate a maximally entangled pure state. Fig. 3 (b) shows the corresponding effective final temperature $T_{\mathrm{II}} \geq T$ of the marginals after the protocol.

B. Bosonic systems

Let us now investigate the optimal generation of entanglement for a bosonic system. Analogously to the fermionic case, we consider two modes of an uncharged, noninteracting bosonic field. We assume that these modes, again labeled S_1 and S_2, have the same frequency ω. The corresponding annihilation and creation operators a_1, $a_1^\dagger$, a_2, and $a_2^\dagger$ satisfy the commutation relations $[a_m, a_n^\dagger] = \delta_{mn}$ and $[a_m, a_n] = 0$, where $m, n = 1, 2$. The system Hamiltonian may be written in terms of these operators (up to a constant) as $H_S = H_{S_1} + H_{S_2} = \omega\left(a_1^\dagger a_1 + a_2^\dagger a_2\right)$. The infinite-dimensional Fock space of these two modes is spanned by the vacuum state $|0\rangle$, which is annihilated by a_1 and a_2, and the particle states, which are obtained by applying the creation operators $a_1^\dagger$ and $a_2^\dagger$ to the vacuum. The bosonic excitations obey Bose-Einstein statistics, where the partition function is given by $\mathcal{Z}_{\mathrm{BE}}(\beta) = \left(1-e^{-\beta}\right)^{-1}$. Note that the temperatures are again given in units of ω and we have set $\hbar = k_{\mathrm{B}} = 1$.

To handle this infinite-dimensional system, we will restrict our analysis of entanglement generation to Gaussian states, which commonly feature in applications in quantum information [37] and quantum computing [38], to name but a few. The correlations of two-mode Gaussian states can be completely described by a real, 4×4 covariance matrix σ_S. This matrix collects the expectation values of quadratic combinations of the mode operators—the second moments—and we may assume that the expectation values of all linear combinations of mode operators—the first moments—vanish. For a given state ρ_S, the components of σ_S are $(\sigma_S)_{mn} = \mathrm{Tr}\left(\{\mathbb{X}_m, \mathbb{X}_n\}\,\rho_S\right)$, with the quadrature operators $\mathbb{X}_{(2n-1)} = (a_n + a_n^\dagger)/\sqrt{2}$ and $\mathbb{X}_{(2n)} = -i(a_n - a_n^\dagger)/\sqrt{2}$, and $m, n = 1, 2$. For the initial thermal state at temperature T that we consider

here, the covariance matrix is proportional to the identity operator, $\sigma_S = \nu(T)\,\mathbb{1}_4$, where the symplectic eigenvalue ν is given by $\nu(T) = \coth(\beta/2)$.

In the first step of the protocol to optimally generate entanglement, the initial temperature is lowered from T to $T_{\rm I} < T$, after which the state is represented by $\sigma_S^{\rm I} = \nu^{\rm I}\,\mathbb{1}_4$, where $\nu^{\rm I} = \nu(T_{\rm I})$. The energy cost for this step is given by

$$\frac{W_{\rm I}}{\omega} = \nu^{\rm I} - \nu(T) - 2\beta^{-1}\Big[f\big(\nu^{\rm I}\big) - f\big(\nu(T)\big)\Big]\,, \qquad (16)$$

where the entropy of a two-mode thermal state represented by σ is expressed as $S(\sigma) = 2f(\nu) = (\nu+1)\ln\big(\frac{\nu+1}{2}\big) - (\nu-1)\ln\big(\frac{\nu-1}{2}\big)$.

In the second step of the protocol, we restrict the entangling unitaries to Gaussian operations, which may be represented as linear transformations of the mode operators. Since the initial covariance matrix is proportional to that of the vacuum, the final covariance matrix must be proportional to that of a pure, two-mode Gaussian state, which is locally equivalent to a two-mode squeezed state. We may therefore conclude that the optimal Gaussian entangling operations for this situation are two-mode squeezing transformations. Moreover, throughout the protocol, the state remains symmetric with respect to the two subsystems, that is, their entropies are identical. For such states, all entanglement measures depend on a single parameter $\tilde{\nu}_-$, the smallest symplectic eigenvalue of the partial transpose. In terms of $\tilde{\nu}_-$, the entanglement of formation takes the form

$$E_{oF} = \begin{cases} \mathfrak{h}(\tilde{\nu}_-)\,, & \text{if } 0 \le \tilde{\nu}_- < 1\,, \\ 0\,, & \text{if } \tilde{\nu}_- \ge 1\,, \end{cases} \qquad (17)$$

where $\mathfrak{h}(x) = h_+(x)\ln\big(h_+(x)\big) - h_-(x)\ln\big(h_-(x)\big)$, and $h_\pm(x) = \frac{(x\pm1)^2}{4x}$. One may also relate $\tilde{\nu}_-$ to the squeezing parameter r of the thermal two-mode squeezed state after step II via $e^{-2r} = \tilde{\nu}_-/\nu^{\rm I}$, while the final state energy is given by $\omega\big(\nu^{\rm I}\cosh(2r) - 1\big)$. With this, the energy cost for step II can be expressed as

$$\frac{W_{\rm II}}{\omega} = \frac{(\nu^{\rm I})^2}{2\tilde{\nu}_-}\Big[\frac{\tilde{\nu}_-}{\nu^{\rm I}} - 1\Big]^2\,. \qquad (18)$$

Conversely, Eq. (18) allows us to express $\tilde{\nu}_-$, and hence E_{oF}, in terms of $\nu^{\rm I}$ and $W_{\rm II} = W - W_{\rm I}$. The results of the numerical optimization of the entanglement of formation over $\nu^{\rm I}$ are shown in Fig. 4. Note that in contrast to the fermionic case, here we find $T_{\rm II} < T$. Another interesting feature of the bosonic system is that for nonzero initial temperatures, entanglement cannot be generated for arbitrarily small amounts of supplied energy [39]. Instead, entanglement is only created when the constraint $(\nu^{\rm I} - 1)^2 < 2W_{\rm II}/\omega$ is satisfied.

Finally, a comment about the optimality of Gaussian operations is in order. As we show in detail in Appendix F, there are two energy regimes. In the low-energy regime, Gaussian operations may be outperformed by non-Gaussian operations in generating entanglement. We provide a protocol which achieves this, and allows leaving the separable states even for arbitrarily small amounts of supplied energy. In the high-energy regime, on the other hand, Gaussian operations are shown to be asymptotically optimal for the generation of entanglement. This can be understood in the following way. When enough energy is supplied, the ground state is reached in the cooling phase. All remaining energy can then be optimally used for Gaussian entangling operations. When large amounts of energy are invested, the fraction of the energy that is suboptimally spent in the cooling stage becomes negligible, vanishing in the limit of an infinite energy supply.

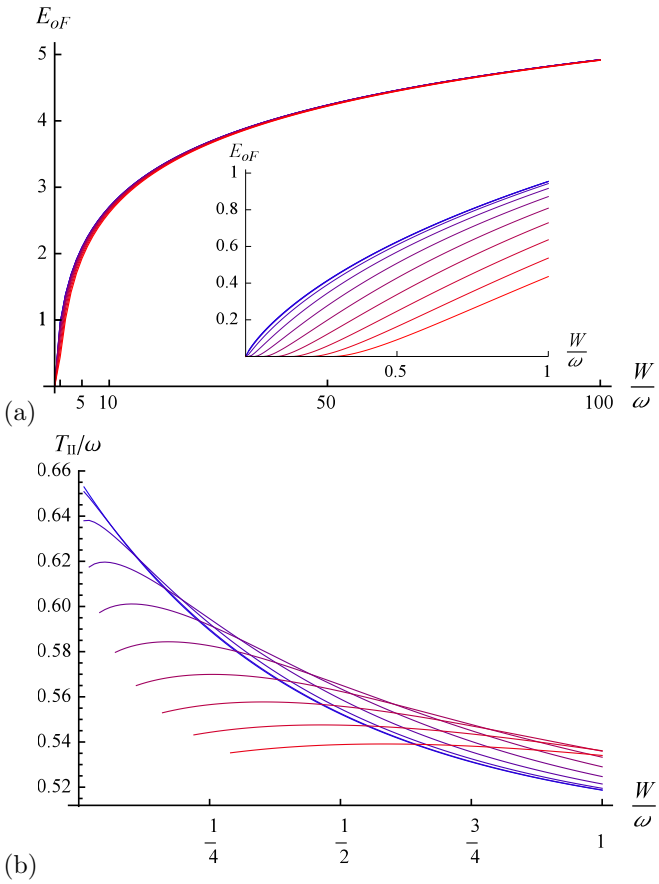

FIG. 4. **Optimal bosonic entanglement**: The curves in Fig. 4 (a) show the optimal amount of entanglement (of formation) that can be generated by Gaussian operations on two bosonic modes, s_1 and s_2, of frequency ω, which are initially in a thermal state of temperature T. The horizontal axis shows the supplied energy W in units of ω. Fig. 4 (b) shows the local temperature $T_{\rm II}$ of the modes after the protocol for values of W for which entanglement can be generated. The curves in both Fig. 4 (a) and (b) are plotted for initial temperatures varying from $T = 0$ to $T = 1$ in steps of 0.1 (top to bottom) in units $\hbar\omega/k_{\rm B}$.

V. CONCLUSION

We have investigated the equivalence between free energy and the ability to create correlations in quantum systems. Any amount of correlation implies that extractable work is present in the system. Conversely, the creation of any amount of correlation comes at the price of investing work. Following this premise, we have introduced protocols that are optimal for the generation of correlations, as well as genuine quantum correlations, at minimal energy cost. For total correlations, as quantified by the mutual information, we have presented a protocol that is optimal for arbitrary bipartite systems.

For the case of genuine quantum correlations — entanglement, the paradigmatic quantum resource — we have focused on two fermionic or two bosonic modes. For both types of systems, we have derived optimal protocols for the generation of entanglement as quantified by the well-known entanglement of formation. In the case of bosons, we have restricted the optimization to the set of Gaussian operations for the sake of feasibility. To place this choice in an appropriate context, we have also discussed explicit protocols that make use of non-Gaussian operations, showing that they can decrease the energy cost when the available energy is small. Nonetheless, our findings further show that Gaussian operations become optimal in the limit of large available energies. A common feature of all the mentioned protocols is their remarkably simple structure. They make use of the interaction with a thermal bath to cool (or heat) the (sub)system, which, interestingly, requires only elementary thermalization processes, before introducing correlations.

Our results connect two important resource theories, revealing the implicit thermodynamical cost and value of quantum correlations. While we have focused our efforts on bipartite quantum systems, the results concerning correlations have the potential for a straightforward generalization to the multipartite case when considering correlations quantified by $S(\rho) - \sum_i S(\rho_i)$ where $\rho_i = \mathrm{Tr}_{j\neq i}(\rho)$. Such considerations are possible extensions of our work, especially when connected to cases of multipartite entanglement generation. Here, the focus on bipartite entanglement has guaranteed the utility of the created resources for quantum communication, whereas future work concerning multipartite entanglement should be approached with great care, as generic generation of entanglement may be less useful than previously believed [40]. Other possible directions inspired by our work include similar considerations for single-shot scenarios as, *e.g.*, in Refs. [15, 31, 41], which effectively means focusing on different entropies in the mutual information, the inclusion of catalytic systems [15], or even the interesting connection with the thermodynamic properties of transformations induced by nonuniform motion [42, 43].

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APPENDIX

A: Preliminaries

Before we present detailed proofs for the main results, let us review some preliminary concepts. First, recall that the free energy of a state ρ is given by

$$F(\rho) = E(\rho) - TS(\rho) = \mathrm{Tr}\big(\rho H\big) + T\,\mathrm{Tr}\big(\rho \ln \rho\big)\,. \tag{A1}$$

For a thermal state, $\tau(\beta) = e^{-\beta H}/\mathcal{Z}$, with the partition function $\mathcal{Z} \in \mathbb{R}$, and $\beta = 1/T$, where we have set $k_{\mathrm{B}} = 1$, the free energy reduces to

$$F\big(\tau(\beta)\big) = -T\,\ln \mathcal{Z}\,. \tag{A2}$$

Moving a thermal state away from equilibrium always requires work, which is given by the free energy difference

$$\Delta F\big(\tau(\beta) \to \rho\big) = F(\rho) - F(\tau) \tag{A3}$$

to the final state ρ. An elementary way to see that $\Delta F \geq 0\ \forall\ \rho$ for initial thermal states is via the relative entropy $S(\rho\|\tau)$, defined as

$$S(\rho\|\tau) = -S(\rho) - \mathrm{Tr}\big(\rho \ln \tau\big)\,. \tag{A4}$$

For thermal states $\tau(\beta)$ we may then write

$$\begin{aligned} T\,S\big(\rho\|\tau(\beta)\big) &= -T\,S(\rho) \,-\, T\,\mathrm{Tr}\big(\rho\ln\tau(\beta)\big) \\ &= -T\,S(\rho) \,+\, \mathrm{Tr}(\rho H) \,+\, T\,\ln\mathcal{Z}\,\mathrm{Tr}(\rho) \\ &= F(\rho) \,-\, F\big(\tau(\beta)\big) \\ &= \Delta F\big(\tau(\beta)\to\rho\big)\,. \end{aligned} \tag{A5}$$

By virtue of Klein's inequality (see, *e.g.*, Ref. [44]), the quantum relative entropy is non-negative, $S(\rho\|\tau)\geq 0$, and vanishes if and only if $\rho=\tau$. Consequently, we can conclude that $\Delta F\big(\tau(\beta)\to\rho\big)\geq 0$.

B: Energy cost of a general unitary

We now give a detailed proof of Eq. (6), where we denote the transformed states of the system, the subsystems, and the bath as ρ_S, ρ_{S_1}, ρ_{S_2}, and ρ_B, respectively. Starting from Eq. (3), the energy differences are rewritten in terms of the changes in free energy and entropy as

$$\begin{aligned} W &= \Delta E_S + \Delta E_B \\ &= \Delta F_S + \Delta F_B \,+\, T\left[S(\rho_S)+S(\rho_B)-S(\tau_S)-S(\tau_B)\right] \\ &= \Delta F_S + \Delta F_B \,+\, T\left[S(\rho_S)+S(\rho_B)-S(\tau_{SB})\right] \\ &= \Delta F_S + \Delta F_B \,+\, T\left[S(\rho_S)+S(\rho_B)-S(\rho_{SB})\right] \\ &= \Delta F_S + \Delta F_B \,+\, T\,\mathcal{I}_{SB}\,, \end{aligned} \tag{B1}$$

where we have made use of the fact that the global unitary leaves the overall entropy unchanged, $S(\rho_{SB}) = S(\tau_{SB})$. To prove the similar result of Eq. (7) for the partition of the system S into its subsystems we first write

$$\begin{aligned} \Delta F_S &= \Delta E_S \,-\, T\,\Delta S_S \\ &= \Delta E_{S_1} + \Delta E_{S_2} - T\left[S(\rho_S)-S(\tau_S)\right]. \end{aligned} \tag{B2}$$

The energy differences of the subsystems may then be expressed as

$$\Delta E_{S_1} \;=\; \Delta F_{S_1} + T\left[S(\rho_{S_1})-S(\tau_{S_1})\right], \tag{B3a}$$

$$\Delta E_{S_2} \;=\; \Delta F_{S_2} + T\left[S(\rho_{S_2})-S(\tau_{S_2})\right]. \tag{B3b}$$

Finally, noting that $S(\tau_{S_1})+S(\tau_{S_2})=S(\tau_S)$, one arrives at

$$\begin{aligned} \Delta F_S &= \Delta F_{S_1} + \Delta F_{S_2} + T\left[S(\rho_{S_1})+S(\rho_{S_2})-S(\rho_S)\right] \\ &= \Delta F_{S_1} \,+\, \Delta F_{S_2} \,+\, T\,\mathcal{I}_{S_1S_2}\,, \end{aligned} \tag{B4}$$

which concludes the proof.

C: Optimal protocol for generating mutual information

Let us now turn our attention to the protocol for the optimal generation of correlations. We prove here that the ultimate bound $W = T\mathcal{I}_{S_1S_2}$ can be achieved, by first proving Eq. (12). The (minimal) energy cost W_{I} for the first step, reducing the system temperature from T to $T_{\mathrm{I}}\leq T$, is given by

$$\begin{aligned} W_{\mathrm{I}} = \Delta F_S\big(\tau_S(\beta)\to\tau_S(\beta_{\mathrm{I}})\big) \;&=\; E\big(\tau_S(\beta_{\mathrm{I}})\big) - E\big(\tau_S(\beta)\big) \\ &- T\left[S\big(\tau_S(\beta_{\mathrm{I}})\big) - S\big(\tau_S(\beta)\big)\right]. \end{aligned} \tag{C1}$$

For the second step we use a unitary operation, which leaves the system entropy invariant, while the subsystems become locally thermal at temperature $T_{\mathrm{II}} = 1/\beta_{\mathrm{II}}$. The average energy of the system after the transformation is hence identical to that of a thermal state $\tau_S(\beta_{\mathrm{II}})$. The minimal energy cost W_{II} is hence given by

$$W_{\mathrm{II}} = E\big(\tau_S(\beta_{\mathrm{II}})\big) - E\big(\tau_S(\beta_{\mathrm{I}})\big)\,. \tag{C2}$$

The correlations of the final state, as measured by the mutual information, are then

$$\begin{aligned} \mathcal{I}_{S_1S_2} &= S\big(\tau_{S_1}(\beta_{\mathrm{II}})\big) + S\big(\tau_{S_2}(\beta_{\mathrm{II}})\big) - S\big(\tau_S(\beta_{\mathrm{I}})\big) \\ &= S\big(\tau_S(\beta_{\mathrm{II}})\big) - S\big(\tau_S(\beta_{\mathrm{I}})\big)\,. \end{aligned} \tag{C3}$$

Using Eq. (C3), the energy costs for both steps can be combined to arrive at

$$\begin{aligned} W = W_{\mathrm{I}} + W_{\mathrm{II}} \;&=\; E\big(\tau_S(\beta_{\mathrm{II}})\big) - E\big(\tau_S(\beta)\big) \\ &- T\left[S\big(\tau_S(\beta_{\mathrm{II}})\big) - S\big(\tau_S(\beta)\big) - \mathcal{I}_{S_1S_2}\right] \\ &= \Delta F_S\big(\tau_S(\beta)\to\tau_S(\beta_{\mathrm{II}})\big) \,+\, T\,\mathcal{I}_{S_1S_2} \\ &= T\left[S\big(\tau_S(\beta_{\mathrm{II}})\|\tau_S(\beta)\big) \,+\, \mathcal{I}_{S_1S_2}\right]. \end{aligned} \tag{C4}$$

Now, if W is split into the contributions W_{I} and W_{II} such that $\beta_{\mathrm{II}}=\beta$, one obtains $T\mathcal{I}_{S_1S_2}=W$, as desired. Interestingly, this is not always achievable. Setting $\beta_{\mathrm{II}}=\beta$ may require W_{I} to become larger than the energy that is necessary to cool down to the ground state. This leads to a surplus of energy for the correlation step. In such a case, T_{II} is larger than the initial temperature T. The transition to this regime occurs when,

$$W = \tilde{W} = \tilde{W}_{\mathrm{I}} \,+\, \tilde{W}_{\mathrm{II}} \;=\; T\,S\big(\tau_S(\beta)\big)\,, \tag{C5}$$

where $\tilde{W}_{\mathrm{I}} = -F\big(\tau_S(\beta)\big)$ corresponds to the energy necessary to cool down to the ground state and $\tilde{W}_{\mathrm{II}} = E\big(\tau_S(\beta)\big)$ is the work necessary to correlate the systems

such that $\beta_{\rm II} = \beta$. After some rearranging, one obtains

$$\mathcal{I}_{S_1S_2} \leq \begin{cases} \beta W & \text{if } \beta W \leq S\big(\tau_S(\beta)\big)\,, \\ S(\tau_S(\beta_{\rm II})) & \text{if } \beta W > S\big(\tau_S(\beta)\big)\,, \end{cases} \tag{C6}$$

where $\beta_{\rm II}$ is given by the implicit relation

$$E\big(\tau_S(\beta_{\rm II})\big) = W + F\big(\tau_S(\beta)\big)\,. \tag{C7}$$

There are thus two fundamentally different regimes for the generation of mutual information.

D: Generation of mutual information between two bosonic modes

Let us examine more closely the scaling of the generated correlations with the input energy. Since the amount of energy that may be used to correlate two fermionic modes is finite, we will focus on the system of two bosonic modes as described in Section IV B. Recall that the system Hamiltonian is given by $H_S = H_{S_1} + H_{S_2}$. Up to a constant, the subsystem Hamiltonians may be expressed in terms of the Fock states $|n_{S_{1(2)}}\rangle = (1/\sqrt{n!})(a^\dagger_{1(2)})^n\,|0\rangle$ as

$$H_{S_{1(2)}} = \sum_{n=0}^{\infty} n\omega\,|n_{S_{1(2)}}\rangle\langle n_{S_{1(2)}}|\,, \tag{D1}$$

and we use units where $\hbar = 1$. Likewise, the initial thermal state $\tau_S(\beta) = \tau_{S_1}(\beta)\otimes\tau_{S_2}(\beta)$ can be expressed in this way, *i.e.*,

$$\tau_{S_{1(2)}}(\beta) = \sum_{n=0}^{\infty} p_n(\beta)\,|n_{S_{1(2)}}\rangle\langle n_{S_{1(2)}}|\,, \tag{D2}$$

where $p_n = (1-e^{-\beta})e^{-n\beta}$, with $\beta = 1/T$, and temperatures in units of ω. The energy and entropy of the thermal state evaluates to

$$E\big(\tau_S(\beta)\big) = \mathrm{Tr}\big(H_S\tau_S(\beta)\big) = \omega\Big[\coth\big(\beta/2\big) - 1\Big]\,, \tag{D3}$$

$$S\big(\tau_S(\beta)\big) = -\mathrm{Tr}\big(\tau_S\ln(\tau_S)\big) = 2f\Big(\coth\big(\beta/2\big)\Big)\,, \tag{D4}$$

where $f(x)$ is the entropic function

$$f(x) = \frac{x+1}{2}\ln\Big(\frac{x+1}{2}\Big) - \frac{x-1}{2}\ln\Big(\frac{x-1}{2}\Big)\,. \tag{D5}$$

As we have argued in Eq. (13), the optimal mutual information that may be generated from such a thermal state using energies W smaller than $S\big(\tau_S(\beta)\big)/\beta$ scales linearly with W.

Let us now consider the regime where the supplied energy W is much larger than $S\big(\tau_S(\beta)\big)/\beta$. After reaching the ground state in the first step of the protocol, all of the excess energy increases the correlations. The energy of the final state is equal to the work invested into the correlation step, *i.e.*, $E\big(\tau_S(\beta_{\rm II})\big) = W_{\rm II}$. From Eq. (D3), we hence find

$$\coth\Big(\frac{\beta_{\rm II}}{2}\Big) = \frac{W_{\rm II}}{\omega} + 1\,. \tag{D6}$$

From Eq. (C6) we infer that the mutual information is given by $\mathcal{I}_{S_1S_2} = S(\tau_S(\beta_{\rm II}))$. Inserting into Eq. (D4) and expanding $f\big((W_{\rm II}/\omega)+1\big)$ into a Taylor-Maclaurin series for $(\omega/W_{\rm II}) \ll 1$, we find

$$\mathcal{I}_{S_1S_2} = 2 + 2\ln\Big(\tfrac{1}{2}\frac{W_{\rm II}}{\omega}\Big) + \mathcal{O}\Big(\frac{\omega}{W_{\rm II}}\Big)\,, \tag{D7}$$

where $\mathcal{O}(x)$ is a quantity such that $\mathcal{O}(x)/x$ remains finite in the limit $x \to 0$. We conclude that for large energy supply, the optimally generated correlations increase only logarithmically with increasing energy, in stark contrast to the linear increase at small energies, see Fig. 4 (a).

E: Optimal protocol for fermionic entanglement of formation

We now present a modification of our previous protocol for the generation of entanglement between two fermionic modes. To optimally convert the supplied energy into fermionic entanglement of formation, the temperatures of the two modes are allowed to change independently of each other in the first step of the protocol. In particular, this entails heating as well as cooling of the individual modes, and the average particle numbers $N^{\rm I}_{S_1}$ and $N^{\rm I}_{S_2}$ may be different from each other. As before, the energy cost $W_{\rm I}$ for this step is given by the free energy difference of the initial thermal and the transformed state.

For step II of the protocol, the two modes are correlated using unitary operations on the system only. As mentioned before, the superselection rules forbid coherent superpositions between even and odd numbers of fermions. In particular, the maximally entangled two-mode pure states for the even parity subspace, $\|\phi^\pm\rangle\rangle = \frac{1}{\sqrt{2}}\big(\|0\rangle\rangle \pm \|1_{S_1}\rangle\rangle\|1_{S_2}\rangle\rangle\big)$, and those for the odd parity subspace, $\|\psi^\pm\rangle\rangle = \frac{1}{\sqrt{2}}\big(\|1_{S_1}\rangle\rangle \pm \|1_{S_2}\rangle\rangle\big)$, may not be interconverted by parity conserving operations. These states may hence be regarded as forming a maximally entangled set [45]. Consequently, the optimally correlating unitary $U_{\rm corr}$ for two modes decomposes into a direct sum of two $SU(2)$ rotations. For each, only one real parameter, denoted by $\theta_{\rm even}$ and $\theta_{\rm odd}$, respectively, is relevant for the amount of generated entanglement. We quantify the entanglement by the superselected entanglement of formation, *i.e.*, the minimum number, per copy, of the aforementioned maximally entangled states respecting superselection rules, that are required to assemble a given two-mode state.

However, note that the imposed superselection rules also prevent local changes of basis for each fermionic mode. The states $\|\phi^\pm\rangle\rangle$ and $\|\psi^\pm\rangle\rangle$ could therefore be

considered to be entangled only in a mathematical sense, that is, the entanglement may not be directly used, for instance, to violate a Bell inequality. Nonetheless, if the entanglement is extracted by swapping it to a bosonic system, it becomes useful in the conventional sense. Since a swap using local unitaries cannot create entanglement, its origin must lie in the original fermionic entanglement. Keeping this argument in mind, a pure state decomposition of the transformed state that requires the fewest copies of the maximally entangled pure states $\|\phi^{\pm}\rangle\rangle$ and $\|\psi^{\pm}\rangle\rangle$ may easily be found, yielding the entanglement of formation

$$E_{oF} = \ln(2)\big[|1 - N^{\mathrm{I}}_{S_1} - N^{\mathrm{I}}_{S_2}|\sin(2\theta_{\mathrm{even}}) + |N^{\mathrm{I}}_{S_1} - N^{\mathrm{I}}_{S_2}|\sin(2\theta_{\mathrm{odd}})\big]\,, \tag{E1}$$

where $0 \leq \theta_{\mathrm{even}}, \theta_{\mathrm{odd}} \leq \pi/4$. Since the odd-subspace rotation shifts excitations of equal frequency, θ_{odd} does not contribute to the energy cost of the second step, which is given by

$$\frac{W_{\mathrm{II}}}{\omega} = 2\big(1 - N^{\mathrm{I}}_{S_1} - N^{\mathrm{I}}_{S_2}\big)\sin^2(\theta_{\mathrm{even}})\,. \tag{E2}$$

We may hence set $\theta_{\mathrm{odd}} = \pi/4$ at no additional expense in energy. We note that this suggests a tradeoff between creating entanglement in the even and odd subspace by heating one mode, while the other is cooled. The entanglement of formation becomes maximal when enough energy is supplied to cool one mode, we assume here S_1, to the ground state, while $\theta_{\mathrm{even}} = \frac{\pi}{4}$. The minimum energy W_{opt} for which this is the case is obtained when the reduced state of the second mode S_2 is maximally mixed. If less energy than W_{opt} is supplied, it is split between cooling and heating the modes S_1 and S_2, respectively, in step I, before correlating them in step II. The resulting state is a mixed state that is entangled both in the even and odd subspace. When $W = W_{\mathrm{opt}}$, the weights of the even and odd subspace entangled states are equal.

As more energy is provided, it may be used to shift the entanglement to one of the subspaces, obtaining a final state with higher purity. When $W = W_{\mathrm{max}}$, where $W_{\mathrm{max}} = W_{\mathrm{opt}} + T\ln(2) = 2T\ln(e^{\beta}+1) - \omega$, the final overall state is pure, but the entropy of both subsystems is maximal. The exact values of $N^{\mathrm{I}}_{S_1}$, $N^{\mathrm{I}}_{S_2}$, and θ_{even} may be determined by numerical optimization for fixed values of W and T. In Fig. 5, the protocol is illustrated for various temperatures, where the excess energy between W_{opt} and W_{max} is used to shift the entanglement towards the even subspace.

Note that the single-mode marginals of the superselected fermionic modes after step I of the protocol are fully determined by the corresponding average particle numbers. In principle, one may therefore consider the first step to involve the preparation of more general, uncorrelated states, for which $1/2 < N^{\mathrm{I}}_{S_1(S_2)} \leq 1$. However, we find that optimality is achieved for particles numbers that are compatible with thermal marginals.

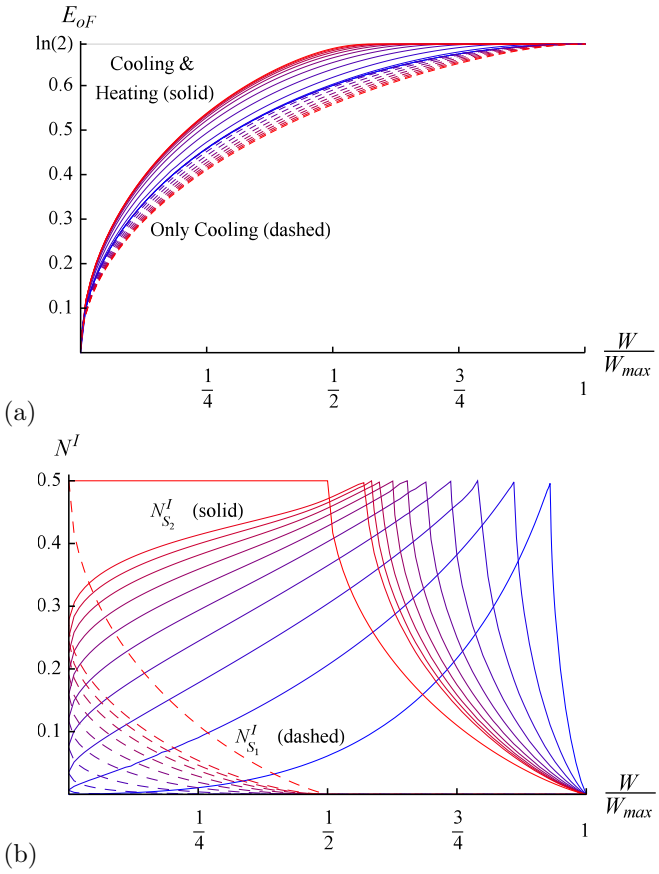

FIG. 5. **Optimal fermionic entanglement**: The solid curves in Fig. 5 (a) show the amount of entanglement (of formation) that can maximally be generated between two fermionic modes, S_1 and S_2, that are initially in a thermal state of temperature T, for a given energy cost W. The horizontal axis shows the relative energy cost, *i.e.*, the fraction of W and the energy cost W_{max}. Fig. 5 (b) shows the average particle numbers $N^{\mathrm{I}}_{S_1}$ (dashed lines) and $N^{\mathrm{I}}_{S_2}$ (solid lines) after the first step of the protocol, where we have assumed $N^{\mathrm{I}}_{S_2} \geq N^{\mathrm{I}}_{S_1}$ without loss of generality. The curves in both Fig. 5 (a) and (b) are plotted for initial temperatures varying from $T = 0$ to $T = 1$ in steps of 0.1 and for the limit $T \to \infty$ (bottom to top) in units $\hbar\omega/k_{\mathrm{B}}$. The dashed curves in Fig. 5 (a) show the corresponding curves of Fig. 3 (a) for temperatures varying from $T = 0$ to $T = 1$ in steps of 0.1 (top to bottom) as a comparison.

F: Optimality of Gaussian operations

Finally, we investigate the optimality of Gaussian operations for the generation of entanglement. As for the mutual information, we identify two energy regimes with qualitatively different behavior. In a certain low-energy regime, we are able to show that Gaussian operations are not optimal. To achieve this, we construct a protocol using specific non-Gaussian unitaries, which outperforms our previously established protocol for Gaussian operations. Nevertheless, in the high-energy regime, Gaussian

operations perform better. Indeed, we show that the entanglement generated by the Gaussian protocol scales optimally with the available energy in this case.

Low-energy regime

Instead of the previously established protocol based on Gaussian operations, we now introduce a scheme to generate entanglement using non-Gaussian operations in the correlation step. That is, after cooling the system to the temperature $T_{\mathrm{I}} = 1/\beta_{\mathrm{I}}$ using the energy W_{I}, we perform a unitary transformation that rotates in the subspace of the two-mode Fock space that is spanned by $|0_{S_1}\rangle\,|0_{S_1}\rangle$ and $|n_{S_1}\rangle\,|n_{S_2}\rangle$, where we recall the notation of Appendix D. One may think of this operation as generating Bell states in the four-dimensional subspace. We conveniently parametrize this rotation by a single, real parameter α, where $0 \leq \alpha \leq \pi/4$, such that

$$|0_{S_1}\rangle|0_{S_2}\rangle \mapsto \cos(\alpha)\,|0_{S_1}\rangle|0_{S_2}\rangle + \sin(\alpha)\,|n_{S_1}\rangle|n_{S_2}\rangle\,, \tag{F1}$$

$$|n_{S_1}\rangle|n_{S_2}\rangle \mapsto \cos(\alpha)\,|n_{S_1}\rangle|n_{S_2}\rangle - \sin(\alpha)\,|0_{S_1}\rangle|0_{S_2}\rangle\,. \tag{F2}$$

The energy cost W_{II} of this rotation is given by

$$W_{\mathrm{II}} = 2n\omega\big(p_0^2 - p_n^2\big)\sin^2(\alpha)\,, \tag{F3}$$

where we now have $p_n = (1-e^{-\beta_{\mathrm{I}}})e^{-n\beta_{\mathrm{I}}}$, with $\beta_{\mathrm{I}} = 1/T_{\mathrm{I}}$, and temperatures in units of ω. Here, the entanglement of formation of the transformed state can be quantified by way of the concurrence[46] of the (unnormalized) state of the subspace spanned by $|0_{S_1}\rangle\,|0_{S_1}\rangle$, $|0_{S_1}\rangle\,|n_{S_1}\rangle$, $|n_{S_1}\rangle\,|0_{S_2}\rangle$, and $|n_{S_1}\rangle\,|n_{S_2}\rangle$, see Refs. [47, 48]. For the concurrence $\mathcal{C}$, we obtain the expression

$$\begin{aligned}\mathcal{C} &= \big(p_0^2 - p_n^2\big)\sin(2\alpha) \;-\; 2p_0p_n \\ &= \sqrt{\tfrac{1}{n}\tfrac{W_{\mathrm{II}}}{\omega}}\sqrt{2\big(p_0^2 - p_n^2\big) - \tfrac{1}{n}\tfrac{W_{\mathrm{II}}}{\omega}} - 2p_0p_n\,.\end{aligned} \tag{F4}$$

Whenever $\mathcal{C} > 0$, entanglement is present, which translates to the condition

$$\frac{W_{\mathrm{II}}}{\omega}\Big(p_0^2 - p_n^2 - \frac{1}{2n}\frac{W_{\mathrm{II}}}{\omega}\Big) > 2np_0^2p_n^2\,. \tag{F5}$$

It can easily be seen that this condition can always be satisfied by choosing n to be large enough. Therefore, *some* entanglement can be generated at an arbitrarily low energy cost given two infinite-dimensional systems. Recall that Gaussian operations require at least the energy $\frac{\omega}{2}(\nu^{\mathrm{I}} - 1)^2$ to leave the separable set. Consequently, Gaussian operations cannot be optimal for entanglement generation in all regimes, although they are optimal for the generation of total correlations. Specifically, the unitary of Eq. (11) can be implemented with Gaussian operations. On the other hand, the amount of entanglement generated by the non-Gaussian protocol we have presented here is bounded. For fixed n, the maximal amount of energy useful for this protocol is $n\big(p_0^2 - p_n^2\big)$, and the corresponding maximal concurrence is given by

$$\mathcal{C}_{\max} = \big(p_0 - p_n\big)^2\,. \tag{F6}$$

In contrast, the entanglement that may be generated by Gaussian operations is unbounded. Our considerations are illustrated in Fig. 6.

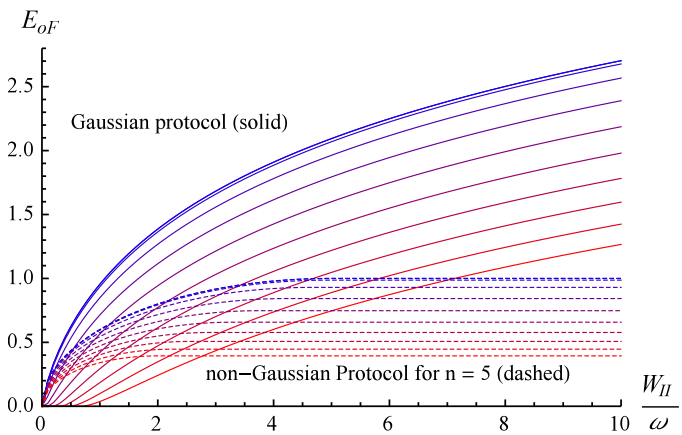

FIG. 6. **Comparison of Gaussian and non-Gaussian operations**: The plot shows the amount of entanglement (of formation) that can maximally be generated between two bosonic modes in step II of the protocol. Both modes are assumed to have been cooled to temperature T_{I} in the first step. Using the energy W_{II}, the solid curves show the optimal entanglement generated by Gaussian operations, while the dashed curves show the amount of entanglement generated by the non-Gaussian protocol. In both cases, the curves are plotted for temperatures varying from $T_{\mathrm{I}} = 0$ to $T_{\mathrm{I}} = 1$ in steps of 0.1 (top to bottom) in units $\hbar\omega/k_{\mathrm{B}}$.

High-energy regime

To study the regime of large energies, we first show that Gaussian operations are optimal to generate entanglement from the ground state. If the state is pure, the entanglement of formation is simply given by the entropy of the local state. For a given amount of work, the unitary maximizing E_{oF} will then be precisely the expression of Eq. (11), as the thermal state maximizes the entropy for a given energy. Given two bosonic modes, this operation can be implemented by a two-mode squeezing operation. In the protocols that we have considered, the first step consists of cooling. Whenever the ground state is reached, the Gaussian correlating operation is optimal. This occurs when $W_{\mathrm{I}} > -F\big(\tau_S(\beta)\big)$, and we conclude that the protocol is certainly optimal when $W \gg -F\big(\tau_S(\beta)\big)$.

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II.2 Energetics of Correlations in Interacting Systems

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Overview: In this paper, we have investigated the creation of correlations (in terms of the mutual information) in systems with symmetric local Hamiltonians that are coupled via an interaction term. The presence of these interactions leads to initial correlations in the thermal state of the system, which not only contribute to the overall correlations created, but, as we show, can also reduce the work cost of additionally created correlations.

Contribution: I developed the idea for this paper with my coauthors, performed the calculations and derivations, and was responsible for writing and composition of the (first draft of the) manuscript.

Energetics of correlations in interacting systems

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A fundamental connection between thermodynamics and information theory arises from the fact that correlations exhibit an inherent work value. For noninteracting systems this translates to a work cost for establishing correlations. Here we investigate the relationship between work and correlations in the presence of interactions that cannot be controlled or removed. For such naturally coupled systems, which are correlated even in thermal equilibrium, we determine general strategies that can reduce the work cost of correlations, and illustrate these for a selection of exemplary physical systems.

I. INTRODUCTION

Quantum information (QI) and quantum thermodynamics (QT) can both be framed as resource theories [1]. Based on the fundamental laws of quantum physics, these theories describe the (minimal) resources needed to perform certain tasks of interest. In order to identify the relevant resources, one first determines which states and operations are freely available, taking into account practical limitations on physical operations. Within QT, thermal systems and energy preserving operations are considered to be "for free", whereas systems out of equilibrium and operations that require external energy constitute resources [2, 3]. In QI, on the other hand, the paradigmatic task is efficient communication. In this context one assumes local operations and classical communication (LOCC) to be for free, whereas entangled quantum systems are resources that enable tasks beyond the restrictions of LOCC [4, 5].

Both of these resource theories can be considered to be simplifications of a more general physical framework: In either case only the restrictions of one area are taken into account. However, especially in quantum systems limitations from both thermodynamics and information theory present themselves simultaneously, which has greatly stimulated investigations of the connection between QI and QT (see Refs. [6–8] for recent reviews). For instance, locality restrictions on the allowed operations can limit the efficiency of thermodynamic processes [9–14], while correlations can enhance the performance of thermodynamic tasks [10, 15–21], and may even change the natural direction of the heat flow [22, 23]. Conversely, a nonzero ambient temperature induces a nonzero entropy, which limits the capacity for establishing (quantum) correlations [24, 25]. To overcome the constraints of LOCC, QI tasks hence require a supply of thermodynamic resources in the form of free energy.

Here, we aim to study the exchange between energy and correlations in the particularly transparent setting considered in Refs. [24, 25]: Given a collection of uncorrelated thermal states at the same temperature T, one is interested in determining the minimal energy W that is needed to create (quantum) correlations. For a bipartite system with access to an auxiliary thermal bath at temperature T, one finds the relation [15, 25–27]

$$W \geq T\Delta\mathcal{I}_{S_1S_2}\,, \tag{1}$$

where $\Delta\mathcal{I}_{S_1S_2}$ is the gain of correlations between the subsystems S_1 and S_2 as measured by the mutual information, and we note that we work in units such that $\hbar = k_{\mathrm{B}} = 1$ throughout this paper. The expression in Eq. (1) represents a fundamental bound on the exchange between energy and correlations, if the subsystems S_1 and S_2 are not interacting, that is, if the systems Hamiltonian is of the form $H_S = H_{S_1} + H_{S_2}$. In this work we relax this assumption, and explore how the relation in Eq. (1) is modified for interacting systems.

An important difference to previous results lies in the fact that thermal states of interacting Hamiltonians are generally already correlated, and may potentially even be entangled [28]. This naturally raises the question of whether the presence of interactions provides advantages for the generation of (additional) correlations. We answer this question affirmatively, by constructing explicit strategies to achieve $W < T\Delta\mathcal{I}_{S_1S_2}$ for some energy range in any finite-dimensional system with arbitrary interacting Hamiltonian. While these procedures can improve on the best protocols for non-interacting systems, they are not necessarily optimal in the sense that other protocols may exist that generate more correlations at the same energy cost. To complement this approach we therefore develop optimal strategies for two physically relevant cases: two interacting, fermionic or bosonic modes.

This paper is structured as follows. We first provide a short summary of the framework for this investigation in Sec. II. We then approach the problem of generating correlations in interacting systems in Sec. III, where we develop general strategies to use the energy

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contained in the interactions to improve upon the bound in Eq. (1), and explicitly demonstrate their applicability in a system of two qubits. In Sec. IV, we then turn to another finite-dimensional example, two interacting, fermionic modes. This system, restricted by superselection rules, is amenable to a numerical approach that we use to determine the optimal conversion of energy into correlations. Finally, in Sec. V, we study the generation of correlations in the infinite-dimensional system of two interacting, bosonic modes.

II. FRAMEWORK

We consider a bipartite system S, made up of subsystems S_1 and S_2, initially at thermal equilibrium at ambient temperature $T = 1/\beta$, described by a thermal state

$$\tau(\beta) = \mathcal{Z}^{-1}(\beta)\, e^{-\beta H_S}\,, \tag{2}$$

where $\mathcal{Z}(\beta) = \mathrm{Tr}(e^{-\beta H_S})$ is the partition function, and H_S is the system Hamiltonian. We further assume the presence of an auxiliary heat bath B, that is, an arbitrarily large system in thermal equilibrium with S. The total Hamiltonian is $H = H_S + H_B$, and the initial state can be written as $\tau_{SB}(\beta) = \tau(\beta) \otimes \tau_B(\beta)$. In order to transform this equilibrium state, we consider arbitrary unitary operations U_{SB} on SB. Since the joint systems SB is closed, these unitaries correspond to the most general operations permissable in this situation. The average work cost of transforming the state of S from $\tau(\beta)$ to a final state $\rho = \mathrm{Tr}_B(U_{SB}\tau_{SB}(\beta)U_{SB}^\dagger)$ is given by,

$$W = \mathrm{Tr}\Big(H\big[U_{SB}\tau_{SB}(\beta)U_{SB}^\dagger - \tau_{SB}(\beta)\big]\Big), \tag{3}$$

which corresponds to the total external energy input (see, e.g., Refs. [29, 30]). In Ref. [27] (see also Refs. [31–33] for the same result in related frameworks), it is shown that W can be bounded by the (nonequilibrium) free energy difference,

$$W(\tau \to \rho) \geq \Delta F_S = F(\rho) - F(\tau), \tag{4}$$

where the free energy with respect to the reservoir at temperature T is

$$F(\rho) = E(\rho) - TS(\rho)\,. \tag{5}$$

Here, $E(\rho) = \mathrm{Tr}\big(H_S\rho\big)$ is the average energy, and $S(\rho) = -\mathrm{Tr}\big(\rho\ln(\rho)\big)$ is the von Neumann entropy. Note that $F(\rho)$ depends only on the state of S and the temperature of B. Equality in (4) can be obtained in a quasistatic process [31–33], in which case the work cost becomes minimal.

We now wish to invest some work W to increase the correlations within the bipartite state of S as much as possible. (Note that B is only an auxiliary system and we do not wish to create correlations between S and B.) In Refs. [24, 25] this problem was considered for noninteracting Hamiltonians, $H_S = H_{S_1} + H_{S_2}$. Here, we want to depart from this paradigm and consider an interacting Hamiltonian of the form

$$H_S = H_{S_1} + H_{S_2} + H_I\,. \tag{6}$$

As discussed above, the work cost of transforming $\tau(\beta)$ to a final state ρ satisfies, $W(\tau \to \rho) \geq \Delta F_S = F(\rho) - F(\tau)$, and equality can be achieved in a quasi-static process and with a sufficiently large bath [31–33]. The task is then to maximize the correlations of ρ under the constraint $F(\rho) - F(\tau) \leq W$, where W is the amount of available work.

Before continuing, note that the main ingredients of the investigations in Refs. [24, 25] are preserved:

(i) The initial state is in thermal equilibrium, and therefore, the energy cost of transforming $\tau(\beta)$ to any final state ρ is nonnegative, $W \geq 0$.

(ii) We assume arbitrary (in particular, unitary) operations can be performed on S and the auxiliary thermal bath, which allows obtaining fundamental bounds on the work cost of correlations.

We quantify the amount of correlations between the subsystems by the mutual information,

$$\mathcal{I}_{S_1S_2}(\rho) = S(\rho_{S_1}) + S(\rho_{S_2}) - S(\rho)\,, \tag{7}$$

where $\rho_{S_1(S_2)} = \mathrm{Tr}_{S_2(S_1)}(\rho)$ are the reduced states of the subsystems. The main quantity of interest throughout this paper will be the correlations gain,

$$\Delta\mathcal{I}_{S_1S_2} = \mathcal{I}_{S_1S_2}(\rho) - \mathcal{I}_{S_1S_2}(\tau). \tag{8}$$

That is, we take a point of view inspired by Landauer's principle, and ask how many units of correlations $\Delta\mathcal{I}_{S_1S_2}$ can be newly generated (on top of the preexisting correlations) at the expense of one unit of energy. Note that, in the noninteracting case, $\Delta\mathcal{I}_{S_1S_2}$ and $\mathcal{I}_{S_1S_2}$ coincide, as the initial thermal state of the Hamiltonian $H_{S_1} + H_{S_2}$ is an uncorrelated product state. In the interacting case, $\Delta\mathcal{I}_{S_1S_2}$ and $\mathcal{I}_{S_1S_2}$ still arise from the same optimization procedure, but $\mathcal{I}_{S_1S_2} \geq \Delta\mathcal{I}_{S_1S_2}$. Since $\Delta\mathcal{I}_{S_1S_2}$ quantifies the amount of correlations generated through the investment of W, we first focus on $\Delta\mathcal{I}_{S_1S_2}$, establishing strategies to achieve $W < T\Delta\mathcal{I}_{S_1S_2}$ in Sec. III.

III. GENERAL CONSIDERATIONS

A. Work cost of generating correlations

Let us now relate the correlation gain $\Delta\mathcal{I}_{S_1S_2}$ to the minimal work cost ΔF_S. Inserting the Hamiltonian from

Eq. (6) into (4) one obtains

$$\Delta F_S = \mathrm{Tr}\big(H_{S_1}[\rho-\tau] + H_{S_2}[\rho-\tau]\big) + \mathrm{Tr}\big(H_I[\rho-\tau]\big) + T\left[S(\tau) - S(\rho)\right]. \tag{9}$$

On the other hand, for the difference in mutual information from Eq. (8) one finds $T\,\Delta\mathcal{I}_{S_1S_2} = T\left[S(\tau) - S(\rho)\right] + T\left[S(\rho_{S_1}) - S(\tau_{S_1})\right] + T\left[S(\rho_{S_2}) - S(\tau_{S_2})\right]$. After some straightforward manipulations we then arrive at

$$\Delta F_S = T\Delta\mathcal{I}_{S_1S_2} + \mathrm{Tr}\big(H_I[\rho-\tau]\big) + \Delta\tilde{F}_{S_1} + \Delta\tilde{F}_{S_2}\,, \tag{10}$$

where the quantities $\tilde{F}_{S_1(S_2)}$ correspond to nonequilibrium free energies with respect to the local Hamiltonians, i.e.,

$$\tilde{F}_{S_i}(\rho) = \mathrm{Tr}\big(H_{S_i}\rho\big) - TS(\rho_{S_i}) \tag{11}$$

with $\rho_{S_1(S_2)} = \mathrm{Tr}_{S_2(S_1)}\rho$. Here, it is important to note that the marginals $\tau_{S_1(S_2)} = \mathrm{Tr}_{S_2(S_1)}\tau$ of the initial thermal state are not themselves thermal states with respect to the local Hamiltonians, $\tau_{S_1(S_2)} \neq \mathcal{Z}_{S_i}^{-1}\exp(-\beta H_{S_i})$. It can therefore be inferred that, while $\Delta F_S \geq 0$ since the initial state τ is at thermal equilibrium with the bath, the quantities $\Delta\tilde{F}_{S_i}$ may have either sign. Only when H_I vanishes are the marginals of τ also thermal states with minimal free energy and $\sum_i \Delta\tilde{F}_{S_i}$ is nonnegative. In this case one obtains (see Ref. [25] for a detailed derivation) the bound

$$T\Delta\mathcal{I}_{S_1S_2}(\rho) \leq \Delta F_S \leq W, \quad \text{if } H_I = 0, \tag{12}$$

which shows that, for noninteracting systems, at least $T\Delta\mathcal{I}_{S_1S_2}$ units of work have to be invested to increase the correlations of the systems by the amount $\Delta\mathcal{I}_{S_1S_2}$.

B. Strategies to utilize the interactions

In this section we determine strategies that can potentially outperform the bound of Eq. (12) by making use of the energy provided by the interactions. We formulate these strategies for arbitrary Hamiltonians of bipartite systems, whose subsystems S_1 and S_2 can be arbitrary finite-dimensional quantum systems, qudits, with Hilbert spaces $\mathcal{H}_{S_1}$ and $\mathcal{H}_{S_2}$, and dimensions d_1 and d_2, respectively. The density operators for such systems can be written in a generalized Bloch-Fano decomposition [34, 35], that is

$$\rho = \frac{1}{d_1d_2}\Big(\mathbb{1}_S + \sum_{m=1}^{d_1^2-1} a_m\,\sigma_m^{S_1}\otimes\mathbb{1}_{S_2} + \sum_{n=1}^{d_2^2-1} b_n\mathbb{1}_{S_1}\otimes\sigma_n^{S_2} + \sum_{m=1}^{d_1^2-1}\sum_{n=1}^{d_2^2-1} c_{mn}\,\sigma_m^{S_1}\otimes\sigma_n^{S_2}\Big), \tag{13}$$

where the Hermitean operators $\sigma_m^{S_i}$ satisfy $\mathrm{Tr}(\sigma_m^{S_i}\sigma_n^{S_i}) = 2\delta_{mn}$ and $\mathrm{Tr}(\sigma_m^{S_i}) = 0$, and the real coefficients a_m, b_n, and t_{mn} are subject to constraints arising from the positivity of ρ. The reduced states are then immediately obtained as

$$\rho_{S_1} = \frac{1}{d_1}\Big(\mathbb{1}_{S_1} + \sum_{m=1}^{d_1^2-1} a_m\,\sigma_m^{S_1}\Big), \tag{14a}$$

$$\rho_{S_2} = \frac{1}{d_2}\Big(\mathbb{1}_{S_2} + \sum_{n=1}^{d_2^2-1} b_n\,\sigma_n^{S_2}\Big). \tag{14b}$$

The Hermitean interaction Hamiltonian can similarly be written as

$$H_I = \sum_{m=1}^{d_1^2-1}\sum_{n=1}^{d_2^2-1} \epsilon_{mn}\,\sigma_m^{S_1}\otimes\sigma_n^{S_2}\,, \tag{15}$$

with real coefficients ϵ_{mn}. Any terms of the form $\mathbb{1}_{S_1}\otimes\sigma_m^{S_2}$ and $\sigma_m^{S_1}\otimes\mathbb{1}_{S_2}$ that may appear in such a decomposition of H_I can be absorbed into the local Hamiltonians H_{S_i}.

Returning to the relation of Eq. (10), notice that the interactions allow surpassing the bound in Eq. (12) whenever $\mathrm{Tr}\big(H_I[\rho-\tau]\big) + \sum_i \Delta\tilde{F}_{S_i}$ is negative. The expansion of Eq. (13) further permits treating each of these terms independently: The terms $\Delta\tilde{F}_{S_i}$ depend only on the local Bloch vector components a_m and b_n, for $i=1$ and $i=2$, respectively, whereas the interaction term $\mathrm{Tr}\big(H_I[\rho-\tau]\big)$ depends only on the correlation tensor c_{mn}. With this we can formulate two complementary strategies. However, it is important to keep in mind that any choice of the coefficients c_{mn}, a_m and b_n is subject to the positivity constraint $\rho \geq 0$.

First, we focus on the local terms $\Delta\tilde{F}_{S_i}$. Defining the local Gibbs states as $\gamma_{S_i} \equiv \mathcal{Z}_{S_i}^{-1}e^{-\beta H_{S_i}}$, which are generically different from the local initial states $\tau_{S_1(S_2)} = \mathrm{Tr}_{S_2(S_1)}\tau$, it is useful to rewrite $\Delta\tilde{F}_{S_i}$ as,

$$\beta\Delta\tilde{F}_{S_i} = \beta\big(F(\rho_{S_i}) - F(\gamma_{S_i})\big) - \beta\big(F(\tau_{S_i}) - F(\gamma_{S_i})\big) = S(\rho_{S_i}\|\gamma_{S_i}) - S(\tau_{S_i}\|\gamma_{S_i})\,, \tag{16}$$

where $S(\rho\|\tau) = -S(\rho) - \mathrm{Tr}(\rho\ln\tau)$ is the relative entropy. Here, we have used that $\beta\big[F(\rho) - F\big(\tau(\beta)\big)\big] = S(\rho\|\tau(\beta))$, which can easily be shown using Eqs. (2) and (5). Since $S(\,.\,\|\,.\,)$ is a measure of distance between two quantum states, the quantities $\Delta\tilde{F}_{S_i}$ are negative whenever the final reduced states ρ_{S_i} are closer to the local Gibbs states γ_{S_i} than the initial state marginals τ_{S_i}. This provides a simple strategy to minimize $\Delta\tilde{F}_{S_i}$: The Bloch coefficients $a_m^{(\rho)}$ and $b_n^{(\rho)}$ of the final state ρ should to be chosen as close as possible to $a_m^{(\gamma)}$ and $b_m^{(\gamma)}$, respectively, where $a_m^{(\gamma)} = \frac{d_1}{2}\mathrm{Tr}(\gamma_{S_1}\sigma_m^{S_1})$ and $b_n^{(\gamma)} = \frac{d_2}{2}\mathrm{Tr}(\gamma_{S_2}\sigma_n^{S_2})$. This strategy ensures that $\Delta\tilde{F}_{S_i} < 0$.

The second strategy entails the minimization of the term $\mathrm{Tr}\big(H_I[\rho-\tau]\big)$. Using Eqs. (13) and (15), we can express it in terms of the correlation tensors $c_{mn}^{(\rho)}$ and

$c_{mn}^{(\tau)}$ of ρ and τ, respectively, obtaining

$$\mathrm{Tr}\big(H_I[\rho-\tau]\big) = \sum_{m=1}^{d_1^2-1}\sum_{n=1}^{d_2^2-1}\big(c_{mn}^{(\rho)} - c_{mn}^{(\tau)}\big)\epsilon_{mn}\,. \tag{17}$$

This relation has a clear geometrical interpretation. Mapping $c_{mn}^{(\rho)}$, $c_{mn}^{(\tau)}$, and ϵ_{mn} to vectors $\mathbf{c}^{(\rho)}$, $\mathbf{c}^{(\tau)}$, and $\boldsymbol{\epsilon}$ in a Euclidean vector space of dimension $(d_1^2-1)(d_2^2-1)$, the condition of Eq. (17) becomes

$$\mathrm{Tr}\big(H_I[\rho-\tau]\big) = (\mathbf{c}^{(\rho)} - \mathbf{c}^{(\tau)})\cdot\boldsymbol{\epsilon}\,. \tag{18}$$

To minimize the expression in (18) it is hence desirable to select the vector $(\mathbf{c}^{(\rho)} - \mathbf{c}^{(\tau)})$ to be as antiparallel as possible to $\boldsymbol{\epsilon}$.

The considerations discussed in this section hence provide two complementary strategies to obtain $\beta W < \Delta\mathcal{I}_{S_1S_2}$, as desired. In general, the choices of $a_m^{(\rho)}$, $b_m^{(\rho)}$, and $c_{mn}^{(\rho)}$ are limited by the positivity constraint, $\rho \geq 0$ (and of course also by the amount of available work, W). In the next section we illustrate possible issues with the positivity of ρ in more detail for a particular example of two interacting qubits.

C. Improved generation of correlations for two qubits

We consider a system of two qubits, coupled by the Hamiltonian

$$H_S = \omega\left(\sigma_z^{S_1} + \sigma_z^{S_2}\right) + \epsilon\,\sigma_z^{S_1}\otimes\sigma_z^{S_2}\,, \tag{19}$$

where $\omega \geq 0$ and $\epsilon \in \mathbb{R}$ can take either sign. In this simple example, the presence of the interaction Hamiltonian $H_I = \epsilon\,\sigma_z^{S_1}\otimes\sigma_z^{S_2}$ does not change the eigenstates of H_S, but the eigenvalues of the noninteracting system are modified to $(\epsilon \pm 2\omega)$ and $-\epsilon$ (twice degenerate). The initial thermal state $\tau(\beta) = e^{-\beta H_S}/\mathcal{Z}$ is hence of the form

$$\tau(\beta) = \mathcal{Z}^{-1}\,\mathrm{diag}\{e^{-\beta(\epsilon+2\omega)}, e^{\beta\epsilon}, e^{\beta\epsilon}, e^{-\beta(\epsilon-2\omega)}\} \tag{20}$$

with $\mathcal{Z} = \mathrm{Tr}\big(e^{-\beta H_S}\big) \geq 0$. The nonzero coefficients of the Bloch decomposition of $\tau(\beta)$ are

$$a_z^{(\tau)} = b_z^{(\tau)} = -2\mathcal{Z}^{-1}e^{-\beta\epsilon}\sinh(2\beta\omega) < 0\,, \tag{21a}$$

$$c_{zz}^{(\tau)} = 1 - \frac{4e^{\beta\epsilon}}{\mathcal{Z}}\,. \tag{21b}$$

To correlate the system, we apply a two-step protocol based on the strategies discussed in Sec. III B. In the first phase of the protocol, step I, we aim to minimize the term $\mathrm{Tr}\big(H_I[\rho-\tau]\big)$. To do this, we transform the state τ to ρ_{I}, such that the local Bloch vector components remain invariant, $a_z^{(\rho_{\mathrm{I}})} = b_z^{(\rho_{\mathrm{I}})} = a_z^{(\tau)}$, while the (nonzero) correlation tensor coefficient is mapped to

$$c_{zz}^{(\rho_{\mathrm{I}})} = c_{zz}^{(\tau)} - \mathrm{sgn}(\epsilon)\,\alpha_{\mathrm{I}}\,, \tag{22}$$

for $\alpha_{\mathrm{I}} \geq 0$. With this, one finds $\mathrm{Tr}\big(H_I[\rho_{\mathrm{I}}-\tau]\big) = -|\epsilon|\alpha_{\mathrm{I}}$ and from Eq. (10) we obtain

$$W_{\mathrm{I}} = T\,\Delta\mathcal{I}_{S_1S_2} - |\epsilon|\alpha_{\mathrm{I}}\,, \tag{23}$$

where we assumed that the process is quasistatic, so that $W = \Delta F_S$. (Note that the same assumption is made also later in step II.) The correlations are hence generated at a work cost that is lower than in the noninteracting case, $W_{\mathrm{I}} \leq T\,\Delta\mathcal{I}_{S_1S_2}$. However, it is crucial to note that the transformation in Eq. (22) is limited by the positivity constraint, $\rho_I \geq 0$, requiring $2|a_z^{(\tau)}| - 1 \leq c_{zz}^{(\rho_{\mathrm{I}})} \leq 1$. Depending on the sign of the interaction term, one of these bounds is reached, when enough energy is supplied. That is, $c_{zz}^{(\rho_{\mathrm{I}})}$ eventually tends towards either $c_{zz}^{(\rho_{\mathrm{I}})} = 2|a_z^{(\tau)}| - 1$ or $c_{zz}^{(\rho_{\mathrm{I}})} = 1$ for $\epsilon > 0$ or $\epsilon < 0$, respectively.

If more energy is available than is needed to saturate the positivity constrain in step I, we may employ the complementary strategy discussed in Sec. III B in step II, the second phase of the protocol. Now, we keep the correlation tensor fixed, while changing the local Bloch vector components to minimize $\Delta\tilde{F}_{S_i}$. This entails moving the marginals closer to the states γ_{S_i} that are locally thermal with respect to H_{S_i}. These local Gibbs states are here given by

$$\gamma_{S_i} = \frac{e^{-\beta H_{S_i}}}{\mathcal{Z}_{S_i}} = \tfrac{1}{2}\big(\mathbb{1}_2 - \tanh(\beta\omega)\sigma_z^{S_i}\big)\,, \tag{24}$$

with $a_z^{(\gamma)} = -\tanh(\beta\omega) < 0$. We hence map ρ_{I} to the state ρ_{II} with Bloch vector components given by

$$a_z^{(\rho_{\mathrm{II}})} = (1-\alpha_{\mathrm{II}})\,a_z^{(\tau)} + \alpha_{\mathrm{II}}\,a_z^{(\gamma)}\,, \tag{25}$$

where $0 \leq \alpha_{\mathrm{II}} \leq 1$. Again, the positivity constraint $\rho_{\mathrm{II}} \geq 0$ must still be taken into account. For $\epsilon < 0$ we find that the full range of α_{II} is compatible with the positivity of ρ_{II}. The work cost of step II is given by $W_{\mathrm{II}} = T\Delta\mathcal{I}_{S_1S_2} + \Delta\tilde{F}_{S_1} + \Delta\tilde{F}_{S_2}$, and, as illustrated in Fig. 1, we indeed find that $\Delta\tilde{F}_{S_i} \leq 0$ for all values of $T \geq 0$, $0 \leq \alpha_{\mathrm{II}} \leq 1$, and $\epsilon < 0$.

For $\epsilon > 0$, on the other hand, the positivity constraints require that $|a_z^{(\rho_{\mathrm{II}})}| \leq |a - z^{(\tau)}|$. Since $a_z^{(\tau)} < 0$ and $a_z^{(\gamma)} = -\tanh(\beta\omega) < 0$, Eq. (25) yields $|a_z^{(\rho_{\mathrm{II}})}| = (1-\alpha_{\mathrm{II}})|a_z^{(\tau)}| + \alpha_{\mathrm{II}}\tanh(\beta\omega) \geq |a_z^{(\tau)}|$. Unfortunately, since $|a_z^{(\tau)}| = \sinh(2\beta\omega)/\big(\cosh(2\beta\omega)+e^{-\beta\epsilon}\big) \leq \tanh(\beta\omega)$, one finds that $|a_z^{(\rho_{\mathrm{II}})}| \geq |a - z^{(\tau)}|$, that is, the positivity constraint does not allow for step II of the protocol to be carried out for $\epsilon > 0$.

In addition to the strategies discussed here, the states obtained after steps I and II may be further correlated until the maximal value of correlation is reached. How-

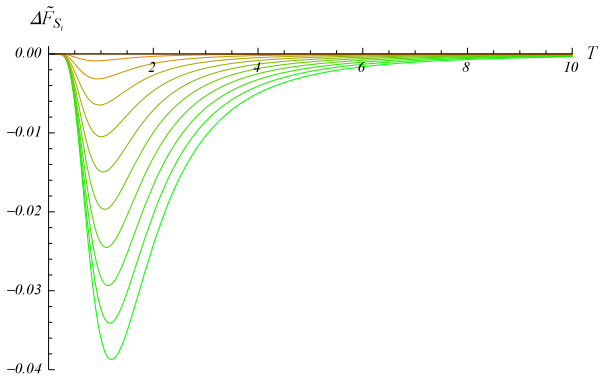

FIG. 1. **Advantage in correlation cost:** During step II of the protocol to generate correlations between two qubits, an advantage over the noninteracting case arises when $\Delta\tilde{F}_{S_i}$ from Eq. (11) becomes negative. $\Delta\tilde{F}_{S_i}$ is plotted here against the temperature T in units of ω (recall that we use units where $\hbar = k_{\mathrm{B}} = 1$) for $\alpha_{\mathrm{II}} = 0.5$, and the different curves correspond to values of ϵ (also in units of ω) from $\epsilon = 0$ (top) to $\epsilon = -1$ (bottom) in steps of 0.1. The advantage increases with increasing coupling strength ϵ, but does not monotonically decrease with the temperature. Instead, the advantage becomes maximal at a finite temperature. Although curves are only shown for a fixed value $\alpha_{\mathrm{II}} = 0.5$, we have checked that other values yield analogous behaviour and the advantage increases monotonically with α_{II}.

ever, the work cost per newly generated unit of correlation beyond this point may be the same, or even higher than in the noninteracting case. We shall shed light on this possibility in the next sections, by studying in detail protocols to correlate bipartite systems of fermions and bosons.

IV. TWO FERMIONIC MODES

In this section, we consider the two systems S_1 and S_2, which are to be correlated, to be two fermionic modes with ladder operators b_1, $b_1^\dagger$ and b_2, $b_2^\dagger$, respectively. Such fermionic systems have been well studied in the context of quantum information processing, see, e.g., Refs. [36–38], but we shall briefly review some key features. The annihilation and creation operators satisfy the usual anticommutation relations $\{b_m, b_n^\dagger\} = \delta_{mn}$ and $\{b_m, b_n\} = 0$. While the Pauli exclusion principle limits the dimension of the corresponding two-mode Fock space to 4, the anticommutation relations nonetheless imply a different subsystem structure as compared to a two-qubit Hilbert space [39]. Despite this inequivalence[1] of qubits and fermionic modes, the marginals and correlation measures for the fermionic system are well-defined when imposing a superselection rule that forbids superpositions of even and odd numbers of fermions [40, 41].

The fermionic system is hence of interest for the following reasons: First, the anticommutation relations and the restrictions of the superselection rule provide a qualitative difference to the qubit case, which makes for an interesting comparison. Second, the low-dimensional Hilbert space is amenable to a numerical treatment, allowing a rather general approach to optimal protocols for the generation of correlations. Third, fermionic fields form a conceptually fundamental ingredient in the current view of matter in the universe in terms of relativistic quantum field theory.

A. Hamiltonian and initial thermal state

Let us now turn to the specific system Hamiltonian that we consider in this section, $H = H_{S_1} + H_{S_2} + H_I$. For the noninteracting part, we consider the standard Hamiltonian for two modes of the same frequency ω, i.e.,

$$H_{S_1} + H_{S_2} = \omega \left(b_1^\dagger b_1 + b_2^\dagger b_2\right) . \tag{26}$$

For the interaction between the modes, we will employ the most general two-mode coupling term that is quadratic in the mode operators, given by

$$\begin{aligned} H_I &= H_{\mathrm{even}} + H_{\mathrm{odd}} \\ &= \epsilon_{\mathrm{even}}\left(b_1 b_2 + b_2^\dagger b_1^\dagger\right) + \epsilon_{\mathrm{odd}}\left(b_1^\dagger b_2 + b_2^\dagger b_1\right) , \end{aligned} \tag{27}$$

where H_{even} couples only the states $\| 0 \rangle\!\rangle$ and $\| 1_1 \rangle\!\rangle \| 1_2 \rangle\!\rangle = b_1^\dagger b_2^\dagger \| 0 \rangle\!\rangle$ in the even subspace, while H_{odd} acts in the odd subspace spanned by $\| 1_1 \rangle\!\rangle = b_1^\dagger \| 0 \rangle\!\rangle$ and $\| 1_2 \rangle\!\rangle = b_2^\dagger \| 0 \rangle\!\rangle$. Here $\| 0 \rangle\!\rangle$ is the vacuum state satisfying $b_i \| 0 \rangle\!\rangle = 0 \ \forall\ i$, and the double-lined ket notation indicates the antisymmetrized tensor product for the excited states, i.e., $\| 1_1 \rangle\!\rangle \| 1_2 \rangle\!\rangle = \| 1_1 \rangle\!\rangle \wedge \| 1_2 \rangle\!\rangle = - \| 1_2 \rangle\!\rangle \| 1_1 \rangle\!\rangle$ (see, e.g., Ref. [39] for more information).

The thermal state $\tau(\beta)$ of H can be computed straightforwardly. The eigenvalues of H read,

$$\lambda_{1,4} = \omega \pm \sqrt{\omega^2 + \epsilon_{\mathrm{even}}^2} , \tag{28a}$$

$$\lambda_{2,3} = \omega \pm \epsilon_{\mathrm{odd}} , \tag{28b}$$

where the labels 3, 4 refer to the negative relative sign,

[1] Note that n-mode fermionic Fock spaces are isomorphic to n-qubit Hilbert spaces via maps such as the Jordan-Wigner transformation. However, local operators in one space are generally mapped to nonlocal ones in the other. The marginals of an n-mode fermionic state ρ are hence generally not isomorphic to those of the n-qubit state $\tilde{\rho}$, even if $\tilde{\rho}$ is related to ρ via an isomorphism.

and the corresponding eigenstates are given by

$$\| \lambda_{1,4} \rangle\!\rangle = \frac{1}{\sqrt{\epsilon_{\text{even}}^2 + \lambda_{1,4}^2}} \Big(\epsilon_{\text{even}} \| 0 \rangle\!\rangle - \lambda_{1,4} \| 1_1 \rangle\!\rangle \| 1_2 \rangle\!\rangle \Big) , \tag{29a}$$

$$\| \lambda_{2,3} \rangle\!\rangle = \frac{1}{\sqrt{2}} \Big(\| 1_2 \rangle\!\rangle \pm \| 1_1 \rangle\!\rangle \Big) . \tag{29b}$$

Then, $\tau(\beta)$ can the be written as,

$$\tau(\beta) = \mathcal{Z}^{-1}(\beta) \sum_i e^{-\beta \lambda_i} \| \lambda_i \rangle\!\rangle \langle\!\langle \lambda_i \| , \tag{30}$$

where the partition function is $\mathcal{Z}(\beta) = \sum_i e^{-\beta \lambda_i}$. It is important to note that $\tau(\beta)$ already contains correlations, which are computed in detail in the Appendix.

B. Generation of Correlations

We now consider the task of correlating $\tau(\beta)$ further. The simple structure of the system (an only four-dimensional Hilbert space that is further restricted by superselection rules) allows us to consider the most general protocols beyond the strategies discussed in Sec. III B. That is, given some available work W, we consider the possibility to transform τ to any state ρ, provided $\Delta F_S(\tau \to \rho) \leq W$ is satisfied. In order to maximize the created correlations, $\Delta \mathcal{I}_{S_1 S_2}$, we conveniently parametrize the final state ρ, and numerically optimize its mutual information $\mathcal{I}_{S_1 S_2}(\rho)$ under the constraint of a maximally available free energy.

Since the final state needs to respect the superselection rule that forbids superpositions of even and odd numbers of fermions [40], the four-dimensional Fock space splits into two two-dimensional spaces. An arbitrary two-mode final state may therefore be written as a convex combination of two density operators, ρ_{even} and ρ_{odd}, corresponding to the subspaces of even and odd fermion numbers, respectively. We hence write

$$\rho = p\, \rho_{\text{even}} + (1-p)\, \rho_{\text{odd}} , \tag{31}$$

where $0 < p < 1$. For each of the two subspaces, we then use a single-qubit Bloch representation, i.e.,

$$\begin{aligned} \rho_{\text{even}} = \tfrac{1}{2} \Big(& \big[1 + z_{\text{even}}\big] \| 0 \rangle\!\rangle \langle\!\langle 0 \| \\ & + \big[1 - z_{\text{even}}\big] \| 1_1 \rangle\!\rangle \| 1_2 \rangle\!\rangle \langle\!\langle 1_2 \| \langle\!\langle 1_1 \| \\ & + \big[(x_{\text{even}} - i y_{\text{even}}) \| 0 \rangle\!\rangle \langle\!\langle 1_2 \| \langle\!\langle 1_1 \| + \text{H. c.} \big] \Big) , \end{aligned} \tag{32a}$$

$$\begin{aligned} \rho_{\text{odd}} = \tfrac{1}{2} \Big(& \big[1 + z_{\text{odd}}\big] \| 1_2 \rangle\!\rangle \langle\!\langle 1_2 \| + \big[1 - z_{\text{odd}}\big] \| 1_1 \rangle\!\rangle \langle\!\langle 1_1 \| \\ & + \big[(x_{\text{odd}} - i y_{\text{odd}}) \| 1_2 \rangle\!\rangle \langle\!\langle 1_1 \| + \text{H. c.} \big] \Big) , \end{aligned} \tag{32b}$$

where the coefficients satisfy $|x_{\text{even,odd}}| \leq 1$, $|y_{\text{even,odd}}| \leq 1$, $|z_{\text{even,odd}}| \leq 1$, and

$$r_{\text{even}}^2 = x_{\text{even}}^2 + y_{\text{even}}^2 + z_{\text{even}}^2 \leq 1 , \tag{33a}$$

$$r_{\text{odd}}^2 = x_{\text{odd}}^2 + y_{\text{odd}}^2 + z_{\text{odd}}^2 \leq 1 . \tag{33b}$$

In this parametrization, the entropy of the final state can easily be obtained via its eigenvalues $\frac{p}{2}(1 \pm r_{\text{even}})$ and $\frac{1-p}{2}(1 \pm r_{\text{odd}})$. The energy of the final state, in turn, is

$$E(\rho) = \omega(1 - p\, z_{\text{even}}) - p\, \epsilon_{\text{even}} x_{\text{even}} + (1-p) \epsilon_{\text{odd}} x_{\text{odd}} . \tag{34}$$

Lastly, the final state marginals are of the form

$$\begin{aligned} \rho_{S_1} = & \tfrac{1}{2} \big(1 + p\, z_{\text{even}} + (1-p) z_{\text{odd}}\big) \| 0 \rangle\!\rangle \langle\!\langle 0 \| \\ & + \tfrac{1}{2} \big(1 - p\, z_{\text{even}} - (1-p) z_{\text{odd}}\big) \| 1_1 \rangle\!\rangle \langle\!\langle 1_1 \| , \end{aligned} \tag{35a}$$

$$\begin{aligned} \rho_{S_2} = & \tfrac{1}{2} \big(1 + p\, z_{\text{even}} - (1-p) z_{\text{odd}}\big) \| 0 \rangle\!\rangle \langle\!\langle 0 \| \\ & + \tfrac{1}{2} \big(1 - p\, z_{\text{even}} + (1-p) z_{\text{odd}}\big) \| 1_2 \rangle\!\rangle \langle\!\langle 1_2 \| . \end{aligned} \tag{35b}$$

For the illustration of the results, it is convenient to specify the amount of available input energy in units of W_{min}, the minimal free energy difference[2] to a maximally correlated state. Taking into account that such a maximally correlated state must be pure, and that the free energy of the initial state is $F(\tau) = -T \ln(\mathcal{Z})$, we find

$$W_{\text{min}} = \omega - \max\{|\epsilon_{\text{even}}|, |\epsilon_{\text{odd}}|\} + T \ln(\mathcal{Z}) . \tag{36}$$

With this, we may numerically evaluate the maximal amount of correlations that can be created at a fixed temperature T and a fixed energy input W/W_{min}. The results of the optimization allows us to compare the energy cost of optimal protocols to generate correlations, for both interacting ($\epsilon_{\text{even,odd}} \neq 0$) and noninteracting systems ($\epsilon_{\text{even,odd}} = 0$). The results are shown in Fig. 2.

In agreement with our considerations in Sec. III B, one observes an initial regime where the interactions provide an advantage. However, at some point, the energy cost of $\Delta \mathcal{I}_{S_1 S_2}$ becomes higher for interacting systems than for noninteracting ones. This is to be expected: Since the interacting system is correlated initially, the maximal value of $\Delta \mathcal{I}_{S_1 S_2}$ is always lower than in the noninteracting case.

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[2] Note that the minimal energy for maximal correlations (W_{min}) depends on the coupling strength. Therefore, the functions $\Delta \mathcal{I}_{\epsilon \neq 0}$ and $\Delta \mathcal{I}_{\epsilon = 0}$, whose difference is plotted in Fig. 2, would be multiplied by different values when converting the plots of Fig. 2 to absolute energy costs. This would result in shifted intersections with the horizontal axes. Nonetheless, since W_{min} is maximal in the absence of interactions, the intersections would all shift to the left, leaving the conclusion unchanged, that the presence of interactions may make the creation of new correlations more expensive, even if the overall amount of correlations is larger in the end.

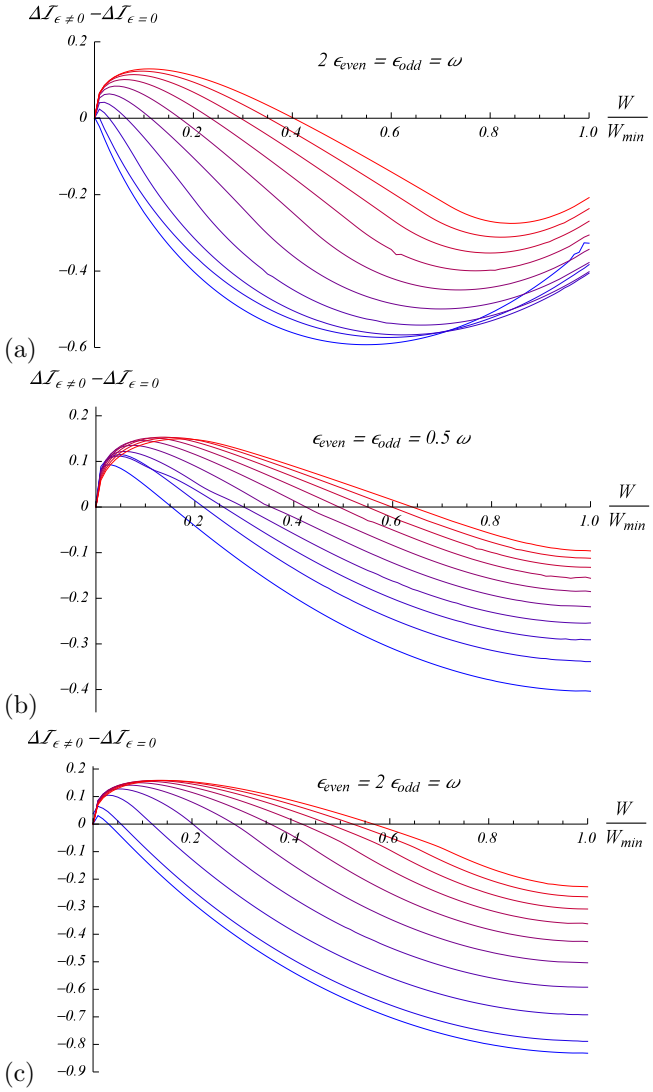

FIG. 2. **Fermionic newly generated correlation cost:** The difference in correlations that can be newly generated for an available energy W (in units of $W_{\min}$) in the presence ($\Delta\mathcal{I}_{\epsilon\neq 0}$) and absence ($\Delta\mathcal{I}_{\epsilon=0}$) of interactions is shown for temperatures $T = 0.1, \ldots, 1$ (in units of $\hbar\omega/k_B$) in steps of 0.1 (blue to red, top to bottom) for the ratios $\epsilon_{\text{even}}/\epsilon_{\text{odd}} = 2, 1$, and 0.5 in (a),(b), and (c), respectively.

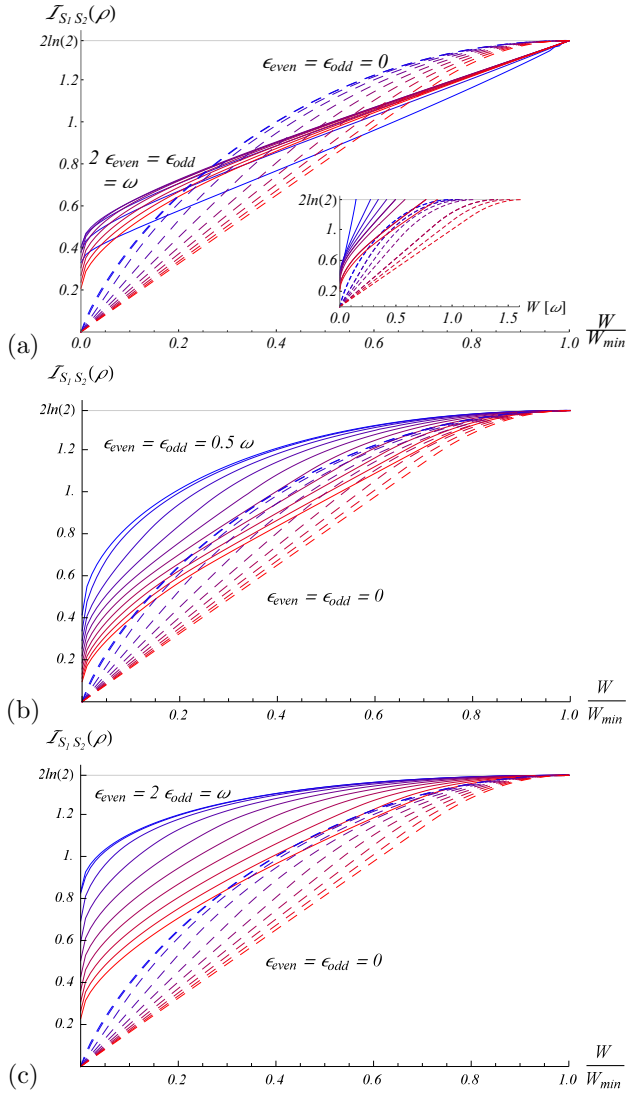

FIG. 3. **Fermionic correlation cost:** The maximal correlation of the final state that is achievable for a fixed input energy W [in units of $W_{\min}$ from Eq. (36)] is shown for temperatures $T = 0.1, \ldots, 1$ (in units of $\hbar\omega/k_B$) in steps of 0.1 (blue to red, top to bottom) for the ratios $\epsilon_{\text{even}}/\epsilon_{\text{odd}} = 2, 1$, and 0.5 in (a),(b), and (c), respectively. In all cases, the achievable final correlation is larger in the presence of interactions (solid lines) than in their absence (dashed lines). In (a) this can be seen from the inset plot, where the horizontal axis is not scaled with $W_{\min}$. For (b) and (c) one may deduce this directly from the plots, since the solid lines are strictly above their corresponding dashed lines, and $W_{\min}$ is maximal when $\epsilon_{\text{even}} = \epsilon_{\text{odd}} = 0$.

For a complete picture of the situation, it is also enlightening to study the behaviour of the total correlations $\mathcal{I}_{S_1 S_2}(\rho)$ for interacting and noninteracting systems, as shown in Fig. 3. In all cases that we have considered, the presence of the interactions leads to a larger amount of final state correlations $\mathcal{I}_{S_1 S_2}(\rho)$, irrespective of the (relative) size and sign of the coupling constants.

V. TWO BOSONIC MODES

In this section we study the creation of correlations for two bosonic modes. We present an example for which we explicitly show that the overall final-state correlation $\mathcal{I}_{S_1 S_2}(\rho)$ is always larger in the presence of interactions. This complements the observations made in Fig. 3, where

we come to same conclusion using a numerical approach.

Let us now consider the simple, yet versatile system of two bosonic modes with creation and annihilation operators a_i and $a_i^\dagger$ $(i = 1, 2)$, respectively. The mode operators satisfy the canonical commutation relations $[a_i, a_j^\dagger] = \delta_{ij}$ and $[a_i, a_j] = 0$. Such systems of two (or more) harmonics oscillators are of fundamental importance to quantum optics and quantum field theory. The correlations between bosonic modes have been extensively studied in continuous variable quantum information (see, e.g., Refs. [42, 43]) but they are also of interest in more specialized lines of research, such as, e.g., studies of entanglement in relativistic quantum field theory, see for example Refs. [44–46]. In addition to the usual free Hamiltonian

$$H_{S_1} + H_{S_2} = \omega \left(a_1^\dagger a_1 + a_2^\dagger a_2\right), \tag{37}$$

where we have assumed that the two modes have the same frequency ω, we will consider the interaction term

$$H_I = \epsilon \left(a_1 a_2 + a_1^\dagger a_2^\dagger\right), \tag{38}$$

with $\epsilon \in \mathbb{R}$. Since the system Hamiltonian $H_S = H_{S_1} + H_{S_2} + H_I$ is quadratic in the mode operators, any thermal state τ of H_S is a Gaussian state that is fully described by its second moments, that is, its covariance matrix Γ_S with components

$$\left(\Gamma_S\right)_{mn} = \mathrm{Tr}\big(\tau\big[\mathbb{X}_m\mathbb{X}_n + \mathbb{X}_n\mathbb{X}_m\big]\big), \tag{39}$$

with the quadrature operators $\mathbb{X}_{2n-1} = \left(a_n + a_n^\dagger\right)/\sqrt{2}$ and $\mathbb{X}_{2n} = -i\left(a_n - a_n^\dagger\right)/\sqrt{2}$. The first moments $\mathrm{Tr}\big(\tau\mathbb{X}_n\big)$, which would normally also enter into Eq. (39), vanish for the state τ. This can easily be seen by diagonalizing H_S using the Bogoliubov transformation

$$c_1 = \cosh(u)\, a_1 + \sinh(u)\, a_2^\dagger, \tag{40a}$$

$$c_2 = \cosh(u)\, a_2 + \sinh(u)\, a_1^\dagger, \tag{40b}$$

where $u = \frac{1}{2}\,\mathrm{artanh}(\epsilon/\omega)$, such that $[c_i, c_j^\dagger] = \delta_{ij}$ and $[c_i, c_j] = 0$. With this transformation, the system Hamiltonian becomes

$$H_S = \tilde{\omega}\left(c_1^\dagger c_1 + c_2^\dagger c_2\right) - 2\tilde{\omega}\sinh^2(u), \tag{41}$$

where $\tilde{\omega} = \sqrt{\omega^2 - \epsilon^2}$. The eigenstates of H_S are therefore the eigenstates of $c_1^\dagger c_1$ and $c_2^\dagger c_2$. Expanding the thermal state $\tau = \mathcal{Z}^{-1} e^{-\beta H_S}$ in terms of these eigenstates one quickly obtains $\mathrm{Tr}\big(\tau c_i\big) = \mathrm{Tr}\big(\tau c_i^\dagger\big) = 0$. Since the Bogoliubov transformation relating the operators $a_i, a_j^\dagger$ and $c_i, c_j^\dagger$ is linear, this implies that also $\mathrm{Tr}\big(\tau a_i\big) = \mathrm{Tr}\big(\tau a_i^\dagger\big) = 0$. The first moments vanish and Γ_S completely describes the state τ.

To assess the properties of the initial state, we then define the covariance matrix $\Gamma_S^{(c)}$ with respect to the operators c_i, in complete analogy to Eq. (39). For the thermal state τ, this 4×4 matrix is proportional to the identity, that is,

$$\Gamma_S^{(c)} = \nu(T)\mathbb{1}_4 = \coth\left(\tfrac{\tilde{\omega}}{2T}\right)\mathbb{1}_4, \tag{42}$$

where the identity $\mathbb{1}_4$ is the covariance matrix of the pure two-mode vacuum state with respect to the operators c_i. The mixedness of the state is hence captured solely by the prefactor $\nu(T) = \coth(\frac{\tilde{\omega}}{2T})$. The matrices $\Gamma_S^{(c)}$ and Γ_S are related by a symplectic transformation $\mathcal{S}$ corresponding to the unitary Bogoliubov transformation of Eq. (40), such that

$$\Gamma_S = \mathcal{S}\,\Gamma_S^{(c)}\mathcal{S}^T. \tag{43}$$

The transformation $\mathcal{S}$ leaves the symplectic form Ω, with components $\Omega_{kl} = -i\,[\mathbb{X}_k, \mathbb{X}_l]$ invariant, $\mathcal{S}\Omega\mathcal{S}^T = \Omega$. Consequently, also the eigenvalues $\nu(T)$ of $|i\Omega\Gamma_S|$, the symplectic eigenvalues are left unchanged by the transformation $\mathcal{S}$. This means that, up to the prefactor $\nu(T)$, the covariance matrix $\Gamma_S = \nu(T)\mathcal{S}\mathcal{S}^T$ represents a pure two-mode state, which is hence locally equivalent to two-mode squeezed state [47]. Due to the presence of $\nu(T)$, the overall state is nonetheless mixed and correlated, but may or may not be entangled, depending on the size of $\nu(T)$ [25, 48].

Any available energy W may then be used to further correlate the system by a combination of cooling [i.e., reducing $\nu(T)$] and two-mode squeezing along the direction in phase space corresponding to the two-mode squeezed state $\mathcal{S}\mathcal{S}^T$. These transformations leave the subsystems in local thermal states with respect to H_{S_1} and H_{S_2} and therefore optimally correlate[3] the subsystems at any given work cost [24]. Consequently, the presence of the interaction Hamiltonian H_I is here equivalent to an increased energy supply in the noninteracting case, and the overall correlations $\mathcal{I}_{S_1S_2}(\rho)$ are always larger than in the noninteracting case at a fixed work cost W.

The correlations that can in principle be generated in this infinite-dimensional Hilbert space are unbounded. However, the energy cost of creating additional correlations increases as the state becomes more correlated. The newly generated correlations $\Delta\mathcal{I}_{S_1S_2}$ may hence be more or less expensive than in the noninteracting case, depending on the initial temperature, coupling strength ϵ, and the available energy.

VI. CONCLUSION

We have investigated the work cost of creating correlations between interacting quantum systems and compared our results to previous studies [24, 25] of the corre-

[3] Note that two-mode squeezing is generally not the optimal entangling transformation and may be outperformed by non-Gaussian transformations [25].

lation cost in noninteracting systems. While the notion of isolated, noninteracting systems may appear more natural from the perspective of quantum communication scenarios, our approach here is motivated by the ubiquity of interactions present in nature. Hence, assuming that the presence of the interactions cannot be avoided or controlled, we find that the interactions can nonetheless be harnessed.

For such naturally occurring interactions we have identified general strategies for finite-dimensional systems to reduce the energy cost of creating correlations. These strategies, which apply to any finite-dimensional bipartite system with arbitrary interaction Hamiltonian, improve on previous bounds for non-interacting systems, at least in some low-energy regime. Nevertheless, the exact relation between the interactions and the correlation cost is complicated. The work cost of correlations strongly depends on the exact configuration of the interaction terms and thus on the underlying physics. To illustrate the general strategies, we therefore choose some exemplary physical systems — qubits, as well as fermionic and bosonic modes — to showcase the usefulness of the interactions.

In our examination, we have focused on the mutual information as a measure of the generated correlations, capturing both classical and genuine quantum correlations. The notoriously difficult case of characterizing the cost of entangling interacting quantum systems, which would provide further insight into the relation between the practically motivated resource theories of QI and QT, is hence left open for future investigation.

Moreover, while we have here considered arbitrary operations on the system, inevitable noise and practical design may favor operations that can be directly implemented through the natural interactions present in the underlying systems. It would hence be interesting to compare such physically motivated protocols with the optimal protocols derived here, including also noncyclic processes where the interactions can be switched on or off at will. Further open questions include the general cost and impact of interactions on single-shot information processing capabilities.

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Appendix: Correlations in a fermionic thermal state

Here we study the correlations present in the initial thermal state τ_β of two fermionic modes, given by Eq. (30). Whereas the thermal states of the noninteracting Hamiltonian (i.e., for $\epsilon_{\text{even,odd}} = 0$) are uncorrelated, the thermal states of interacting Hamiltonians feature some correlations. For instance, consider the ground state, that is, the limit $T \to 0$ ($\beta \to \infty$), which arises as the eigenstate of the Hamiltonian with the smallest eigenvalue. As can be seen from Eq. (28), this can be either $\| \lambda_3 \rangle\!\rangle$ or $\| \lambda_4 \rangle\!\rangle$, depending on the relative sizes of $|\epsilon_{\text{odd}}|$ and $\sqrt{\omega^2 + \epsilon_{\text{even}}^2}$. That is,

$$\tau(\beta \to \infty) = \begin{cases} \rho_3 & \text{if } |\epsilon_{\text{odd}}| > \sqrt{\omega^2 + \epsilon_{\text{even}}^2} \\ \frac{1}{2}\big(\rho_3 + \rho_4\big) & \text{if } |\epsilon_{\text{odd}}| = \sqrt{\omega^2 + \epsilon_{\text{even}}^2} \\ \rho_4 & \text{if } |\epsilon_{\text{odd}}| < \sqrt{\omega^2 + \epsilon_{\text{even}}^2} \end{cases}, \tag{A.1}$$

with $\rho_3 = \| \lambda_3 \rangle\!\rangle\langle\!\langle \lambda_3 \|$ and $\rho_4 = \| \lambda_4 \rangle\!\rangle\langle\!\langle \lambda_4 \|$. Both $\| \lambda_3 \rangle\!\rangle$ and $\| \lambda_4 \rangle\!\rangle$ are correlated, but only the former state is maximally correlated. It is hence expected that the relative sizes of the coupling constants strongly influence the initial amount of correlations, see Fig. 4. To evaluate the mutual information of Eq. (7), we still need to specify the reduced density operators. These are found to be diagonal, with matching matrix elements, i.e.,

$$\tau_{S_1}(\beta) = \tfrac{1}{2}(1+\tau_0)\, \| 0 \rangle\!\rangle\langle\!\langle 0 \| + \tfrac{1}{2}(1-\tau_0)\, \| 1_1 \rangle\!\rangle\langle\!\langle 1_1 \| \,, \tag{A.2a}$$

$$\tau_{S_2}(\beta) = \tfrac{1}{2}(1+\tau_0)\, \| 0 \rangle\!\rangle\langle\!\langle 0 \| + \tfrac{1}{2}(1-\tau_0)\, \| 1_2 \rangle\!\rangle\langle\!\langle 1_2 \| \,, \tag{A.2b}$$

with the coefficient τ_0 given by

$$\tau_0 = \frac{\omega \, \sinh\!\big(\beta\sqrt{\omega^2 + \epsilon_{\text{even}}^2}\,\big)}{\sqrt{\omega^2 + \epsilon_{\text{even}}^2}\Big(\cosh\!\big(\beta\epsilon_{\text{odd}}\big) + \cosh\!\big(\beta\sqrt{\omega^2 + \epsilon_{\text{even}}^2}\big)\Big)} \,. \tag{A.3}$$

With the eigenvalues of the thermal state given by $e^{-\beta\lambda_i}$ and those of the marginals by $\frac{1}{2}(1\pm\tau_0)$ one can then easily evaluate the entropies $S(\tau)$, $S(\tau_{S_1})$ and $S(\tau_{S_2})$, and hence the mutual information, shown in Fig. 4.

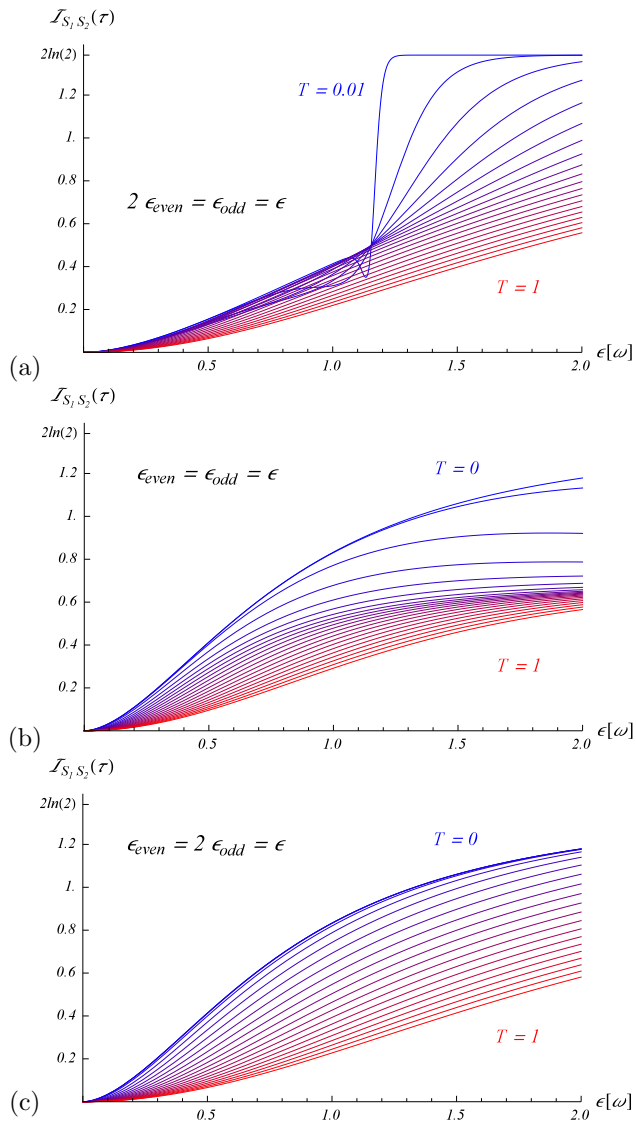

FIG. 4. **Fermionic thermal state correlation:** The correlation of the initial thermal state $\tau(\beta)$, as measured by the mutual information $\mathcal{I}_{S_1 S_2}$, depend on the (relative) and absolute sizes of the couplings ϵ_{even} and ϵ_{odd}. The correlation is plotted for temperatures $T = 0(0.01), \ldots, 1$ (in units of $\hbar\omega/k_B$) in steps of 0.05 (blue to red, top to bottom) for the ratios $\epsilon_{\text{even}}/\epsilon_{\text{odd}} = 2, 1$, and 0.5 in (a),(b), and (c), respectively.

II.3 Passivity and Practical Work Extraction using Gaussian Operations

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Overview: We have studied the problem of work extraction from bosonic modes using Gaussian operations. A central output of the paper is establishing the notion of Gaussian passivity, describing states from which no work can be extracted via Gaussian unitaries realized by cyclic Hamiltonian processes. The main results of the paper are a full characterization of all Gaussian-passive states in terms of their first and second statistical moments (Theorem 1 and Corollary 1), an explicit protocol for Gaussian work extraction that reaches Gaussian passivity, and a proof that the potential gap between Gaussian passivity and passivity is maximal when only the system entropy, first and second moments of the state are known (Theorem 2 and Lemma 1).

Contribution: Together with my coauthors I derived Theorem 1 and the Gaussian work-extraction protocol. I formulated the hypothesis for the maximal gap between passivity and Gaussian passivity and proved Theorem 2 and Lemma 1. I wrote significant parts of the manuscript.

Passivity and practical work extraction using Gaussian operations

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Quantum states that can yield work in a cyclical Hamiltonian process form one of the primary resources in the context of quantum thermodynamics. Conversely, states whose average energy cannot be lowered by unitary transformations are called passive. However, while work may be extracted from non-passive states using arbitrary unitaries, the latter may be hard to realize in practice. It is therefore pertinent to consider the passivity of states under restricted classes of operations that can be feasibly implemented. Here, we ask how restrictive the class of Gaussian unitaries is for the task of work extraction. We investigate the notion of Gaussian passivity, that is, we present necessary and sufficient criteria identifying all states whose energy cannot be lowered by Gaussian unitaries. For all other states we give a prescription for the Gaussian operations that extract the maximal amount of energy. Finally, we show that the gap between passivity and Gaussian passivity is maximal, i.e., Gaussian-passive states may still have a maximal amount of energy that is extractable by arbitrary unitaries, even under entropy constraints.

I. INTRODUCTION

At the very core of quantum thermodynamics — which has recently seen a surge in interest from the quantum information community [1–3]— lies the task of extracting work from quantum systems. Ideally, this is achieved by reversible cyclic processes that can be represented by unitary transformations, which, in turn, form the most basic primitive of a cyclically operating engine. However, from so-called *passive* states [4] no work can be extracted unitarily if only a single copy of the system is available. Viewing quantum thermodynamics as a resource theory of work extraction [5, 6], it is therefore tempting to view passive states as being freely available. Curiously, most passive states still contain extractable work, that can only be accessed by processing multiple copies using entangling operations [7]. This has sparked interested in the role of entanglement in work extraction [8] and more generally in the structure of passive states [9]. States from which no energy can be extracted unitarily, no matter how many copies are available, are called completely passive, and without further conserved charges the unique completely passive state is the so-called thermal state. Naturally, the extraction of work from non-passive quantum states using arbitrary unitaries has therefore been the subject of many fruitful investigations (see, e.g., Refs. [10–13]). This has provided useful insights into the role of coherence, correlations, and entanglement for work extraction [14–17], and conversely, about the work cost for establishing correlations [18–20] and coherence [21].

However, the unitary operations that extract the maximal amount of work from a given non-passive state may be difficult to realize in practice. For example, the global entangling unitaries required to extract work from passive but not completely passive states are not feasible under realistic conditions, which already leads to a discrepancy between theoretically extractable work and what is practically achievable. Consequently, the characterization of states as non-completely passive may fail to accurately represent how useful a quantum system is for thermodynamical work extraction in practice. It is hence of interest to study the *ergotropy* [10], i.e., the maximal amount of work that can be unitarily extracted in a Hamiltonian process, under the restriction to practically realizable transformations. An important example of such a constraint in continuous variable systems is the class of Gaussian unitary operations, which, although being standard operations in quantum optical systems (see, e.g., Refs. [22, 23]), represents a significant restriction of the set of all possible transformations. This manifests, amongst other things, in the fact that Gaussian operations are not universal for quantum computation [24].

It is the aim of this paper to investigate this interesting dichotomy between what is possible in principle and what is practically feasible within quantum thermodynamics, focusing on Gaussian unitary transformations. We provide a full characterization of Gaussian passivity for multi-mode states, i.e., we give necessary and sufficient conditions to identify all (not necessarily Gaussian) quantum states from which no work can be extracted using Gaussian transformations. This characterization only requires knowledge of the first and second statistical moments of the state in question, independently of whether the state itself is Gaussian or not, and imme-

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diately provides a protocol for optimal Gaussian work extraction given any number of modes. Finally, we show that the gap between passivity and Gaussian passivity can be maximal if only the first and second moments of the state are known. That is, we show that the first and second moments of any Gaussian-passive state (which need not be a Gaussian state, and hence is not to be confused with a passive Gaussian state) are in principle compatible with those of a state of maximal ergotropy, even under entropy constraints.

To examine the usefulness of Gaussian transformations in the context of thermodynamic work extraction, we first review the notion of passivity in Section II. In Section III we introduce the notion of Gaussian passivity and formulate our main result, the characterization of all Gaussian-passive states, before examining the ergotropic gap in Section IV. In Section V we finally discuss the consequences and applications of our results.

II. PASSIVE STATES

On an elementary level, controlled engines perform their tasks based on repeated cycles, during which the system dynamics are typically changed through an external control. The resulting time evolution of the system is hence governed by a time-dependent Hamiltonian $H(t)$. At the end of each cycle of duration τ the system is returned to its initial Hamiltonian, i.e., $H(n\tau) = H(0) \equiv H_0$ for any integer n, leading to unitary dynamics perturbed only by the necessary interactions with the environment. In this sense the unitary orbits of the input quantum states determine the fundamental limits of operation of cyclic machines, which is one of the reasons that unitary work extraction has recently attracted attention [7–9, 17, 25, 26].

Within this paradigm, the elementary processes that we consider here can in principle generate work if the average energy of a given system can be reduced by unitary operations. That is, if for a system with Hilbert space $\mathcal{H}$ described by a density operator $\rho \in L(\mathcal{H})$ there exists a unitary operator $U \in L(\mathcal{H})$, such that

$$\tilde{E} = \mathrm{Tr}\big(H_0 U \rho U^\dagger\big) < \mathrm{Tr}\big(H_0 \rho\big) = E\,. \tag{1}$$

States for which the average energy cannot be reduced by unitaries are called *passive*. All passive states ρ_{pass} are diagonal in the eigenbasis $\{|n\rangle\}$ of H_0 with probability weights decreasing (not necessarily strictly) with increasing energy [4], that is, passive states ρ_{pass} can be written as

$$\rho_{\text{pass}} = \sum_{n=0}^{d-1} p_n \, |n\rangle\langle n| \,, \tag{2}$$

where $p_n \leq p_m$ when $E_n \geq E_m$, and $d = \dim(\mathcal{H})$, and, as usual, $0 \leq p_n \leq 1$ and $\sum_n p_n = 1$, while the energy eigenstates satisfy

$$H_0 \, |n\rangle = E_n \, |n\rangle \,. \tag{3}$$

To see this, simply consider a two-dimensional subspace spanned by $|m\rangle$ and $|n\rangle$ with $E_m < E_n$. To decrease the average energy using a unitary on this subspace, the corresponding probability weights must satisfy $p_m < p_n$. Any state for which this isn't the case for any two-dimensional subspace is passive, and is of the form of Eq. (2).

An example for a passive state in continuous variable systems is a product state of two thermal states of two bosonic modes at the same frequency ω and temperature $T = 1/\beta$ (in units where $\hbar = k_{\text{B}} = 1$). A bosonic mode is represented by annihilation and creation operators a and $a^\dagger$, satisfying $[a\,, a^\dagger] = 1$, and a Hamiltonian $H_0 = \omega\, a^\dagger a$. The operator a annihilates the vacuum state, i.e., $a\,|0\rangle = 0$, and the eigenstates of the Hamiltonian, the Fock states, are obtained by applying the creation operators $|n\rangle = (a^\dagger)^n/\sqrt{n!}\,|0\rangle$. A thermal state $\tau(\beta)$ of temperature $T = 1/\beta$ is given by

$$\tau(\beta) = \frac{e^{-\beta H_0}}{\mathcal{Z}}\,, \tag{4}$$

where $\mathcal{Z} = \mathrm{Tr}(e^{-\beta H_0})$ is the partition function. In the Fock basis the thermal state reads

$$\tau(\beta) = (1 - e^{-\beta\omega}) \sum_n e^{-n\beta\omega} \, |n\rangle\langle n| \,. \tag{5}$$

Since thermal states are the only completely passive states, any resource state for a cyclic engine must be out of thermal equilibrium. The most elementary engine is hence a heat engine, which only needs access to two thermal baths at different temperatures. This is arguably the simplest out-of-equilibrium resource: Given two thermal baths at equal temperature, increasing the temperature of one of them can be achieved by increasing the energy of one of the systems without requiring any knowledge of its microstates.

At this point, it seems prudent to restate the above observation about these elementary heat engines in a more technical manner by reminding the reader of the elementary fact that, for two bosonic modes at the same frequency, the product state of two thermal states with different temperatures is not passive. Since we will refer to it later in the manuscript, it is instructive to examine one potential strategy to prove this statement. Consider two bosonic modes at the same frequency ω, with temperatures T_a and $T_b > T_a$, respectively. The product state of the two thermal states is

$$\begin{aligned}\tau(T_a, T_b) = {} & (1 - e^{-\omega/T_a})(1 - e^{-\omega/T_b}) \\ & \times \sum_{m,n} e^{-\omega(\frac{m}{T_a} + \frac{n}{T_b})} \, |m\rangle\langle m| \otimes |n\rangle\langle n| \,.\end{aligned} \tag{6}$$

Up to a common prefactor, the matrix elements are

$$e^{-\omega(\frac{m}{T_a}+\frac{n}{T_b})} = e^{-\frac{\omega}{T_a T_b}(mT_b+nT_a)} . \tag{7}$$

The state is not passive if there exist pairs of non-negative integers m, n and m', n', such that

$$m\,T_b + n\,T_a > m'\,T_b + n'\,T_a\,, \tag{8}$$

while $m' + n' > m + n$. Now consider, e.g., the case where $m = n = x/2$, while $m' = 0$ and $n' = x + 1$ for some even non-negative integer x. In this case, the second inequality is trivially true for all x and the first condition is

$$x(T_a + T_b)/2 > (x + 1)\,T_a\,, \tag{9}$$

which implies $x > 2T_a/(T_b - T_a) > 0$. So by picking x large enough, one can always find a two-dimensional subspace, in which a unitary can reduce the average energy, proving that the state $\tau(T_a, T_b)$ of Eq. (6) is not passive.

However, practically realizing these unitaries on arbitrary (two-dimensional) subspaces of the overall Hilbert space may prove to be practically unachievable. Therefore, it is of interest to investigate the limitations of work extraction by operations that can be feasibly implemented.

III. GAUSSIAN-PASSIVE STATES

While the fact that $\tau(T_a, T_b)$ of Eq. (6) is not passive in principle allows the construction of a heat engine the necessary unitaries to extract energy may be difficult to realize in practice. A set of operations that are in general easier to implement are Gaussian unitaries, which are generated by Hamiltonians that are at most quadratic in the mode operators, and transform Gaussian states to Gaussian states. These, in turn, are states whose characteristic Wigner function is Gaussian (see, e.g., Ref. [23] for a detailed review). Such states are fully described by their first and second statistical moments, that is, the expectation values of linear and quadratic combinations of the quadrature operators $\mathbb{X}_i$, where $\mathbb{X}_{2n-1} = \big(a_n + a_n^\dagger\big)/\sqrt{2}$ and $\mathbb{X}_{2n} = -i\big(a_n - a_n^\dagger\big)/\sqrt{2}$, and $n = 1, 2, \ldots, N$ labels the N modes of the system in question. The second moments are collected in the real, symmetric, and positive definite $2N \times 2N$ covariance matrix Γ, with components

$$\Gamma_{ij} = \langle\, \mathbb{X}_i\mathbb{X}_j + \mathbb{X}_j\mathbb{X}_i \,\rangle \,-\, 2\,\langle\, \mathbb{X}_i \,\rangle\,\langle\, \mathbb{X}_j \,\rangle\,, \tag{10}$$

where $\langle A \rangle = \mathrm{Tr}(A\rho)$ is the expectation value of the operator A in the state ρ. For example, the thermal state of a single bosonic mode that we have encountered in Eq. (5) belongs to the class of Gaussian states and is of particular interest for us here. Its first moments vanish, $\langle\, \mathbb{X}_i \,\rangle = 0$, while the covariance matrix is given by $\Gamma_{\text{thermal}} = \coth\big(\beta\omega/2\big)\mathbb{1}$.

Gaussian unitaries are described by affine maps (S, ξ) : $\mathbb{X} \mapsto S\mathbb{X} + \xi$, where $\xi \in \mathbb{R}^{2N}$ represent phase space displacements, and S are real, symplectic matrices. By definition, a symplectic operation S leaves invariant the symplectic form Ω with components $\Omega_{mn} = i\,[\,\mathbb{X}_m\,, \mathbb{X}_n\,] = \delta_{m,n-1} - \delta_{n,m+1}$, i.e.,

$$S\,\Omega\,S^T = \Omega\,. \tag{11}$$

Displacements, represented by the unitary Weyl operators $D(\xi) = \exp\big(i\sqrt{2}\mathbb{X}^T\Omega\xi\big)$, do not affect the covariance matrix but rather shift the first moments. While all of the mentioned transformations preserve the Gaussian character of Gaussian states, one can of course also consider the effects of Gaussian transformations on any arbitrary state via the effect on the corresponding covariance matrix and vector of first moments. We are now interested in determining for which (not necessarily Gaussian) states of two noninteracting bosonic modes with frequencies ω_a and ω_b (w.l.o.g. we assume $\omega_b \geq \omega_a$), energy can be extracted using only Gaussian operations. In analogy to the previous terminology we call states *Gaussian-passive* if their average energy cannot be reduced by Gaussian unitaries. The first important step in analyzing this property is the ability to identify Gaussian-passive states, which is established by the following theorem.

Theorem 1. *Any (not necessarily Gaussian) state of two noninteracting bosonic modes with frequencies ω_a and $\omega_b \geq \omega_a$ is Gaussian-passive if and only if its first moments vanish, $\langle\, \mathbb{X} \,\rangle = 0$, and its covariance matrix Γ is either*

(i) in Williamson normal form [27]

$$\Gamma = \mathrm{diag}\{\nu_a, \nu_a, \nu_b, \nu_b\}$$

with $\nu_a \geq \nu_b$ for $\omega_a < \omega_b$. Or, in the case of equal frequencies $\omega_a = \omega_b$, the state may also be

(ii) in standard form [28, 29]

$$\Gamma = \begin{pmatrix} a\mathbb{1} & C \\ C & b\mathbb{1} \end{pmatrix}$$

with $C = c\mathbb{1}$, if $\omega_a = \omega_b$.

Proof. To prove Theorem 1, we will proceed in the following way. We will start with the most general combination of first moments $\langle\, \mathbb{X} \,\rangle$ and second moments Γ that any initial state may have, before successively applying Gaussian operations (steps P1-P4) to reduce the average energy. When we reach a state whose energy cannot be further reduced by Gaussian unitaries, we compare its energy to that of the initial state and identify under which conditions the energy has been lowered with respect to the initial state. These conditions will finally result in the identification of the characteristics of Gaussian-passive states as stated in clauses (i) and (ii) above.

(P1) **Displacements**: As we consider noninteracting bosonic modes, the average energy of a two-mode

state is given by the sum of the average energies of the individual modes. For a single mode with frequency ω_a and ladder operators a and $a^\dagger$, a state described by the density operator ρ has the average energy $E(\rho) = \omega_a \mathrm{Tr}(\rho\, a^\dagger a)$, which can be written in terms of the state's covariance matrix Γ_a and vector of first moments $\langle\, \mathbb{X}_a\, \rangle$ as

$$E(\rho) = \omega_a \Big(\frac{1}{4}\big[\mathrm{Tr}(\Gamma_a) - 2\big] + \frac{1}{4}\|\langle\, \mathbb{X}_a \rangle\|^2 \Big)\,, \tag{12}$$

where $\|\cdot\|$ is the (Euclidean) norm. Since displacements change the first moments but leave the second moments invariant, the energy of the state can always be decreased by shifting $\langle\, \mathbb{X}_a \rangle$ to the null vector. Conversely, every state with nonvanishing first moments cannot be Gaussian-passive, since its energy can be lowered by appropriate displacements. From here on we may hence consider only states for which the energy has been reduced by displacements as much as possible, such that for each mode one has $\langle\, \mathbb{X}_a \rangle = 0$. In the following, one may then apply Gaussian unitaries represented by symplectic transformations S, which leave the zero first moments invariant.

(P2) **Local symplectic operations**: In the next step, we note that every two-mode covariance matrix Γ can be brought to the standard form Γ_{st} [28, 29] by local symplectic operations $S_{\mathrm{loc}} = S_{\mathrm{loc},a} \oplus S_{\mathrm{loc},b}$, that is,

$$S_{\mathrm{loc}}\, \Gamma\, S_{\mathrm{loc}}^T = \Gamma_{\mathrm{st}} = \begin{pmatrix} a\,\mathbb{1} & C \\ C & b\,\mathbb{1} \end{pmatrix}\,, \tag{13}$$

where $C = \mathrm{diag}\{c_1, c_2\}$. Each of the single-mode symplectic operations $S_{\mathrm{loc},i}$ $(i = a, b)$ can be decomposed into phase rotations and single-mode squeezing as

$$S_{\mathrm{loc},i} = R(\theta_i)\, S(r_i)\, R(\phi_i)\,. \tag{14}$$

For some real angles θ_i and ϕ_i, and real squeezing parameters r_i, these local operations take the form

$$R(\theta_i) = \begin{pmatrix} \cos\theta_i & \sin\theta_i \\ -\sin\theta_i & \cos\theta_i \end{pmatrix}\,, \quad S(r_i) = \begin{pmatrix} e^{-r_i} & 0 \\ 0 & e^{r_i} \end{pmatrix}\,. \tag{15}$$

Conversely, this means that we can write the covariance matrix Γ as

$$\Gamma = (S_{\mathrm{loc}}^{-1})\, \Gamma_{\mathrm{st}}\, (S_{\mathrm{loc}}^{-1})^T\,, \tag{16}$$

where the inverse operations are also local symplectic transformations $S_{\mathrm{loc}}^{-1} = S_{\mathrm{loc},a}^{-1} \oplus S_{\mathrm{loc},b}^{-1}$. The single-mode inverses $S_{\mathrm{loc},i}^{-1}$ are simply

$$S_{\mathrm{loc},i}^{-1} = R(-\phi_i)\, S(-r_i)\, R(-\theta_i)\,. \tag{17}$$

This allows us to express the energy of the state described by the covariance matrix Γ as

$$E(\Gamma) = \frac{\omega_a}{2}\big(a\cosh(2r_a) - 1\big) + \frac{\omega_b}{2}\big(b\cosh(2r_b) - 1\big)\,. \tag{18}$$

Since $\cosh(2r_i) \geq 1$, it becomes clear that the energy of a state with covariance matrix Γ can be lowered by local symplectic operations until Γ reaches the standard form. Consequently, states for which $\Gamma \neq \Gamma_{\mathrm{st}}$ are not Gaussian-passive, whereas states with covariance matrices in the standard form may still have energy that can be reduced by global symplectic transformations.

(P3) **Two-mode squeezing**: After using local Gaussian operations to extract as much energy as possible, one is hence left with a state whose covariance matrix is in the standard form of Eq. (13). The local covariance matrices of each mode are then proportional to the identity, $a\mathbb{1}$ and $b\mathbb{1}$, but the off-diagonal block C may have two different diagonal elements c_1 and c_2. If this is the case, we can apply a two-mode squeezing operation to reduce the energy and bring the covariance matrix to a form in which the off-diagonal block is proportional to the identity as well. The symplectic representation of this global transformation is

$$S_{\mathrm{TMS}} = \begin{pmatrix} \cosh(r)\mathbb{1} & \sinh(r)\sigma_z \\ \sinh(r)\sigma_z & \cosh(r)\mathbb{1} \end{pmatrix}\,, \tag{19}$$

where $\sigma_z = \mathrm{diag}\{1, -1\}$ is the usual Pauli matrix and the squeezing parameter r that achieves equal off-diagonal elements is given by

$$r = -\tfrac{1}{2}\,\mathrm{artanh}\Big(\frac{c_1 - c_2}{a + b}\Big)\,. \tag{20}$$

To show that this transformation always reduces the average energy, we compute the energy $E(\tilde{\Gamma})$ associated to the two-mode squeezed covariance matrix $\tilde{\Gamma} = S_{\mathrm{TMS}}\Gamma_{\mathrm{st}}S_{\mathrm{TMS}}^T$ and find

$$\begin{aligned} E(\tilde{\Gamma}) = {} & \frac{\omega_a}{2}\big[a\cosh^2(r) + b\sinh^2(r)\big] \\ & + \frac{\omega_b}{2}\big[b\cosh^2(r) + a\sinh^2(r)\big] \\ & + \frac{\omega_a + \omega_b}{4}\big[(c_1 - c_2)\sinh(2r) - 1\big]\,. \end{aligned} \tag{21}$$

We then take the derivative with respect to r and set $\partial E(\tilde{\Gamma})/\partial r = 0$, which provides the condition

$$(a + b)\sinh(2r) + (c_1 - c_2)\cosh(2r) = 0\,, \tag{22}$$

which in turn is solved by r from Eq. (20). It is then easy to check that for this value of r we have $\partial^2 E(\tilde{\Gamma})/\partial r^2 > 0$, indicating that the energy is minimal for the specified value of the squeezing param-

eter. The two-mode squeezing transformation with this strength hence reduces the energy. While the off-diagonal block of the covariance matrix is proportional to the identity after this operation, the local covariance matrices are generally not of this form, albeit still being diagonal. We can then use local rotations $R(\vartheta,\varphi) = R(\vartheta) \oplus R(\varphi)$, which leave the energy invariant, to bring the covariance matrix back to the standard form where every 2×2 subblock is now proportional to the identity, i.e.,

$$\widehat{\Gamma} \;=\; R(\vartheta,\varphi)\,\tilde{\Gamma}\,R^T(\vartheta,\varphi) = \begin{pmatrix} \tilde{a}\,\mathbb{1} & c\,\mathbb{1} \\ c\,\mathbb{1} & \tilde{b}\,\mathbb{1} \end{pmatrix} . \tag{23}$$

In some circumstances, the third step of the protocol can be seen as the conversion of Gaussian entanglement into work. Note that the previous two steps consist of local unitaries, and hence leave any entanglement measure invariant. If the initial state is a Gaussian state, the form of $\widehat{\Gamma}$ in Eq. (23) further indicates that no more entanglement is present after step P3, since a nonnegative determinant of the 2×2 off-diagonal block is a sufficient separability criterion for two-mode Gaussian states [29]. For any Gaussian state, the presence of entanglement hence indicates that the energy can be lowered by Gaussian unitaries in the third step. However, the fact that the energy of a Gaussian state can be lowered in step P3, does not imply that the initial state is entangled [19]. Moreover, if the initial state is not Gaussian, the final state after step P3 may still be entangled in general.

(P4) **Beam splitting**: Having reached a state with a covariance matrix as in Eq. (23), we have exhausted all local Gaussian operations as well as two-mode squeezing to lower the energy. In particular, at this point we know that applying any local or global squeezing transformation can only increase the energy. This leaves only the beam splitting transformation as a last Gaussian unitary that we still have at our disposal. This transformation, represented by the global orthogonal symplectic matrix

$$S_{\mathrm{BS}}(\theta) = \begin{pmatrix} \cos(\theta)\,\mathbb{1} & \sin(\theta)\,\mathbb{1} \\ \sin(\theta)\,\mathbb{1} & -\cos(\theta)\,\mathbb{1} \end{pmatrix} \tag{24}$$

for real values of θ, is an optically passive transformation. That is, it leaves the average excitation number unchanged. If the frequencies of the two modes are the same, $\omega_a = \omega_b$, then such a transformation obviously also leaves the average energy unchanged. In this case, the energy of the state cannot be further lowered by any Gaussian unitary and we conclude that the state is hence Gaussian-passive, which proves clause (ii) of Theorem 1.

If the frequencies are not the same we may assume w.l.o.g. that $\omega_a < \omega_b$. Then, the energy can be lowered by shifting as many excitations as possible to the lower frequency mode. To prove this rigorously, we compute the average energy of

$$\Gamma_{\mathrm{GP}} \;=\; S_{\mathrm{BS}}(\theta)\,\widehat{\Gamma}\,S_{\mathrm{BS}}^T(\theta)\,, \tag{25}$$

for which we find

$$E(\Gamma_{\mathrm{GP}}) = \frac{\omega_a}{2}\big[a\cos^2(\theta) + b\sin^2(\theta) + c\sin(2\theta) - 1\big] + \frac{\omega_b}{2}\big[b\cos^2(\theta) + a\sin^2(\theta) - \sin(2\theta) - 1\big]. \tag{26}$$

Similarly as for the two-mode squeezing we then set $\partial E(\Gamma_{\mathrm{GP}})\partial\theta = 0$ and find that the energy is minimized when

$$\theta = \begin{cases} \frac{1}{2}\arctan(\frac{2c}{a-b}) & \text{if } a \geq b \\ \frac{1}{2}\arctan(\frac{2c}{a-b}) + \frac{\pi}{2} & \text{if } a < b \end{cases} . \tag{27}$$

The resulting covariance matrix $\Gamma_{\mathrm{GP}} = \mathrm{diag}\{\nu_a,\nu_a,\nu_b,\nu_b\}$ is in Williamson normal form [27], its eigenvalues coincide with its symplectic eigenvalues, and the lower frequency mode now has the higher population, $\nu_a \geq \nu_b$. Any further Gaussian unitary applied to this final state would bring the covariance matrix (and/or the first moments) to a form that would allow reducing the energy via one (or several) of the steps P1-P4. The corresponding symplectic operations leave the symplectic eigenvalues invariant. Consequently, the second moments of the initial state uniquely determine the associated Gaussian-passive covariance matrix Γ_{GP}. That is, Γ_{GP} is the only covariance matrix with symplectic spectrum $\{\nu_a,\nu_a,\nu_b,\nu_b\}$ whose energy cannot be lowered by Gaussian unitaries. (If $\omega_a = \omega_b$, the Gaussian-passive covariance matrix is not unique, but is determined only up to arbitrary optically passive transformations.) We therefore arrive at the conclusion that the state associated to the covariance matrix Γ_{GP} is Gaussian-passive. Any state whose covariance matrix is not of this form can be subjected to one (or several) of the steps P1-P4 to reduce its average energy, and is hence not Gaussian-passive, which concludes the proof. □

Note that for any given initial state (which need not be Gaussian), the corresponding Gaussian-passive state is not unique, because the operations P1-P4 do not commute. For instance, applying the operations of step P1 after any of the other steps leads to different final states that have the same first and second moments, and hence the same final energy. The symplectic eigenvalues of the initial state hence uniquely define the lowest energy that can be reached via Gaussian unitaries, but several (non-Gaussian) states (equivalent up to energy conserving Gaussian unitaries) may be compatible with the corresponding Gaussian-passive covariance matrix.

A corollary that follows immediately from Theorem 1

concerns the extension to an arbitrary number of modes.

Corollary 1. *An arbitrary state of n bosonic modes is Gaussian-passive if and only if all of its two-mode marginals are Gaussian-passive.*

Proof. To prove this statement, simply note that all Gaussian unitaries can be decomposed into sequences of operations on one or two modes. Consequently, if a state admits no two-mode marginal whose energy can be lowered by Gaussian unitaries, then the overall state must be Gaussian-passive. □

An interesting example for a Gaussian-passive state of two modes with different frequencies is that of a product of single-mode thermal states, in which each mode has a different temperature. In this case the symplectic eigenvalues are $\nu_i = \coth(\frac{\omega_i}{2T_i})$ and for $T_b \neq 0$ the condition $\nu_a > \nu_b$ for Gaussian passivity can be expressed as

$$\frac{\omega_a}{\omega_b} < \frac{T_a}{T_b}\,. \tag{28}$$

Now, recall from Section II that we know that within the framework of general operations the product states of two thermal states at different temperatures is not passive, regardless of the frequencies of the modes involved. However, as we have seen, such a state may nonetheless be Gaussian-passive depending on the relation between the local temperatures and frequencies.

Here, a word of caution is in order. Since (Gaussian) unitaries leave the purity $\mathrm{Tr}(\rho^2) = 1/\sqrt{\det(\Gamma)}$ unchanged, one may be tempted to (falsely) conclude that the existence of (Gaussian) states that have the same purity as a given Gaussian-passive state but a lower average energy means that one may further reduce the energy of Gaussian-passive states beyond what is stated in Theorem 1. For example, for $\nu_a\nu_b < \omega_b/\omega_a$ the Gaussian state with covariance matrix

$$\Gamma' = \begin{pmatrix} \nu_a\nu_b\,\mathbb{1} & 0 \\ 0 & \mathbb{1} \end{pmatrix}\,, \tag{29}$$

has the same purity as the Gaussian-passive state specified in clause (i). However, such states cannot be reached by Gaussian unitaries if their symplectic eigenvalues ($\nu_a\nu_b$ and 1 in the example) do not match those of the original state. In general, there may not even exist a non-Gaussian unitary (even if it preservers the Gaussian character of the specific state in question) that transforms the corresponding states into each other. Finally, note that all passive states are obviously Gaussian-passive, but the converse is not true.

IV. THE GAP BETWEEN PASSIVITY AND GAUSSIAN PASSIVITY

Given the characterization of a given state as Gaussian-passive, it is now natural to ask how much extractable energy is potentially sacrificed by the restriction to Gaussian unitary orbits, rather than general unitary transformations. Suppose that one only has knowledge of and access to the first and second moments of an arbitrary state of two bosonic modes. With this information, which can practically be easily obtained in several ways (see, e.g., Ref. [30]), one may use Gaussian unitaries to lower the energy of the state until reaching a Gaussian-passive state. One may then wonder how much more energy could have been extracted if general unitary operations could be applied. The answer to this question of course depends on the particular state in question. So far, we have only fixed the first and second moments, which identifies states uniquely only if they are Gaussian. It is hence crucial to understand which (non-Gaussian) states are in general compatible with a given set of first and second moments. A first important observation can be phrased in the following lemma.

Lemma 1. *The first and second moments of any Gaussian-passive state are compatible with a (non-Gaussian) pure state for which the entire energy is extractable by unitary transformations.*

Proof. To prove the lemma first note that any Gaussian-passive state of an arbitrary number of modes with different frequencies (clause (i) of Theorem 1) has a locally thermal covariance matrix with different effective temperatures for each mode. In this case it is therefore enough to consider a single mode in a thermal state with arbitrary temperature, and show that there exists a pure state with the same first and second moments. If such a pure state exists for a single mode for any temperature, then one can certainly find pairs of states of this kind whose tensor product is compatible with a Gaussian-passive, locally thermal two-mode state.

In the case that the covariance matrix has nonzero off-diagonal blocks, i.e., if clause (ii) of Theorem 1 applies, the covariance matrix can be brought to the locally thermal form by an energy conserving, Gaussian unitary, that is, a beam splitting transformation with angle θ given by Eq. (27). Then, as before, one is required to find a pure state that matches the resulting locally thermal covariance matrix. Applying the inverse of the beam splitting operation to this state, one finally obtains a pure two-mode state compatible also for Gaussian-passive states with non-diagonal covariance matrices.

To identify the pure states in question, recall that the first moments of a Gaussian-passive state must vanish. This is also the case for all Fock states $|n\rangle = (a^\dagger)^n/\sqrt{n!}\,|0\rangle$. Indeed, this is even true for all superpositions of Fock states that differ by two or more excitations, for instance, all states of the form $\sum_k c_k\,|n + m\,k\rangle$ for any $n, m \in \mathbb{N}_0$ and $m \geq 2$. Restricting to this family of states we are interested in identifying those members that also have the second moments of a thermal state.

This is achieved by considering states that are superpositions of Fock states that differ by three (or more) excitations ($m \geq 3$), for instance, $\sqrt{p}\,|n\rangle + \sqrt{1-p}\,|n+3\rangle$ for $0 \leq p \leq 1$. For such a state the covariance matrix takes the form of a thermal state $\Gamma = \nu\mathbb{1}$, where $\nu = \big(2pn + 2(1-p)(n+3) + 1\big)$. By selecting the discrete value $n \in \mathbb{N}_0$ and the continuous parameter p appropriately, the second moments of this state can be chosen to match those of the desired Gaussian-passive state. The thus constructed state ρ is clearly pure, and its (non-equilibrium) free energy $F(\rho) = E(\rho) - TS(\rho)$, where $S(\rho) = -\mathrm{Tr}\big[\rho \ln(\rho)\big]$ is the von Neumann entropy, is hence identical to its average energy $E(\rho)$. The latter can of course be lowered to zero by a (non-Gaussian) unitary by rotating the pure state towards the vacuum state. □

As we have seen in Lemma 1, if only the first and second moments of a state are known and the state is Gaussian-passive, in principle all (or none) of the state's energy may be extractable. In other words the gap between the free energy, i.e., the energy extractable by general unitary transformations, and the energy that can be extracted using only Gaussian unitaries is maximal. However, for such a maximal gap both the initial and final state must be pure, since we are applying only (Gaussian) unitary transformations, which leave the spectrum (and hence the entropy) unchanged. As most machines operate at an ambient temperature that is above zero and the second law implies that it is highly unlikely for any state to fall below the entropy of the corresponding thermal state, a maximal gap in the above sense may not occur in practice.

It is therefore reasonable to assume that, in addition to the first and second moments, also a lower bound on the entropy of the state is known. Given some nonzero entropy $S(\rho) = -\mathrm{Tr}\big[\rho \ln(\rho)\big]$, the average energy of the state is bounded from below and may not be lowered arbitrarily[1]. In such a case, it is of interest to ask whether the free energy gap is still maximal. That is, we ask: Does every Gaussian-passive state with entropy[2] S_0 admit a state ρ that has the same entropy, $S(\rho) = S_0$, and the same first and second moments $\langle\, \mathbb{X} \,\rangle = 0$ and Γ, but whose energy $E(\rho)$ [which is determined by $\langle\, \mathbb{X} \,\rangle$ and Γ via Eq. (12)] may be lowered to the minimal value E_0 that is compatible with S_0 using (arbitrary) unitary transformations?

Theorem 2. *The first and second moments of any Gaussian-passive state with entropy S_0 are compatible with a (non-Gaussian) state of the same entropy for which the maximal amount of energy (the energy difference to the thermal state of entropy S_0) is extractable by unitary transformations.*

Proof. For a Gaussian-passive state with fixed first and second moments ($\langle\, \mathbb{X} \,\rangle = 0$ and Γ), the energy is also fixed, see Eq. (12). In addition, we assume that the entropy of the initial (Gaussian-passive) state is S_0. Clearly, any previous Gaussian unitaries or possible general unitary transformations on the closed system that are yet to be carried out must leave this entropy invariant. On the other hand, the state ρ that minimizes the energy $E(\rho)$ at a fixed entropy S_0 is the thermal state of Eq. (4). Since we cannot change the spectrum using unitary transformations, we hence have to show that for every Gaussian-passive state at entropy S_0 there exists a state ρ that has the same spectrum as a thermal state of entropy S_0, but whose first and second moments (and hence its energy) match those of the Gaussian-passive state.

The strategy to show that this is possible is to start from the thermal state and manipulate it using unitary transformations to reach the desired first and second moments. In this way the spectrum of the state is preserved. In particular, we know that the spectrum is also invariant under the possible application of an energy-conserving beam splitting operation in the case that the covariance matrix of the Gaussian-passive state is not diagonal (clause (ii) of Theorem 1). Consequently, we can again focus on proving the statement of Theorem 2 for single-mode Gaussian-passive states with thermal covariance matrices, as we have argued in the proof of Lemma 1. For each of these local single-mode covariance matrices the diagonal elements are identical and linear functions of the energy, see Eq. (12). The first moments as well as the off-diagonals of the covariance matrix of both the thermal state and the initial state vanish. We therefore restrict to rotations in subspaces of Fock states that differ by three (or more) excitations to keep it that way.

We now just have to show that this method allows increasing the energy of the thermal state to reach the energy of any single-mode Gaussian-passive state, which also fixes the desired nonzero second moments. For any specified energy this can be achieved by continuously rotating in the subspace spanned by the states $|0\rangle$ and $|n\rangle$ for some sufficiently large $n \geq 3$. Since the thermal state is (i) diagonal in the Fock basis, (ii) the eigenvalues are strictly decreasing with increasing n, and (iii) the Hilbert space is infinite-dimensional, one may reach arbitrarily large energies at a fixed entropy. Finally, because a Gaussian-passive state with the same first and second moments (and therefore same energy), and with the same entropy S_0 as the Gaussian-passive initial state can be reached unitarily from the minimal energy thermal state, the converse must also be true. □

It is quite remarkable to note that the proof of Theorem 2 makes use of the infinite-dimensionality of the Hilbert space, which is reminiscent of the famous Hilbert

[1] We implicitly assume that the spectrum of the Hamiltonian is bounded from below, i.e., a ground state exists.

[2] Note that the entropy is not determined by the second moments alone, since the Gaussian-passive state need not be a Gaussian state.

hotel paradox (see, e.g., Ref. [31, p. 17]). The fact that the Hilbert space is infinite-dimensional is crucial to give the necessary freedom to be able to unitarily increase the energy of any thermal state to arbitrary values without introducing nonzero first moments or off-diagonal second moments. In any finite dimension this is not possible. In practice, one may encounter systems that are effectively finite-dimensional, which would place limitations on the applicability of Theorem 2. This could lead to a potential reduction of the ergotropy gap.

Nonetheless, it is interesting to observe that the infinite dimensions of the Hilbert space may even allow extending the statement of Theorem 2 to cases where more than the first two statistical moments of the Gaussian-passive state are known. For instance, suppose an expectation value of a cubic combination of mode operators such as $\langle\, a^3 \,\rangle$ was known. In this example, one could rotate in a subspace spanned by two Fock states separated by three excitations (e.g., $|k\rangle$ and $|k+3\rangle$ for some appropriate value of k) to arrange for the desired expectation value without changing the lower order moments, energy, or entropy.

V. CONCLUSION

In this article, we have investigated the fundamental thermodynamic problem of work extraction from continuous-variable quantum systems under the restriction to Gaussian unitaries. These operations can typically be easily implemented in quantum optics experiments, whereas the general unitary transformations that may be required to extract work from a given non-passive state may be extremely challenging to realize. To capture the limitations of this restricted class of operations for the task at hand we have introduced the notion of Gaussian passivity. We have given necessary and sufficient criteria for identifying Gaussian-passive states (whose energy may not be reduced by Gaussian unitaries) based on the first and second statistical moments of an arbitrary number of modes. Furthermore, we have shown that although the first two statistical moments provide complete information about the Gaussian ergotropy (the maximal amount of energy extractable in a Gaussian unitary process), the gap to the non-Gaussian ergotropy may always be maximal if the state is not fully known, even under entropy constraints.

This trade-off between usefulness and severe limitation of Gaussian operations comes as no surprise and is a recurring feature in continuous-variable quantum information. For instance, Gaussian operations are known not to be universal for computational tasks [24]. Similar properties have also been described in a quantum thermodynamical framework of converting work and correlations. There it was found that, while Gaussian operations provide optimal scaling for the creation of entanglement using large input energies, they cannot create entanglement with finite energy at arbitrary temperatures [19, 32].

While uncovering and quantifying the restrictiveness of Gaussian operations in the thermodynamic context, our results also provide practical strategies for the implementation of quantum heat engines based on Gaussian operations in quantum optical architectures. In particular, the steps P1-P4 of the proof of Theorem 1 can be viewed as a set of instructions for Gaussian work extraction.

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II.4 Precision and Work Fluctuations in Gaussian Battery Charging

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Overview: This publication introduces the idea of studying the charging precision (variance of the final energy) and work fluctuations in the context of battery charging and studies this problem for bosonic modes. As central technical outputs, the optimal (and worst-case) protocols for unitary (realized by cyclic Hamiltonian processes) battery charging of initially thermal batteries with arbitrary input energy are derived for both precision and fluctuations. Similarly, the corresponding optimal (and worst-case) protocols are derived when restricting to Gaussian unitaries.

Contribution: The idea for this research was developed jointly with Marcus Huber. I derived the protocols, performed the calculations and largely wrote the manuscript.

Precision and Work Fluctuations in Gaussian Battery Charging

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One of the most fundamental tasks in quantum thermodynamics is extracting energy from one system and subsequently storing this energy in an appropriate battery. Both of these steps, work extraction and charging, can be viewed as cyclic Hamiltonian processes acting on individual quantum systems. Interestingly, so-called passive states exist, whose energy cannot be lowered by unitary operations, but it is safe to assume that the energy of any not fully charged battery may be increased unitarily. However, unitaries raising the average energy by the same amount may differ in qualities such as their precision, fluctuations, and charging power. Moreover, some unitaries may be extremely difficult to realize in practice. It is hence of crucial importance to understand the qualities that can be expected from practically implementable transformations. Here, we consider the limitations on charging batteries when restricting to the feasibly realizable family of Gaussian unitaries. We derive optimal protocols for general unitary operations as well as for the restriction to easier implementable Gaussian unitaries. We find that practical Gaussian battery charging, while performing significantly less well than is possible in principle, still offers asymptotically vanishing relative charge variances and fluctuations.

1 Introduction

Quantum thermodynamics (QT) deals with the manipulation and transfer of energy and entropy at the quantum scale. How well one can transfer energy depends greatly on the information one has about a system [1, 2, 3]. Consequently, the system entropy quantifying this information is rendered an important quantity for achievable state transformations [4]. At fixed energy, the entropy is maximized for thermal states, which allows for the definition of thermal equilibrium characterized by the emergent notion of temperature. A system in such a thermal equilibrium with an environment at temperature T is thermodynamically useless in the sense that its energy cannot be extracted as work [5, 6]. Therefore, much effort has been invested into understanding the emergence of equilibration and thermalization in quantum systems [7]. At the same time, quantifying extractable energy and identifying achievable transformations crucially depends on the control one assumes to have about microscopic degrees of freedom. For instance, acting only upon individual quantum systems from whom work is to be extracted gives rise to the notion of passive states [8] which cannot yield any work in cyclic Hamiltonian processes, even if the entropy at a given energy is far below the thermal entropy [9]. However, even non-passive states may still require complex operations and precise control over large Hilbert spaces that make them practically unfeasible sources of work. A recent focus of thermodynamic resource theories has thus been to investigate the role of precise control and practically implementable operations for achieving desired work extraction [10, 11, 12, 13] and refrigeration [14].

The resource-theoretic view on quantum thermodynamics of course extends beyond the task of work extraction, and generally aims to identify the ultimate limitations of all single-shot processes [5, 15, 16]. More specifically, viewing quantum thermodynamics as a resource theory entails either the ability to perform any unitary operation induced by a Hamiltonian $H(t)$ with control parameter t (i.e., the case of "driven" or controlled operations), or applying arbitrary "thermal operations", i.e., global energy conserving operations on the chosen system and arbitrary many auxiliary systems. When these auxiliary systems can feature coherence w.r.t. the energy eigenbasis (i.e., if coherent "batteries" are provided), these paradigms become equivalent [17, 18]. However, neither paradigm limits the complexity of the allowed operations, requiring arbitrary coherent energy shifts in subsystems from which energy is extracted or in which energy is stored [19, 20]. This can lead to genuine quantum advantages, e.g., for the charging power of N-qubit batteries [21, 22] and for small, finite-

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dimensional systems (e.g., few-qubit registers) such full control over the quantum systems may reasonably be expected. However, for larger systems such as registers of many qubits, arbitrary global operations may be difficult to realize and call for more specialized practical solutions [23]. In particular, this applies to infinite-dimensional quantum systems such as (ensembles of) harmonic oscillators. Besides the paradigmatic two-dimensional Hilbert spaces of qubits often favoured in information-theoretic approaches to quantum mechanics, harmonic oscillators play a crucial role for the description of physical systems in quantum optics and quantum field theory. Indeed, all current realistic proposals for and implementations of quantum machines involve at least one Hilbert space corresponding to a harmonic oscillator. Examples for such systems include superconducting resonators [24, 25], modes of the electromagnetic field in a cavity [26], or vibrational modes of trapped ions [27, 28]. It is hence of conceptual significance to understand the fundamental as well as the practical limitations for thermodynamic tasks in such infinite-dimensional continuous-variable (CV) systems. In particular, full control over such systems automatically implies the ability to create coherence between energy levels with arbitrarily large separation.

In contrast, a class of operations that can typically be realized comparatively simply in quantum optical realizations of CV systems is that of Gaussian unitaries [29]. In the context of driven quantum systems these operations naturally appear as most straightforwardly implementable in the hierarchy of driving Hamiltonians since they require $H(t)$ to be at most quadratic in the system's creation and annihilation operators. Indeed, most natural interactions are appropriately described by such "bipartite terms" (usually resonant energy exchanges), whereas the creation of higher order terms is a challenge that is usually addressed only in a perturbative way. For the task of work extraction, the restriction of thermodynamic operations on CV systems to Gaussian transformations brings about the notion of *Gaussian passivity* [10], which encompasses states that are potentially non-passive, but are passive w.r.t. Gaussian transformations. Once work has been extracted, one would of course also like to put it to use, potentially at a later time. This requires the previously obtained energy to be stored and distributed. It is hence expected that practical limitations applying to work extraction —in particular, the restriction to Gaussian operations —will also be relevant for these tasks.

Following the full characterization of Gaussian passivity [10], we hence aim to quantify the limitations imposed by the restriction to Gaussian unitaries on the task of energy transfer to suitable quantum optical storage devices, i.e., charging batteries. More precisely, we consider ensembles of harmonic oscillators as batteries. These batteries are assumed to

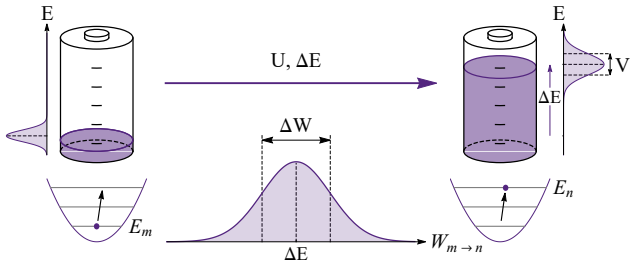

Figure 1: **Quantum battery charging:** The average energy of an initially thermal battery is unitarily increased by ΔE. The fluctuations of the final charge and of the energy supply can be quantified by the variance V of the energy distribution in the final battery, and by the average square deviation $(\Delta W)^2$ of the transitions $W_{m\to n} = E_m - E_n$ between the levels m and n from the average energy supply ΔE.

be initially uncharged in the sense that they contain no extractable work. That is, we consider the empty batteries to be in thermal equilibrium with the environment, and describe them by thermal states at the ambient temperature. We then study the task of unitarily increasing their energy by a fixed increment ΔE. Although such unitaries always exist in infinite-dimensional Hilbert spaces, unitaries achieving a given energy increase are not uniquely determined by ΔE, and may offer different charging precision, speed, and energy fluctuations during the charging process.

Here, we focus on two quantities characterizing the reliability of the charging process, as illustrated in Fig. 1. First, the charging precision, represented by the energy variance V of the battery, which is of interest since it is desirable that a charged battery is able to deliver the expected energy, not hazardously much more energy or disappointingly much less. Second, we consider the fluctuations during the charging process, captured by $(\Delta W)^2$ the average square deviation of the energy transitions from the average energy supply. While the variance quantifies the usefulness of the battery in terms of potential fluctuations occurring when discharging the loaded battery, the variance loosely speaking only captures half of the problem. That is, taking into account the initial distribution of energies one may also be interested in the energy fluctuations during the charging process. The resulting distribution is often called "fluctuating work" [30, 31, 32] and characterizes the distribution of work if one were to measure the battery in the energy eigenbasis at the beginning and end of the charging protocol. For both of these characteristics we determine the ultimate limitations during arbitrary unitary charging processes by designing optimal protocols. We then specialize to Gaussian unitaries, for which we identify the optimal and worst charging protocols. In comparison, we find that Gaussian unitaries perform significantly less well than is possible

in principle. Nonetheless, Gaussian battery charging can asymptotically achieve vanishing relative fluctuations $V/\Delta E$ and $(\Delta W)^2/\Delta E$ for large input energies by way of simple combinations of displacements and single-mode squeezing. Our results hence provide insights into both the fundamental and practical limitations of charging quantum optical batteries.

This article is structured as follows. In Sec. 2, we set the stage for the investigation and define the quantities of interest. We then present an investigation of the fundamental limitation of charging quantum batteries using arbitrary unitaries in Sec. 3, before we restrict to Gaussian transformations in Sec. 4. Finally, we draw conclusions in Sec. 5.

2 Charging a quantum battery

As battery systems to be charged we consider a number of bosonic modes (i.e., an ensemble of harmonic oscillators) initially in thermal states $\tau(\beta) = \exp(-\beta H)/\mathcal{Z}$, where $\mathcal{Z} = \mathrm{Tr}\big(\exp(-\beta H)\big)$ is the partition function, $\beta = 1/T$ is the inverse temperature of the battery (we use units where $\hbar = k_{\mathrm{B}} = 1$ throughout), and $H = \sum_j \omega_j a_j^\dagger a_j$ is the system Hamiltonian. The mode operators a_j and $a_j^\dagger$ satisfy the usual commutation relations $[a_j\,, a_k^\dagger] = \delta_{jk}$ and $[a_j\,, a_k] = 0$. For such a non-interacting Hamiltonian, the initial state is a product state $\tau(\beta) = \bigotimes_i \tau_i(\beta)$. The single-mode Gibbs states $\tau_i(\beta)$ can be written as

$$\tau_i(\beta) = (1 - e^{-\beta\omega_i}) \sum_n e^{-n\beta\omega_i} \,|n_i\rangle\langle n_i| \tag{1}$$

with respect to their respective Fock bases $\{|n_i\rangle\}$, where $a_i\,|n_i\rangle = \sqrt{n_i}\,|(n-1)_i\rangle$ and $a_i^\dagger\,|n_i\rangle = \sqrt{(n+1)_i}\,|(n+1)_i\rangle$. This choice of initial state ensures that the batteries are truly empty at first, i.e., the initial state is passive for any number of such batteries because the Gibbs state is completely passive (uniquely at fixed energy).

We are then interested in applying a unitary transformation U to raise the average energy by ΔE, transforming the initial state $\tau(\beta)$ to a final state $\rho = U\tau U^\dagger$, i.e.,

$$\begin{aligned}\Delta E &= E(\rho) - E\big(\tau(\beta)\big) = \mathrm{Tr}\big(H[\rho - \tau(\beta)]\big)\\ &= \mathrm{Tr}(H\rho) - \sum_n \frac{\omega_n}{e^{\beta\omega_n} - 1}.\end{aligned} \tag{2}$$

We quantify the charging precision via the increase of the standard deviation of the system Hamiltonian, that is, one of the quantities that we are interested in is

$$\Delta\sigma = \sqrt{V(\rho)} - \sqrt{V(\tau)}, \tag{3}$$

where the variance w.r.t. H is given by

$$V(\rho) = \big(\Delta H(\rho)\big)^2 = \mathrm{Tr}(H^2\rho) - \big(\mathrm{Tr}(H\rho)\big)^2. \tag{4}$$

Besides the precision of the final battery charge, one may also care about other quantities, for instance, the energy fluctuations[1] of the charging process. That is, we consider the average squared deviation from the average energy increase, given by

$$(\Delta W)^2 = \sum_{m,n} p_{m\to n}(W_{m\to n} - \Delta E)^2\,, \tag{5}$$

where $W_{m\to n} = E_n - E_m$ is the work relating the m-th and n-th energy levels, with $H\,|n\rangle = E_n\,|n\rangle$, and

$$p_{m\to n} = p_m\,|\langle n|\,U\,|m\rangle|^2 \tag{6}$$

is the probability of a transition from the m-th to the n-th energy eigenstate starting from the initial state $\tau(\beta)$ with diagonal elements $p_n = \langle n|\,\tau\,|n\rangle$. To better understand the quantity ΔW it is useful to note that we can write the squared work fluctuation as the variance of the operator $H_\Delta = \tilde{H} - H$ in the thermal state, where $\tilde{H} = U^\dagger H U$, i.e.,

$$\begin{aligned}(\Delta W)^2 &= \langle H_\Delta^2\rangle_\tau - \langle H_\Delta\rangle_\tau^2\\ &= (\Delta\tilde{H}_\tau)^2 + (\Delta H_\tau)^2 - 2\,\mathrm{Cov}(\tilde{H}, H)\,,\end{aligned} \tag{7}$$

where the covariance is given by

$$\mathrm{Cov}(\tilde{H}, H) = \tfrac{1}{2}\langle\{\tilde{H}, H\}_+\rangle - \langle\tilde{H}\rangle\langle H\rangle\,, \tag{8}$$

and $\{\tilde{H}, H\}_+ = \tilde{H}H + H\tilde{H}$ denotes the anticommutator. In general, the operators $\tilde{H}$ and H need not commute, but since the initial thermal state is diagonal in the energy eigenbasis, we can further simplify Eq. (7) and obtain

$$(\Delta W)^2 = V(\rho) + V(\tau) - 2\left[\mathrm{Tr}(\tilde{H}H\tau) - E(\tau)E(\rho)\right]. \tag{9}$$

The squared increase of the standard deviation, in comparison, can be written as

$$(\Delta\sigma)^2 = V(\rho) + V(\tau) - 2\sqrt{V(\rho)V(\tau)}\,. \tag{10}$$

Since one can write $E(\tau)E(\rho) = \mathrm{Tr}(U^\dagger H U\,\langle H\rangle_\tau\,\tau)$, it is easy to see the charging precision and fluctuations coincide when the initial state is an eigenstate of the Hamiltonian, because $\langle H\rangle_\tau\,\tau = E_n\,|n\rangle\langle n| = H\tau$ and $V(\tau) = 0$. In this case (which, in our scenario only occurs for the ground state since our initial state is a thermal state), one has $(\Delta W)^2 = (\Delta\sigma)^2 = V(\rho)$.

3 Fundamental Limits for Battery Charging

In this section, we investigate the fundamental limits on the charging precision and fluctuations. As

[1] Note that we use a definition for energy fluctuations suitable for the task at hand, which differs from fluctuations in the sense of thermodynamical fluctuation relations [33].

we shall see, optimal protocols can be constructed that minimize either the variance of the final energy or the fluctuations during the charging process, but these do not coincide for finite temperatures. However, the involved operations are often rather complicated in the sense that they require very specific interventions in particular subspaces of the infinite-dimensional Hilbert space, tailored to the initial temperature and energy supply. The results obtained in this section hence illustrate what is in principle possible and provide a benchmark for the precision and fluctuations achievable with Gaussian unitaries.

3.A Fundamental limits for zero temperature

Let us first consider a simple example to set the stage for a further, in-depth investigation. To this end, we consider a single-mode battery that is initially in the ground state, i.e., $H = \omega a^\dagger a$ and $\tau = |0\rangle\langle 0|$. In this case, the work fluctuations and charging precision coincide and are given by

$$(\Delta\sigma)^2 = (\Delta W)^2 = (\Delta H(\rho))^2, \tag{11}$$

and $\rho = U|0\rangle\langle 0|U^\dagger = |\psi\rangle\langle\psi|$ is a pure state. Since the Hilbert space in question is infinite-dimensional the energy variance of the final state is not bounded from above. This can be seen by choosing a superposition of the form $|\psi\rangle = \sqrt{q}|0\rangle + \sqrt{1-q}|k\rangle$ with $k = (1-q)^{-1}\Delta\epsilon$, such that $\frac{\langle H\rangle_\psi}{\omega} = \frac{\Delta E}{\omega} \equiv \Delta\epsilon$. A simple calculation then reveals that

$$\left(\tfrac{\Delta\sigma}{\omega}\right)^2 = \Delta\epsilon(k - \Delta\epsilon). \tag{12}$$

In other words, for any chosen energy ΔE one can make k (and hence $\Delta\sigma = \Delta W$) arbitrarily large by simultaneously choosing q sufficiently close to 1. So for arbitrarily small energies, the energy variance and the fluctuations during the charging process may increase by an arbitrary amount. However, it is also clear that this is an artefact of the infinite-dimensional character of the system. If the dimension d of the system is finite (or there is some cutoff energy), then the maximal variance is obtained for a superposition of the eigenstates $|0\rangle$ and $|d-1\rangle$, with minimal and maximal eigenvalues, respectively, resulting in

$$\tfrac{(\Delta\sigma_{\mathrm{max,d}})^2}{\omega^2} = \Delta\epsilon\big((d-1) - \Delta\epsilon\big)\,. \tag{13}$$

The minimal achievable variance for any given energy is obtained by unitarily rotating to a superposition of the two energy eigenstates $|n\rangle$ and $|n+1\rangle$ that are closest to the available energy, i.e., such that $n \leq \Delta\epsilon \leq n+1$. More specifically, we have $|\psi\rangle = U|0\rangle = \sqrt{p}|n\rangle + \sqrt{1-p}|n+1\rangle$ with

$$p = \lceil\Delta\epsilon\rceil - \Delta\epsilon\,, \tag{14}$$

resulting, after some algebra, in a variance of

$$\left(\tfrac{\Delta\sigma_{\min}}{\omega}\right)^2 = \big(\Delta\epsilon - \lfloor\Delta\epsilon\rfloor\big)\big(\lceil\Delta\epsilon\rceil - \Delta\epsilon\big). \tag{15}$$

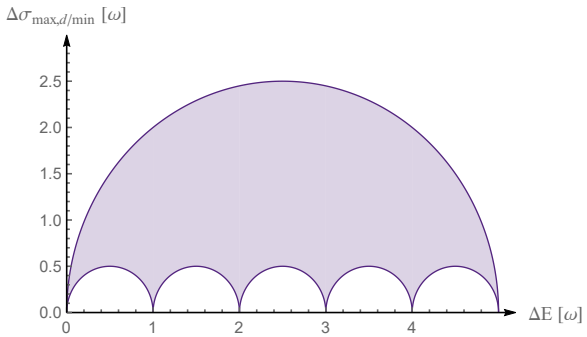

Figure 2: **Unrestricted battery charging:** The maximal and minimal variances of the energy that are in principle possible for a battery that starts in its ground state and is being charged by ΔE are shown (in units of ω, with $\hbar = 1$) for a system of dimension $d = 6$. When the Hilbert space is infinite-dimensional, the lower bound periodically repeats, but the upper bound is no longer finite for any value of ΔE. Since the initial temperature vanishes, the bounds shown also apply to the charging fluctuation ΔW.

Crucially, $\Delta\sigma_{\min} = 0$ whenever ΔE is an integer multiple of the oscillator frequency, and the maximal value of $\Delta\sigma_{\min}$ is $\frac{\omega}{2}$, as illustrated in Fig. 2.

3.B Fundamental precision limits for arbitrary temperatures

Having understood the simple case of optimally charging a battery initially in the ground state, we now want to move on to the case of thermal battery states. On the one hand, the worst-case scenario immediately carries over from the situation discussed in the previous section. That is, in an infinite-dimensional system one may always find a unitary transformation that increases the energy by an arbitrarily small amount, while increasing the variance arbitrarily strongly. This can be seen by just noting that the two-level rotation used to rotate between the ground state and the level $|k\rangle$ can also be applied to thermal states. The only difference is that the corresponding probability weights are now different from 1 and 0 initially. In contrast, the upper bound for the variance in a finite-dimensional system is always finite.

The optimally achievable charging precision, on the other hand, requires a more intricate analysis. The task at hand is to specify the state of minimal energy variance $V(\rho)$ at a fixed average energy within the unitary orbit of a thermal state at a given temperature. In general, we cannot give a closed expression relating this minimal variance to the energy input and the initial temperature. However, one may formulate a protocol that provides (one of) these minimal variance states. Here, we will give a short, intuitive description of this protocol, and provide a detailed step-by-step account in Appendix A.1.

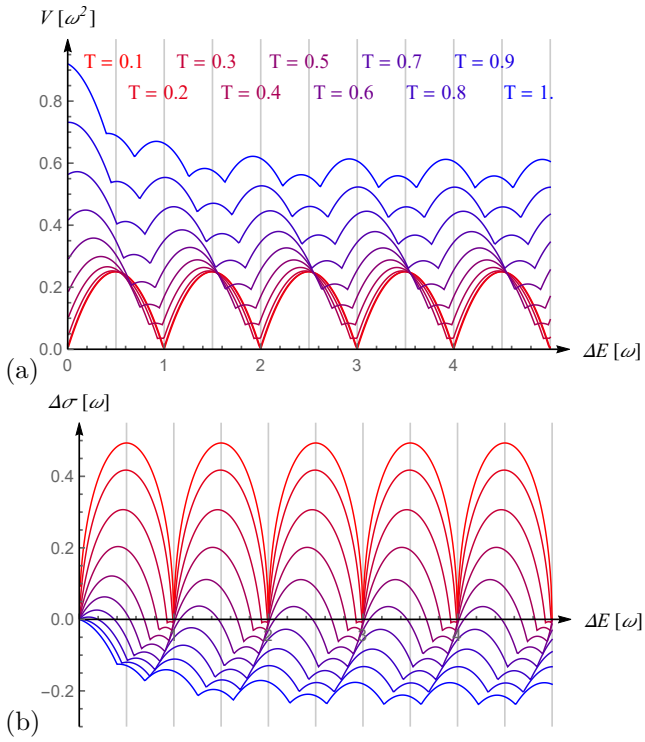

Figure 3: **Optimal precision charging of thermal battery:** The minimal variance $V(\rho)$ (in units of ω^2) and the optimal standard deviation change $\Delta\sigma$ (in units of ω) that are in principle achievable for charging a quantum battery at initial temperature $T = 0.1$ to $T = 1$ (in steps of 0.1 and units of ω, bottom to top in (a) and top to bottom in (b)) are plotted against the energy input $\Delta E/\omega$ in (a) and (b), respectively.

Let us now briefly explain the working principle of the optimal-precision charging protocol. First, recall that the initial thermal state has a density operator $\tau(\beta)$ that is diagonal in the energy eigenbasis with probability weights p_n decreasing with increasing energies E_n. The average energy $E\big(\tau(\beta)\big)$ is determined by the initial temperature and we hence know the target energy $E(\rho) = E(\tau) + \Delta E$ for any energy input. We can then naively apply two-level rotations to reorder the probability weights on the diagonal such that the largest weight p_0 is shifted to the eigenstate whose energy is closest to the target energy, the second largest weight is shifted to the second-closest eigenstate to $E(\rho)$, and so on. This procedure results in the unique state $\tilde{\rho}(\beta)$ within the unitary orbit of $\tau(\beta)$ whose average squared deviation $\tilde{V}$ from the target energy is minimal.

Unfortunately, this state does not generally have the desired target average energy, i.e., $\tilde{E} = E(\tilde{\rho}) \neq E(\rho)$. Consequently, the average squared deviation from $E(\rho)$ is generally not equal to the energy-variance, $\tilde{V} \neq V$. Moreover, both the cases $\tilde{E} > E$ and $\tilde{E} < E$ can occur and one therefore has to adjust the energy accordingly. This can be done by sequences of two-level rotations that change the energy by $\Delta\tilde{E}$ and increase the average squared deviation from E by $\Delta\tilde{V}$. An ordering of these operations that is optimal is obtained when performing them in the order of increasing values of $\Delta\tilde{V}/|\Delta\tilde{E}|$, starting with the smallest, i.e., when the increase of $\tilde{V}$ per unit energy change is as small as possible. One carries on with this protocol until the desired target energy is reached, in which case the final value of $\tilde{V}$ becomes the variance of the energy V. The resulting variances for given energy input for a harmonic oscillator are illustrated in Fig. 3. It can be seen that for higher initial temperatures this optimal protocol can lead to decreasing variances in the battery state. Also note that the working principle of this optimal protocol is unchanged if one considers a finite-dimensional system instead and differences only arise because of the finite maximal energy input at any given temperature.

3.C Fundamental precision limits for multi-mode batteries

After obtaining the fundamental limits on the precision of charging a single-mode battery, it is of course natural to ask which possibilities arise when several such batteries are available. The worst case scenario for multiple modes trivially translates from our previous analysis. Since the variance for any given energy input is not bounded from above for single-mode batteries, the same is also true for many modes.

To understand what can be achieved in the best case for multiple batteries, let us first consider the two-mode case, i.e., two batteries labelled A and B that are initially in a thermal state $\tau_A \otimes \tau_B$. We are now interested in an increase of the average energy $E(\rho) = \mathrm{Tr}\big(\rho H\big)$ w.r.t. $E(\tau)$, where the bipartite Hamiltonian is $H = H_A + H_B = \omega_A a_A^\dagger a_A + \omega_B a_B^\dagger a_B$. The energy variance is then given by

$$(\Delta H)^2 = (\Delta H_A)^2 + (\Delta H_B)^2 + 2\,\mathrm{Cov}(H_A, H_B), \tag{16}$$

where the covariance of Eq. (8) for the local (and hence commuting) operators H_A and H_B is

$$\mathrm{Cov}(H_A, H_B) = \langle H_A \otimes H_B \rangle - \langle H_A \rangle \langle H_B \rangle . \tag{17}$$

For a local unitary charging protocol, i.e., where $U = U_A \otimes U_B$, the initial thermal states remain uncorrelated and the covariance vanishes. That is, the final state ρ_{AB} is a product state $\rho_{AB} = \rho_A \otimes \rho_B$. In such a case not only the average energies but also the variances are additive. Inspection of Fig. 3 then shows that having two or more batteries available can be beneficial even when they are charged independently. For instance, when the supplied energy would lead to a local maximum of the variance if all energy is stored in one battery, it may be prudent to reduce the energy supply to this battery to reach a (local) minimum instead. The remnant energy can then be stored in a

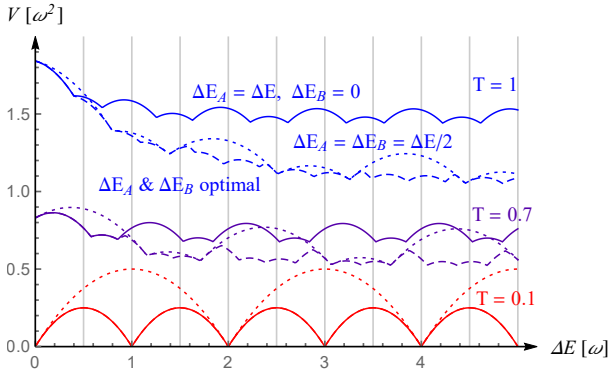

Figure 4: **Precision improvement for two-mode batteries:** The charging precision in terms of the overall variance $V(\rho)$ (in units of ω^2) is shown for a battery consisting of two modes with equal frequencies ω for sample temperatures of $T = 0.1$ (red, bottom), $T = 0.7$ (purple, middle) to $T = 1$ (blue, top) in units of ω. For each temperature, three curves are shown corresponding to different local unitary charging protocols pertaining to different distribution of the overall energy input ΔE into the energy increases ΔE_A and ΔE_B of the two modes labelled A and B, respectively. The solid curves indicate that all energy is stored in one of the modes only, $\Delta E_1 = \Delta E$, $\Delta E_2 = 0$. Dotted lines correspond to equal charging energies for both modes, $\Delta E_A = \Delta E_B = \Delta E/2$, and dashed lines represent optimally splitting the charge between both modes. For the sake of numerical optimization we have chosen integer multiples of $\omega/20$ as indivisible units of energy charge, meaning optimality here means the optimal choice of $m, n \in \mathbb{N}_0$ such that $\Delta E_A = m\frac{\omega}{20}$, $\Delta E_B = n\frac{\omega}{20}$, and $(m+n)\frac{\omega}{20} = \Delta E$. Note that for the lowest temperature shown ($T = 0.1$), the solid and dashed lines are virtually indistinguishable, meaning that there is no (distinguishable) advantage in splitting the energy between the modes. However, such an advantage is clearly visible for higher temperatures.

second battery. When the two modes have the same frequency and the initial temperature is nonzero the resulting overall variance is then smaller than or equal to that of charging only one battery, as we can see from Fig. 4. In short, the availability of several battery modes at potentially different frequencies hence provides a certain flexibility to reach local minima of the variances of the individual batteries, but the exact performance for a given set of battery modes requires to be worked out on a case-by-case basis.

For unitaries that are not local and can correlate the two batteries, the situation is even more involved but in principle such unitaries may help to achieve an even better performance. To see this, let us return to the optimal protocol of the last section. In the first step of this protocol, the probability weights of the initial thermal state are reordered to create a distribution that is as narrow as possible around the target energy. The resulting state is diagonal in the energy eigenbasis. Since this is a product basis w.r.t the tensor product structure of different modes, the state is still uncorrelated. However, in the second step, where the energy of the distribution is adjusted to the target energy, two-level rotations with optimal ratios $\Delta\tilde{V}/|\Delta\tilde{E}|$ may occur between states $|m,n\rangle$ and $|m',n'\rangle$ with $m \neq m'$ and $n \neq n'$ and hence correlate the systems. For batteries at different frequencies there can thus be an advantage in introducing (specific) correlations, whereas a situation as just described can always be avoided for batteries with equal frequencies. In the general case of arbitrary frequencies it is interesting to note though, that the creation of correlations may be marginally helpful but is not the key ingredient. This is in contrast to recent results on the charging power, where the ability to create quantum correlations, i.e., access to entangling operations (albeit not necessarily the actual creation of entanglement) can be extremely useful [21, 22].

To reach optimality it nonetheless remains to be determined how the energy can be optimally split between the oscillators, or invested in correlations. Unfortunately, this is difficult to answer in general, and is even rather complicated for uncorrelated charging due to the non-monotonic behaviour of the optimal single-mode charging protocol illustrated in Fig. 3, which is illustrated in Fig. 4. There, the specific optimal splitting depends on the initial temperature, the specific energy input, and the (number and) frequencies of the battery modes involved. The optimal performance hence has to be determined on a case-by-case basis. However, one can state quite generally that the optimal final variance of the joint system is never larger than the optimal variance when all the energy is stored only in one of the modes. In other words, having several battery modes available is never detrimental. Indeed, having more empty batteries at different frequencies at one's disposal can be considered a nontrivial resource for precise charging.

Having discussed which charging precisions can be achieved in principle, let us briefly turn to the fundamental limitations arising for the charging fluctuations.

3.D Fundamental fluctuation limits for arbitrary temperatures

To complete the investigation of the fundamental restrictions of charging a quantum battery, let us consider a protocol that minimizes the fluctuations ΔW. For simplicity, let us start with the case where the input energy is exactly one unit, $\Delta\epsilon = 1$. Then the infinite-dimensional Hilbert space allows keeping the fluctuations arbitrarily small. To achieve this, we perform a unitary permutation operation on the first N energy levels that shifts the weight $p_n = (1 - e^{-\beta\omega})e^{-n\beta\omega}$ from the level n to the level $n+1$ for $n = 0, \ldots, N-1$, while the last weight p_N is shifted to the ground state level. In the limit $N \to \infty$, the energy is increased by $\Delta E = \omega$ and since $\langle m | U | n \rangle = \delta_{m,n+1}$, the fluctuations vanish.

When the input energy is less than one unit, i.e., when $0 < \Delta\epsilon < 1$, the fluctuations do not vanish, but can be minimized in a simple way. Suppose that we perform the same permutation as before, but start shifting weights upwards at some finite $n = k$ rather than at $n = 0$, such that a vanishingly small weight is placed on the k-th level. The corresponding final state energy (in units of ω) would be

$$\tilde{\epsilon} = \epsilon_0 + \tilde{\Delta\epsilon} = \sum_{n=0}^{k-1} n\, p_n + \sum_{n=k+1}^{\infty} n\, p_{n-1} = \sum_{n=0}^{\infty} n\, p_n + \sum_{n=k}^{\infty} p_n, \tag{18}$$

where $\epsilon_0 = \sum_{n=0}^{\infty} n p_n = E(\tau)/\omega$. The increase w.r.t. the initial state would hence be

$$\tilde{\Delta\epsilon} = \sum_{n=k}^{\infty} p_n = \sum_{n=k}^{\infty} (1 - e^{-\beta\omega}) e^{-n\beta\omega} = e^{-k\beta\omega}. \tag{19}$$

Now, generally, $(\beta\omega)^{-1} \ln(1/\Delta\epsilon)$ is not an integer and, consequently, the energy shift upwards starting from $k = \tilde{k} := \lceil (\beta\omega)^{-1} \ln(1/\Delta\epsilon) \rceil$ is not enough, $\Delta\epsilon_{\mathrm{I}} := e^{-\tilde{k}\beta\omega} \leq \Delta\epsilon$. However, if we perform the shift from $\tilde{k}$ onwards nonetheless, the difference $\Delta\epsilon_{\mathrm{II}} = \Delta\epsilon - \Delta\epsilon_{\mathrm{I}}$ can be obtained by continuously rotating between the level $\tilde{k} - 1$ and the (now effectively unoccupied) level $\tilde{k}$, i.e., by a mapping

$$(p_{\tilde{k}-1}, 0) \mapsto (\cos^2\theta\, p_{\tilde{k}-1}, \sin^2\theta\, p_{\tilde{k}-1}). \tag{20}$$

The corresponding rotation angle θ is given by

$$\theta = \arcsin \sqrt{\frac{\Delta\epsilon_{\mathrm{II}}}{p_{\tilde{k}-1}}} = \arcsin \sqrt{\frac{e^{\tilde{k}\beta\omega}\Delta\epsilon - 1}{e^{\beta\omega} - 1}}. \tag{21}$$

This protocol is optimal since each (finite size) weight is shifted by either 0 or 1 units of energy, i.e., the shifts closest to $\Delta\epsilon$ since $0 \leq \Delta\epsilon \leq 1$. Explicitly, we can calculate the corresponding fluctuations by splitting the contributions for the differently shifted weights, i.e.,

$$(\Delta W)^2 = (\Delta W_{<\tilde{k}-2})^2 + (\Delta W_{\tilde{k}-1})^2 + (\Delta W_{\geq\tilde{k}})^2. \tag{22}$$

For $n = 0, \ldots, \tilde{k} - 2$ we have $p_{m\to n} = p_m \delta_{mn}$ and $W_{m\to n} = 0$ and hence $(\Delta W_{<\tilde{k}-2})^2 = \sum_{n=0}^{\tilde{k}-2} p_n (\Delta\epsilon)^2$. For the level $\tilde{k} - 1$ we have

$$\begin{aligned}\left(\frac{\Delta W_{\tilde{k}-1}}{\omega}\right)^2 &= p_{\tilde{k}-1\to\tilde{k}-1}(\Delta\epsilon)^2 + p_{\tilde{k}-1\to\tilde{k}}(1-\Delta\epsilon)^2 \\ &= p_{\tilde{k}-1}\big((\Delta\epsilon)^2 + \sin^2\theta\,[1 - 2\Delta\epsilon]\big),\end{aligned} \tag{23}$$

where we have used (20). The remaining shifts from $\tilde{k}$ upwards give rise to $(\Delta W_{\geq\tilde{k}})^2 = \sum_{n=\tilde{k}}^{\infty} p_n (1-\Delta\epsilon)^2 = \Delta\epsilon_{\mathrm{I}}(1-\Delta\epsilon)^2$. When summing up these contributions, substituting $\sin^2\theta = \Delta\epsilon_{\mathrm{II}}/p_{\tilde{k}-1}$ from Eq. (21), and noting that $\Delta\epsilon = \Delta\epsilon_{\mathrm{I}} + \Delta\epsilon_{\mathrm{II}}$, we find

$$\left(\frac{\Delta W_{\min}}{\omega}\right)^2 = \Delta\epsilon(1 - \Delta\epsilon) \tag{24}$$

for $0 \leq \Delta\epsilon \leq 1$. Finally, consider the case where $\Delta\epsilon > 1$. Then we perform the protocol just described, but replace $\Delta\epsilon$ with the difference $\Delta\epsilon - \lfloor\Delta\epsilon\rfloor$ to the lower integer value. The remaining energy is now an integer multiple of ω and can be gained by shifting the entire distribution upwards by $\lfloor\Delta\epsilon\rfloor$ units, whilst filling the gaps with vanishing contributions from arbitrarily high levels (as described for $\Delta\epsilon = 1$ at the beginning of this section). Since the last integer shift does not add any fluctuations, we arrive at the optimal value

$$\left(\frac{\Delta W_{\min}}{\omega}\right)^2 = \big(\Delta\epsilon - \lfloor\Delta\epsilon\rfloor\big)\big(\lceil\Delta\epsilon\rceil - \Delta\epsilon\big). \tag{25}$$

Note that this expression for the minimal fluctuations at arbitrary temperatures coincides with the expression for the minimal variance $(\Delta\sigma_{\min}/\omega)^2$ achievable at zero temperature, as given in Eq. (15) and illustrated (by the lower curve) in Fig. 3, but for finite temperatures the protocol minimizing the fluctuations does not minimize the variance, and vice versa. As a remark, note that in contrast to the optimal precision protocol, the protocol for minimal fluctuations does not translate directly to the finite-dimensional case.

As for the case of the variance, let us now turn to the case of several modes, starting with two. Here it is first important to note that a second battery can be added without increasing the fluctuations since for local unitaries one finds

$$(\Delta W)^2[\Delta E] = (\Delta W_A)^2[\Delta E_A] + (\Delta W_B)^2[\Delta E_B], \tag{26}$$

where $\Delta E = \Delta E_A + \Delta E_B$. Second, one may note that the protocol described above can now achieve vanishing fluctuations also for energies $\Delta E = m\omega_A + n\omega_B$ for $m, n \in \mathbb{N}_0$, not just for integer multiples of a single frequency. In addition, the optimization of the energy splitting between the two modes can lead to lower fluctuations as compared to only charging one of the batteries also for energy values that lie in between two choices of m and n. All of this can be done using only local unitary charging. Correlating unitaries again only play a minor role in the sense that they may be employed in optimizing the second part of the protocol, where the missing energy $\Delta\epsilon_{\mathrm{II}}$ is added. The presence of multiple modes as batteries to be charged can hence be considered to be helpful.

However, as before, the exact optimal protocols for multiple modes depend on the respective frequencies, temperatures, and on the input energy, and hence require case-by-case analyses. It thus becomes ever more clear that the operations to optimize either the variance or fluctuations are generally complicated and require extreme levels of control over the infinite-dimensional systems we consider here. It is hence of great interest to turn to practical operations such as Gaussian unitaries, and investigate their limitations for realistic battery charging.

4 Battery Charging Using Gaussian Unitaries

4.A Preliminaries: Phase space and Gaussian states

In the following, we want to study the restrictions imposed on the battery charging scenario when only Gaussian unitaries are used, i.e., unitary operations that map Gaussian states to Gaussian states. To examine this class of states, note that any quantum state ρ in the Hilbert space $\mathcal{L}_2(\mathbb{R}^N, dx)$, i.e., the space of square-integrable (with respect to the Lebesque measure dx) functions over $\mathbb{R}^N$, can be assigned a *Wigner function* $\mathcal{W}(x,p)$ given by

$$\mathcal{W}(x,p) = \frac{1}{(2\pi)^N} \int dy\, e^{-ipy} \langle x + \tfrac{y}{2} | \rho | x - \tfrac{y}{2} \rangle \,, \tag{27}$$

where $x, y, p \in \mathbb{R}^N$, with $x = (x_1, x_2, \ldots, x_N)^T$ and $p = (p_1, p_2, \ldots, p_N)$ are appropriate position and momentum coordinates and $\hat{x} = (\hat{x}_1, \hat{x}_2, \ldots, \hat{x}_N)^T$ and $\hat{p} = (\hat{p}_1, \hat{p}_2, \ldots, \hat{p}_N)^T$ are the corresponding position and momentum operators, respectively. The eigenstates $|x\rangle$ and $|p\rangle$ of these operators, respectively, satisfy

$$\hat{x} \,|x\rangle = x \,|x\rangle \,, \tag{28}$$

$$\hat{p} \,|p\rangle = p \,|p\rangle \,. \tag{29}$$

It is convenient to collect x and p into a single phase space coordinate $\xi = (x_1, p_1, x_2, p_2, \ldots, x_N, p_N)^T \in \mathbb{R}^{2N}$, and corresponding quadrature operators $\mathbb{X}_i$, where

$$\mathbb{X}_{2n-1} = \hat{x}_n = \tfrac{1}{\sqrt{2}}\big(a_n + a_n^\dagger\big), \tag{30}$$

$$\mathbb{X}_{2n} = \hat{p}_n = \tfrac{-i}{\sqrt{2}}\big(a_n - a_n^\dagger\big). \tag{31}$$

The commutation relation $[a_m, a_n^\dagger] = \delta_{mn}$ then implies the canonical commutator $[\hat{x}_m, \hat{p}_n] = i\delta_{mn}$, and vice versa. For the Wigner function, the normalization of the density operator translates to the condition

$$\int dx\, dp\, \mathcal{W}(x,p) = \int d\xi\, \mathcal{W}(\xi) = 1 \,. \tag{32}$$

Expectation values of Hilbert space operators $\hat{G}$ can be computed from the Wigner function via

$$\langle \hat{G} \rangle_\rho = \mathrm{Tr}\big(\hat{G}\rho\big) = \int dx\, dp\, \mathcal{W}(x,p)\, g(x,p) \,, \tag{33}$$

where the Wigner transform $g(x,p)$ of the operator $\hat{G}$ is given by

$$g(x,p) = \int dy\, e^{ipy} \langle x - \tfrac{y}{2} | \hat{G} | x + \tfrac{y}{2} \rangle \,. \tag{34}$$

With these basic definitions at hand, we can now return to Gaussian states and operations.

Gaussian states are defined as those states in $\mathcal{H}$ whose Wigner function is a multivariate Gaussian, i.e., of the form

$$\mathcal{W}(\xi) = \tfrac{1}{\pi^N \sqrt{\det(\Gamma)}} \exp\big[-(\xi - \overline{\mathbb{X}})^T \Gamma^{-1} (\xi - \overline{\mathbb{X}})\big] \,, \tag{35}$$

for some vector $\overline{\mathbb{X}} \in \mathbb{R}^{2N}$ and a real, symmetric $2N \times 2N$ matrix Γ. These quantities are called the first and second statistical moments of a quantum state, where the vector of first moments is simply $\overline{\mathbb{X}} = \langle \mathbb{X} \rangle_\rho$ and the components of the covariance matrix are given by

$$\Gamma_{ij} = \langle \mathbb{X}_i \mathbb{X}_j + \mathbb{X}_j \mathbb{X}_i \rangle - 2 \langle \mathbb{X}_i \rangle \langle \mathbb{X}_j \rangle \,. \tag{36}$$

Note that we have included a conventional factor of 2 in the definition of the covariance matrix w.r.t. the actual covariances of the operators, compare, e.g., Eq. (17). Via Eq. (35) Gaussian states are hence fully determined by $\overline{\mathbb{X}}$ and Γ.

Gaussian unitaries, which map the set of Gaussian states onto itself, are represented by affine maps $(S, \xi) : \mathbb{X} \mapsto S\mathbb{X} + \xi$. Here $\xi \in \mathbb{R}^{2N}$ are displacements in phase space represented by the unitary Weyl operators $D(\xi) = \exp\big(i\mathbb{X}^T \Omega \xi\big)$, which can shift the first moments, but leave the covariance matrix unchanged. The objects S are real, symplectic $2N \times 2N$ matrices which leave the symplectic form Ω invariant, i.e.,

$$S\, \Omega\, S^T = \Omega \,. \tag{37}$$

The components of Ω are given by $\Omega_{mn} = i\,[\mathbb{X}_m, \mathbb{X}_n] = \delta_{m,n-1} - \delta_{n,m+1}$. For more information on Gaussian operations and states see, e.g., Refs. [34, 29].

4.B Charging precision for single-mode Gaussian unitaries

We now want to study the previous situation of precisely charging quantum batteries based on harmonic oscillators under the restriction to Gaussian unitaries. To this end, first note that any initial thermal state $\tau(\beta)$ is Gaussian for all temperatures (for the usual Hamiltonian $H = \sum_n \omega_n \hat{N}_n$ with $\hat{N}_n = a_n^\dagger a_n$). The corresponding first moments vanish, $\overline{\mathbb{X}} = 0$ and the covariance matrix is diagonal,

$$\Gamma\big(\tau(\beta)\big) = \bigoplus_{n=1}^{N} \Gamma_n(\beta), \tag{38}$$

where the single-mode covariance matrices are given by $\Gamma_n(\beta) = \coth\big(\beta\omega_n/2\big)\mathbb{1}_2$. In particular, when the temperature is zero, we have the ground state with $\Gamma_{\mathrm{vac}} = \mathbb{1}$, and the corresponding Wigner function $\mathcal{W}(x,p) = \frac{1}{\pi}\exp[-(x^2 + p^2)]$.

To determine the energy of any Gaussian state for the Hamiltonian $H = \sum_n \omega_n a_n^\dagger a_n$ we do not need to use Eq. (33). Inspection of the first moments $\overline{\mathbb{X}}^{(n)} =$

$(\overline{\mathbb{X}}_{2n-1}, \overline{\mathbb{X}}_{2n})^T = \langle\, (\hat{x}_n, \hat{p}_n)^T \,\rangle$ of each mode and local covariances simply reveals[2] that

$$E(\rho) = \sum_{n=1}^{N} \omega_n \Big(\frac{1}{4}\big[\mathrm{Tr}(\Gamma_n) - 2\big] + \frac{1}{2}\|\overline{\mathbb{X}}^{(n)}\|^2 \Big)\,. \tag{39}$$

However, to compute the variance $V(\rho)$ we require also the expectation value of H^2. For our single-mode example (for notational convenience we drop the mode label n on all quantities from now on) we have $H = \omega\hat{N}$ with $\hat{N} = a^\dagger a$. We hence need to find the Wigner transform of $\hat{N}^2$. With some straightforward calculations which are shown in detail in Appendix A.2, one obtains the expression

$$N^2(x,p) = \tfrac{1}{4}\big(x^2 + p^2 - 1\big)^2 - \tfrac{1}{4}. \tag{40}$$

With this, we can compute the expectation value $\langle\, \hat{N}^2 \,\rangle$ for arbitrary single-mode Gaussian states in terms of the corresponding first and second moments. Since we are dealing with a mode-local operator, we can use the single-mode version of Eq. (35) to do so, i.e., using only the vector $\overline{\mathbb{X}} \in \mathbb{R}^2$ and the 2×2 covariance matrix Γ. After some lengthy but straightforward algebra we find that for any single-mode Gaussian state

$$\begin{aligned} \langle\, \hat{N}^2 \,\rangle &= \int dx\,dp\, \mathcal{W}(x,p) N^2(x,p) \\ &= \Big(\tfrac{1}{4}\big[\mathrm{Tr}(\Gamma) - 2\big] + \tfrac{1}{2}\|\overline{\mathbb{X}}\|^2 \Big)^2 \\ &\quad + \tfrac{1}{2}\overline{\mathbb{X}}^T \Gamma\, \overline{\mathbb{X}} + \tfrac{1}{8}\big[\mathrm{Tr}(\Gamma^2) - 2\big]\,. \end{aligned} \tag{41}$$

Since the first term on the right-hand side of Eq. (41) is just the squared expectation value of $\hat{N}$ for a Gaussian state [compare with Eq. (39)], we immediately obtain the variance

$$(\Delta\hat{N})^2 = \tfrac{1}{2}\overline{\mathbb{X}}^T \Gamma\, \overline{\mathbb{X}} + \tfrac{1}{8}\big[\mathrm{Tr}(\Gamma^2) - 2\big]. \tag{42}$$

With this knowledge at hand, we can now return to our problem of raising the energy of a single-mode battery using Gaussian unitaries. For single-mode batteries that are initially at a finite temperature, the initial energy and corresponding variance can be calculated from Eqs. (39) and (42) by noting that the corresponding first moments vanish, $\overline{\mathbb{X}} = 0$ and the covariance matrix is $\Gamma(\beta) = \coth\big(\beta\omega/2\big)\mathbb{1}_2$. With this we have

$$E\big(\tau(\beta)\big) = \tfrac{\omega}{2}\big[\coth\big(\tfrac{\beta\omega}{2}\big) - 1\big]\,, \tag{43}$$

$$V(\tau) = \big(\Delta H(\tau)\big)^2 = \tfrac{\omega^2}{4}\big[\coth^2\big(\tfrac{\beta\omega}{2}\big) - 1\big]\,. \tag{44}$$

We can then apply Gaussian unitaries to these states. For instance, we may consider single-mode displacements to raise the energy of initial thermal states. For vanishing temperature, the action of the corresponding Weyl displacement operators $D(\xi)$ on the vacuum creates coherent states $D(\xi)\,|\,0\,\rangle = |\,\alpha\,\rangle = e^{-|\alpha|^2/2}\sum_j \frac{\alpha^j}{\sqrt{j!}}\,|\,j\,\rangle$, where $\xi = \sqrt{2}(\mathrm{Re}(\alpha), \mathrm{Im}(\alpha))^T \in \mathbb{R}^2$ and $\alpha \in \mathbb{C}$. Since displacements do not alter the covariance matrix, the latter remains that of a single-mode thermal state, while the first moments are transformed to $\overline{\mathbb{X}}_i = \xi_i$. We hence have

$$\tfrac{\Delta E}{\omega} = \tfrac{1}{2}\|\overline{\mathbb{X}}\|^2 = |\alpha|^2\,, \tag{45a}$$

$$(\Delta\hat{N})^2 = \tfrac{1}{2}\coth\big(\tfrac{\beta\omega}{2}\big)\|\overline{\mathbb{X}}\|^2 + \tfrac{V(\tau)}{\omega^2}. \tag{45b}$$

For displaced thermal states we consequently find

$$\tfrac{\Delta\sigma}{\omega} = \sqrt{\coth\big(\tfrac{\beta\omega}{2}\big)\tfrac{\Delta E}{\omega} + \tfrac{V(\tau)}{\omega^2}} - \sqrt{\tfrac{V(\tau)}{\omega^2}}, \tag{46}$$

i.e., an asymptotic increase of the energy standard deviation with the square-root of the energy increase. As we shall see, pure displacements are neither optimal (minimal $\Delta\sigma$ for given ΔE), nor the worst possible Gaussian operations for battery charging, but nonetheless, make for an interesting comparison. This is illustrated in Fig. 5, where we have also included results for the optimal and worst operations, which we shall derive next.

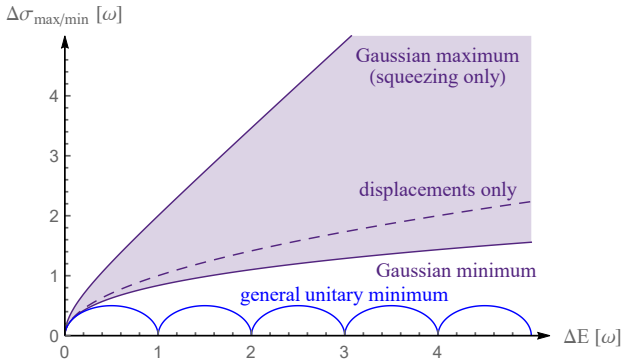

Figure 5: **Ground state battery charging:** The maximal and minimal variances of the energy that are possible using Gaussian unitaries for a battery that has been charged by ΔE and starting in its ground state are shown (in units of ω, with $\hbar = 1$). For reference, the performance of pure displacements and the lower bound for arbitrary unitaries are also shown.

4.C Optimal and worst-case Gaussian charging precision

Let us now investigate these best-case and worst-case Gaussian operations. The action of an arbitrary local Gaussian unitary U_G results in some (generally nonzero) first moments $\xi = \overline{\mathbb{X}}(U_G \tau U_G^\dagger)$, while $\Gamma(\beta)$ is mapped to the covariance matrix $\tilde{\Gamma}$ of an arbitrary single-mode Gaussian state with the same mixedness via a local symplectic operation S_{loc}, i.e., $\tilde{\Gamma} = S_{\mathrm{loc}}\Gamma S_{\mathrm{loc}}^T$. Any single-mode symplectic operation

[2] Note that there is a typographical error in the prefactor of $\|\overline{\mathbb{X}}^{(n)}\|^2$ in Ref. [10, Eq. (12)].

can be decomposed [35, 29] into (phase) rotations R and single-mode squeezing transformations $S(r)$ as

$$S_{\text{loc}} = R(\theta)\, S(r)\, R(\phi)\,, \tag{47}$$

where θ, ϕ are real rotation angles, $r \in \mathbb{R}$ is the squeezing parameter, and

$$R(\theta) = \begin{pmatrix} \cos\theta & \sin\theta \\ -\sin\theta & \cos\theta \end{pmatrix}, \quad S(r) = \begin{pmatrix} e^{-r} & 0 \\ 0 & e^{r} \end{pmatrix}. \tag{48}$$

With this, the transformed covariance matrix can be written as

$$\tilde{\Gamma} = \coth\!\left(\tfrac{\beta\omega}{2}\right) R(\theta)\, S(2r)\, R(\theta)^T\,, \tag{49}$$

and the corresponding average energy of the state $\rho = U_G \tau U_G^\dagger$ evaluates to

$$E(\rho) = \tfrac{\omega}{2}\left[\coth\!\left(\tfrac{\beta\omega}{2}\right) \cosh(2r) - 1 + \|\xi\|^2\right]. \tag{50}$$

Combining this with Eq. (43), we then have the energy input

$$\begin{aligned} \Delta E &= E(\rho) - E(\tau) \\ &= \tfrac{\omega}{2}\left[\coth\!\left(\tfrac{\beta\omega}{2}\right)\left(\cosh(2r) - 1\right) + \|\xi\|^2\right]. \end{aligned} \tag{51}$$

For the variance of the energy of the final state we first inspect the term $\xi^T \tilde{\Gamma} \xi$ and note that for our intents we can absorb the rotation $R(\theta)$ into the choice of the first moments, since $\|\xi\|^2 = \|R^T(\theta)\xi\|^2$. We hence find

$$\begin{aligned} V(\rho) = \left(\Delta H(\rho)\right)^2 = \tfrac{\omega^2}{4}\Big[&\coth^2\!\left(\tfrac{\beta\omega}{2}\right) \cosh(4r) - 1 \\ &+ 2\coth\!\left(\tfrac{\beta\omega}{2}\right) \xi^T S(2r) \xi\Big]. \end{aligned} \tag{52}$$

We then proceed in the following way. First, we note that ΔE is a function of r and $\|\xi\|^2$, whereas $V(\rho)$ depends on ξ only via the term $\xi^T S(2r)\xi = \xi_1^2 e^{-2r} + \xi_2^2 e^{2r}$. Since $e^{-2r} \leq e^{2r}$ for $r \geq 0$, the maximal and minimal values of $V(\rho)$ for fixed ΔE and $T = 1/\beta$ must be attained for combinations of $r \geq 0$ and ξ with $\xi_1 = 0$ and $\xi_2 = 0$, respectively. Conversely, this means that the remaining quantities ξ_2^2 and ξ_1^2 in Eq. (52) can be identified with $\|\xi\|^2$, i.e., from Eq. (51) we have

$$\tfrac{2\Delta E}{\omega} - \coth\!\left(\tfrac{\beta\omega}{2}\right)\!\left(\cosh(2r) - 1\right) = \begin{cases} \xi_2^2 \text{ if } V \text{ maximal}, \\ \xi_1^2 \text{ if } V \text{ minimal}. \end{cases} \tag{53}$$

We can thus define two functions $V_+(r)$ and $V_-(r)$, given by

$$\begin{aligned} \tfrac{V_\pm(r)}{\omega^2} = \tfrac{1}{4}&\left[\coth\!\left(\tfrac{\beta\omega}{2}\right)^2 \cosh(4r) - 1\right] \\ &+ e^{\pm 2r}\left[\tfrac{\Delta E}{\omega} - \tfrac{1}{2}\coth\!\left(\tfrac{\beta\omega}{2}\right)\!\left(\cosh(2r) - 1\right)\right], \end{aligned} \tag{54}$$

which correspond to the respective restrictions of the final state variance $V(\rho)$. Moreover, when the initial temperature and input energy are fixed these functions depend only on r, allowing one to straightforwardly determine the maxima $\max_r V_+(r) = \max_r V$ and minima $\min_r V_-(r) = \min_r V$, respectively. As we show in detail in Appendix A.3, for every fixed $\Delta E \geq 0$ and $T \geq 0$, there exist unique values $r_\pm \geq 0$, such that $\max_r V_+(r) = V_+(r_+) = \max_r V = V(r_+)$ and $\min_r V_-(r) = V_-(r_-) = \min_r V = V(r_-)$.

IV.C.1 Worst-case Gaussian charging precision

In particular, we find (see Appendix A.3.I for details) that for any temperature and energy input, the maximal values of $\Delta\sigma$ are obtained for $\xi = 0$, that is, when all energy is transferred to the battery via single-mode squeezing. In this case both ΔE and $\Delta\sigma$ are functions of r only, and we can use Eq. (51)to relate the two quantities directly. In other words, we find

$$r_+ = \tfrac{1}{2}\,\text{arcosh}\left(2\tfrac{\Delta E}{\omega}/\coth(\beta\omega/2) + 1\right). \tag{55}$$

along with the maximal variance increase

$$\tfrac{\Delta\sigma_{\max}}{\omega} = \sqrt{2\tfrac{\Delta E}{\omega}\left(\tfrac{\Delta E}{\omega} + \coth(\tfrac{\beta\omega}{2})\right) + \tfrac{V(\tau)}{\omega^2}} - \sqrt{\tfrac{V(\tau)}{\omega^2}}. \tag{56}$$

As we see, in this case the energy standard deviation increases linearly with the energy input in the asymptotic regime (as $\Delta E \to \infty$).

IV.C.2 Optimal Gaussian charging precision

While the worst-case Gaussian unitary transformation has thus been identified as pure single-mode squeezing, the Gaussian unitary transformation that minimizes the variance of the energy can be identified as a combination of squeezing and displacement that depends on the input energy and temperature, see Appendix A.3.II. That is, in our conventions, the optimal performance is achieved for $\xi_2 = 0$ and generally nonzero values of ξ_1 and $r = r_-$, where the latter is determined by the condition $\partial V_-/\partial r|_{r=r_-} = 0$, which implies

$$\tfrac{\Delta E}{\omega} = \tfrac{1}{2}\coth(\tfrac{\beta\omega}{2})\left(e^{2r_-}\cosh(4r_-) - 1\right). \tag{57}$$

Inserting Eq. (57) into V_- from Eq. (54) then permits us to write

$$\tfrac{V_-(r_-)}{\omega} = \tfrac{1}{2}\coth^2\!\left(\tfrac{\beta\omega}{2}\right)\left[e^{2r_-}\sinh(2r_-) + \tfrac{1}{2}\left(\cosh(4r_-) - 1\right)\right]. \tag{58}$$

Although a closed expression for $r_-(\Delta E, \beta)$ cannot be given, we show in Appendix A.3.II that $r_-(\Delta E, \beta) \geq 0$ exists and is unique and can thus easily be determined numerically for any given ΔE and $T = 1/\beta$ via the implicit formula in Eq. (57). The value r_- obtained in this way can then be inserted into Eq. (58) to arrive at the minimal variance. Results for the bounds on $\Delta\sigma$ for a initial thermal states are shown in Fig. 6 for a range of temperatures and input energies.

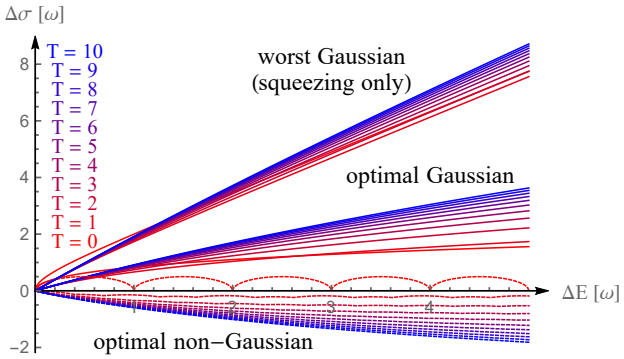

Figure 6: **Gaussian vs. optimal precision charging:** The standard deviation change $\Delta\sigma$ (in units of ω) is shown as a function of the input energy ΔE (in units of ω) for the worst-case (squeezing only, upper group of curves) and optimal (middle group of curves) single-mode Gaussian unitaries, as compared with the corresponding optimal non-Gaussian values (lower group of curves) obtained via the optimal protocol discussed in Section 3.B. Each group of curves corresponds to temperatures $T = 0$ to 10 in steps of 1 (in units of ω).

Here it is interesting to note that, while the upper bound is achieved for pure squeezing transformations, the lower bound is a combination of squeezing and displacements. In the optimal case, the energies $\Delta E_{\rm sq}$ and $\Delta E_{\rm d}$ invested into squeezing and displacement, respectively, are expressed via the optimal squeezing parameter r_- as

$$\frac{\Delta E_{\rm d}}{\omega} = \tfrac{1}{2}\xi_1^2 = \tfrac{1}{2}\coth(\tfrac{\beta\omega}{2})e^{4r_-}\sinh(2r_-)\,, \tag{59}$$

$$\frac{\Delta E_{\rm sq}}{\omega} = \frac{\Delta E - \Delta E_{\rm d}}{\omega} = \tfrac{1}{2}\coth(\tfrac{\beta\omega}{2})\big(\cosh(2r_-)-1\big)\,. \tag{60}$$

In the limit of large energy supplies, $\Delta E \to \infty$ (at fixed temperature this implies $r_- \to \infty$), we hence have $\frac{\Delta E_{\rm d}}{\Delta E_{\rm sq}} \to e^{4r_-}$. That is, the energy invested into squeezing grows much less strongly than that invested into displacements.

Moreover, note that while pure displacement asymptotically (i.e., for $\Delta E \to \infty$) leads to a linear scaling of the final variance with ΔE, that is, $V(\rho)/\Delta E \to \omega\coth(\frac{\beta\omega}{2})$ as $\Delta E \to \infty$, see Eq. (46), the optimal local strategy provides a more favourable scaling behaviour even though most of the energy is invested into displacement. More specifically, considering the relative variance $V_-/\Delta E$ by combining Eqs. (57) and (58), and taking the limit $\Delta E \to \infty$ (corresponding to $r_- \to \infty$), one finds that $V_-/\Delta E \to 0$, i.e., the optimal variance scales sub-linearly with the input energy.

4.D Charging precision for multi-mode Gaussian unitaries

Let us now finally turn to the case of bounding the charging precision of a multi-mode battery under the restriction to Gaussian unitaries. As we have discussed in Section 3.C, in a general optimal protocol for the charging precision, correlations between the individual battery systems can be helpful in principle. However, this appears to be the case only if one can selectively rotate between specific energetically desirable levels. For Gaussian unitaries, such specialized operations with, in a manner of speaking, surgical precision are out of the question. In particular, one may view any multi-mode Gaussian operation as a combination of local operations and beam splitting [35]. The latter may shift the average excitation numbers between the modes but may do little more. Another aspect of introducing correlations during the charging process is that any energy stored in this way also has to be extracted globally from the joint system if optimality is to be preserved. In other words, introducing correlations raises the effective local temperatures (and hence the local entropies), reducing the local free energy. In the spirit of restricting to practical operations, we shall hence consider only local Gaussian operations from now on.

Nonetheless, it may sometimes be useful to split the energy supply between different modes in specific ways, depending on the initial temperature and energy supply. To understand why it is useful, consider the (non-optimal) case of charging two modes labelled A and B (with frequencies ω_A and ω_B, respectively) via pure displacements. For such local operations, no correlations are introduced. If the energy is split in such a way that for some real $p \in [0,1]$ the energy $p\Delta E$ is stored in the mode A and $(1-p)\Delta E$ in the mode B, inspection of Eq. (46) reveals that the variance of the final state ρ_{AB} behaves as

$$V(\rho_{AB}) = \big(p\nu_A\omega_A + (1-p)\nu_B\omega_B\big)\Delta E + V(\tau_A) + V(\tau_B) \tag{61}$$

with $\nu_i = \coth\big(\frac{\beta\omega_i}{2}\big)$ for $i = A, B$. When the two modes have the same frequencies, $\omega_A = \omega_B$, the variance becomes independent of p, i.e., $V(\rho_{AB}) = \nu_A\omega_A\Delta E + 2V(\tau_A) = \nu_B\omega_B\Delta E + 2V(\tau_B)$, and it does not matter how the energy is split. Otherwise, that is, when $\omega_A \neq \omega_B$, it becomes beneficial to store all the energy in the lower frequency mode. Now, recall that this is the case for pure displacements, which are not optimal. The optimal strategy, in contrast, provides an increase of the variance that is sub-linear with the input energy. In this case it matters how the energy is split for all frequency combinations. E.g., for $\omega_A = \omega_B$ it becomes optimal to evenly divide the energy between the two batteries. In general, the optimal energy per battery is determined by the number and temperature of the batteries and their respective frequencies.

The worst case local scenario is obtained when the energy is used only for single-mode squeezing, where the splitting of the energy between the modes again depends on the specific situation. For instance, when

the frequencies of both modes are the same, investing the same energy in both batteries via squeezing will lead to the largest variance. This is because the local variances increase stronger than linearly with the input energy for single-mode squeezing.

4.E Charging fluctuations for single-mode Gaussian unitaries

At last, let us turn to the question of bounding the possible fluctuations that may appear in Gaussian battery charging. From Eq. (9) we already know how to express the energies and variances of ρ and τ in terms of the temperature and final first and second moments. However, we still need to calculate $\mathrm{Tr}(\tilde{H}H\tau)$ for arbitrary Gaussian unitaries, where we restrict to local operations, as before. For a single mode with frequency ω we may write the term in question as

$$\tfrac{1}{\omega^2}\mathrm{Tr}(\tilde{H}H\tau) = \sum_n p_n\, n\, \langle n\,|\, U^\dagger a^\dagger a\, U\,|\, n\rangle\ . \tag{62}$$

In principle, an arbitrary single-mode Gaussian unitary U_G may be decomposed into a combination of single-mode squeezing operations, local rotations, and displacements, none of which commute with each other. In spite of this, for any unitary and any initial state ρ_o we can find a single-mode Gaussian unitary $\tilde{U}_G = D(-\xi)U_G$ such that the first moments of $\tilde{U}_G\rho_o\tilde{U}_G^\dagger$ vanish. Conversely, this means that for the purpose of calculating $\langle n\,|\, U^\dagger a^\dagger a\, U\,|\, n\rangle$ we may assume that U_G may be written as $U_G = D(\xi)\tilde{U}_G$, where $D(\xi)$ is a pure displacement and $\tilde{U}_G$ leaves the origin of the phase space invariant. Consequently, we can use the Bloch-Messiah decomposition [35] to write $\tilde{U}_G$ as a combination of single-mode squeezing $U_\mathrm{S}(r)$ and local rotations $R(\theta)$, i.e.,

$$\tilde{U}_G = R(\theta)U_\mathrm{S}(r)R(\phi). \tag{63}$$

Inserting into Eq. (62) we then find that the rotations either act on the rotationally invariant Fock states (the phases cancel), or can be absorbed into the direction of the displacement (using the same symbol ξ in a slight abuse of notation). We hence find

$$\langle n\,|\, U_G^\dagger a^\dagger a\, U_G\,|\, n\rangle = \langle n\,|\, U_\mathrm{S}(r)^\dagger D(\xi)^\dagger a^\dagger a\, D(\xi)U_\mathrm{S}(r)\,|\, n\rangle\ .$$

We then use the simple relations

$$U_\mathrm{S}(r)^\dagger a\, U_\mathrm{S}(r) = \cosh(r)\, a + \sinh(r)\, a^\dagger, \tag{64a}$$

$$D(\xi)^\dagger a\, D(\xi) = a + \tfrac{\xi}{\sqrt{2}} \tag{64b}$$

to obtain the desired matrix element

$$\langle n\,|\, U_G^\dagger a^\dagger a\, U_G\,|\, n\rangle = n\cosh(2r) + \sinh^2(r) + \tfrac{1}{2}\|\xi\|^2. \tag{65}$$

We can then reinsert this result into Eq. (62) and evaluate the sum over n. Further inserting into the squared work fluctuations of Eq. (9), and combining this with the expressions for the variances and average energies from Eqs. (43), (44), and (51) we obtain

$$\left(\tfrac{\Delta W}{\omega}\right)^2 = \tfrac{V(\rho)}{\omega^2} + \tfrac{V(\tau)}{\omega^2} - 2\tfrac{E(\tau)}{\omega}\left(1 + \tfrac{E(\tau)}{\omega}\cosh(2r)\right), \tag{66}$$

where $V(\rho)$ is given by Eq. (52). Here, we note that, apart from $V(\rho)$, no dependency on the displacement ξ appears in Eq. (66). Consequently, we may argue as in Section 4.B, that for any given value of r, the maximal and minimal function values (here of $(\Delta W/\omega)^2$) for fixed initial temperature and fixed ΔE are attained for combinations of (single-mode) squeezing $r \geq 0$ and displacements ξ with $\xi_1 = 0$ and $\xi_2 = 0$, respectively. In other words, we are interested in determining the maximum of $\Delta W_+(r)$ and the minimum of $\Delta W_-(r)$, where

$$\left(\tfrac{\Delta W_\pm(r)}{\omega}\right)^2 = \tfrac{V_\pm(r)}{\omega^2} + \tfrac{V(\tau)}{\omega^2} - 2\tfrac{E(\tau)}{\omega}\left(1 + \tfrac{E(\tau)}{\omega}\cosh(2r)\right), \tag{67}$$

and $V_\pm(r)$ is given by Eq. (54). As we show in detail in Appendices A.3.III and A.3.IV, both extremal values exist and are unique for any given initial temperature $T = 1/\beta$ and input energy $\Delta E \geq 0$. Once again, the corresponding extremal squeezing parameters $\tilde{r}_\pm$ (which are in general different from the optimal squeezing parameters $r_\pm$ for the charging precision) are only given implicitly, i.e., by the conditions $\partial\Delta W_\pm/\partial r|_{r=\tilde{r}_\pm} = 0$, which can be expressed as

$$\begin{aligned}\tfrac{\Delta E}{\omega} = \tfrac{1}{2}\Big[&\coth(\beta\omega/2)\left(e^{\mp 2\tilde{r}_\pm}\cosh(4\tilde{r}_\pm) - 1\right) \\ &\pm \tfrac{(\coth(\beta\omega/2)-1)^2}{\coth(\beta\omega/2)}\, e^{\mp 2\tilde{r}_\pm}\sinh(2\tilde{r}_\pm)\Big],\end{aligned} \tag{68}$$

respectively. Nonetheless, $\tilde{r}_\pm$ and hence the exact optimal and worst single-mode Gaussian fluctuations can easily be obtained numerically, which we have done for some sample values shown in Fig. 7. Interestingly, the additional terms appearing in Eq. (66) besides the variances lead not only to a different optimum in terms of the relative strengths of squeezing and displacements, but also mean that the worst case is now also attained for nonzero displacements. In particular, we find that the energy $\Delta E_\mathrm{d}^\pm$ invested into displacement in the extremal cases is given by

$$\tfrac{\Delta E_\mathrm{d}^\pm}{\omega} = \tfrac{1-e^{\mp 2\tilde{r}_\pm}}{2}\left[\tfrac{(\coth(\beta\omega/2)-1)^2}{\coth(\beta\omega/2)} - \coth(\tfrac{\beta\omega}{2})e^{\mp 2\tilde{r}_\pm}\right], \tag{69}$$

while the energy invested into squeezing is $\Delta E_\mathrm{sq}^\pm = \Delta E - \Delta E_\mathrm{d}^\pm$. In the limit of large energy supplies, i.e., $\Delta E \to \infty$ (corresponding to $\tilde{r}_\pm \to \infty$ at fixed temperature), we have

$$\lim_{\tilde{r}_+\to\infty} \tfrac{\Delta E_\mathrm{d}^+}{\omega} = \frac{(\coth(\beta\omega/2)-1)^2}{4\coth(\beta\omega/2)} = \text{const.}, \tag{70}$$

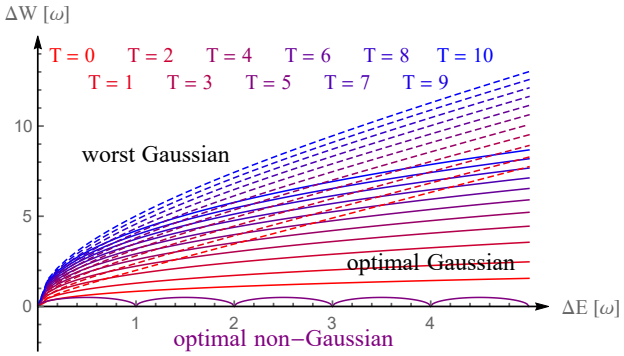

Figure 7: **Gaussian vs. optimal fluctuation charging:** The minimal (solid) and maximal (dashed) fluctuations ΔW (in units of ω) achievable with Gaussian unitaries are shown for charging quantum batteries initially at temperatures $T = 0$ (red) to $T = 10$ (blue) (in steps of 1) for given energy input ΔE (in units of ω). The periodic purple curve at the bottom indicates the minimal fluctuations that are in principle achievable, as described in Section 3.D.

while $\Delta E^{+}_{\mathrm{sq}} \to \infty$. One thus finds that the worst-case Gaussian strategy invests almost all energy into squeezing asymptotically. Conversely, for the minimal fluctuations we have $\Delta E^{-}_{\mathrm{d}}/\Delta E^{-}_{\mathrm{sq}} \to e^{-4\tilde{r}_-}$ as $\tilde{r}_- \to \infty$. In the limit of large input energies it is thus optimal to invest almost all energy into displacement to minimize the fluctuations.

The crucial feature to note is that the optimal Gaussian strategy results in a sub-linear increase of the fluctuations with the input energy. Similarly as before for the Gaussian strategy optimizing the charging precision, we can consider the limit $\Delta E \to \infty$ of the relative fluctuations $\Delta W^2_-/\Delta E$. For the optimal strategy, Eq. (68) tells us that this corresponds to the limit $\tilde{r}_- \to \infty$, for which $\Delta W^2_-/\Delta E(\tilde{r}_-) \to 0$, since ΔW^2_- and ΔE grow as $e^{4\tilde{r}_\pm}$ and $e^{6\tilde{r}_\pm}$, respectively, in this limit.

Finally, let us briefly comment on the Gaussian multi-mode scenario. Much like before for the charging precision, using Gaussian operations that generate correlations seems to be practically irrelevant since any energy stored in such global correlations could not be accessed locally. Nonetheless it can again be useful to split the energy in specific ways (depending on the respective frequencies) between two (or more) batteries, since the optimal protocol brings about a sublinear increase of $(\Delta W)^2$ with the input energy.

5 Conclusion

In this work, we have investigated fundamental and practical limitations on the precision of charging quantum batteries and on the work fluctuations occurring during the charging process. The battery systems we consider are infinite-dimensional bosonic systems, i.e., collections of harmonic oscillators, which are paradigmatic in the theoretical description of physical systems in quantum optics and quantum field theory and are hence of high conceptual significance. We assume these systems to be initially thermalized at the ambient temperature. That is, from the point of view of a resource theory of extractable work, empty batteries are considered to be for free, as no work can be extracted from them. We find that, on the one hand, neither the fluctuations nor the precision of the charge for any finite energy input are bounded from above in principle when increasing the average energy of such batteries. On the other hand, we are able to provide lower bounds for both quantities, presenting the respective optimal protocols minimizing the energy variance or fluctuations at given energies and temperatures.

In general, these optimal protocols, though theoretically easily describable, are practically difficult to realize, since they require sequences of precise interventions in particular subspaces of the corresponding infinite-dimensional Hilbert spaces. Therefore, it is interesting to understand which limitations apply in scenarios where the energy storage is performed using practically realizable transformation. A set of operations that can usually be implemented comparably simply in such systems is the family of Gaussian unitaries. Here, we have determined the optimal and worst-case Gaussian operations for charging quantum batteries. We find that energy increase via pure single-mode squeezing is the least favourable operation if one wishes to obtain a precise charge for single-mode batteries, whereas the optimal precision, as well as the smallest and largest fluctuations within the restricted set of Gaussian operations are obtained for combinations of squeezing and displacements. For multiple modes, the situation becomes more complicated in principle, but it can be said that it is in general useful to have access to multiple batteries, and that correlations between them are not necessarily detrimental, but also only helpful indirectly.

Overall, we conclude that while the optimal Gaussian operations do not achieve results comparable in quality with optimal non-Gaussian protocols, the worst performance achieved with Gaussian operations still produces finite variances and fluctuations, whereas this is not guaranteed in general. In particular, the relative variance and relative fluctuations w.r.t. the energy input asymptotically vanish for large energy supply for the optimal Gaussian charging operations. Gaussian unitaries are hence nonetheless practically useful for battery charging. In a sense, Gaussian operations hence represent a trade-off between performance versus reliability and practicality. This is reminiscent of similar contrasts between usefulness and severe limitations of Gaussian operations for tasks in quantum information, e.g., the non-universality of Gaussian operations for quantum computation [36]. Another observation of this kind can also be made in

a different quantum thermodynamical context, where Gaussian operations achieve optimal scaling for the entanglement creation for large energy inputs, but fail to create entanglement in thermal states of finite temperatures when the energy input is too small [37, 38].

This work hence adds to recent efforts [10] of understanding the usefulness of Gaussian operations for quantum thermodynamical tasks, providing investigations of Gaussian unitary work extraction and energy increase. Nonetheless, future work may expand on a number of open questions. For instance, we have here mostly focused on individual batteries since any work stored in joint systems would also require joint extraction. In other words, the role of correlations for work fluctuations and charging precision may be of interest, in particular, in relation to recent results on the work-cost of creating correlations [37, 39, 40, 41, 42]. In addition, it would also be of interest to consider the consequences of restricting to Gaussian operations for the charging speed (or charging power) as considered in Ref. [21, 22]. Finally, we note that, while some of the results presented here (e.g., the optimal precision charging protocol) directly translate to finite-dimensional systems, other aspects of this work are applicable only to the infinite-dimensional case. An in-depth investigation of the fundamental and practical limitations of precision and fluctuations in charging finite-dimensional systems, although certainly of interest [23], goes beyond the scope of this paper.

Acknowledgments

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Appendix

A.1 Optimal precision charging protocol

In this appendix, we give a detailed description of a unitary battery charging protocol that raises the average energy of an initial single-mode thermal state whilst keeping the energy variance of the final state minimal. The initial thermal state with density operator $\tau(\beta)$ is diagonal in the energy basis. The corresponding diagonal elements are the probability weights $p_n = (1-e^{-\beta\omega})e^{-n\beta\omega}$, which are decreasing with increasing energies $E_n = n\omega$. The initial average energy $E\big(\tau(\beta)\big) = \epsilon_o\omega$, where $\epsilon_o = e^{-\beta\omega}(1-e^{-\beta\omega})^{-1}$ and the initial energy variance $V\big(\tau(\beta)\big) = \omega^2 e^{-\beta\omega}(1-e^{-\beta\omega})^{-2}$ are determined by the initial temperature $T = 1/\beta$. We are then interested in increasing the average energy by an amount $\Delta E = \omega\Delta\epsilon$ to reach a state ρ with $E(\rho) = \omega\epsilon = E(\tau) + \Delta E$. In particular, we aim to achieve this increase unitarily, i.e., such that $\rho = U\tau U^\dagger$. Moreover, we want to keep the energy variance of ρ minimal. In other words, we would like to determine the (non-unique) minimal energy-variance state ρ with given average energy $\omega\epsilon$ in the unitary orbit of $\tau(\beta)$.

The protocol that we present here to obtain such a state consists of two parts (I & II). Each of these parts can be described as a series of (unitary) two-level rotations, ensuring that the final state is within the unitary orbit of the initial state. The two-level rotations between pairs of energy levels are used to appropriately shift and reorder the probability weights p_n of the initial state. As we shall explain, part I of the protocol reaches the unique state $\tilde{\rho}$ in the unitary orbit of $\tau(\beta)$ that minimizes the average squared distance to the target energy. The state $\tilde{\rho}$ is diagonal in the energy eigenbasis, and arises from a permutation of the weights p_n that assigns positions with increasing distance to the target energy to weights with decreasing size. However, the state obtained in this way does not have the desired target energy, i.e., in general $E(\tilde{\rho}) \neq E(\rho)$. During part II of the protocol, this deviation of the average energy is corrected in such a way that the target energy is reached whilst only minimally increasing the average squared distance to it.

A.1.I Part I of the protocol

In part I, we first identify the energy level (labelled k) that is closest to the desired target energy, i.e., we define

$$k = \begin{cases} \lfloor\epsilon\rfloor & \text{if } \epsilon - \lfloor\epsilon\rfloor < \lceil\epsilon\rceil - \epsilon \\ \lceil\epsilon\rceil & \text{if } \epsilon - \lfloor\epsilon\rfloor \geq \lceil\epsilon\rceil - \epsilon \end{cases}, \tag{A.1}$$

where we distinguish between two cases, depending on whether ϵ is closer to the energy level above or below its value. The probability weights for the case where $k = \lfloor\epsilon\rfloor$ are illustrated in Fig. A.1 (a). Part I of the protocol then consists of a reordering of the weights p_n such that the largest weight p_0 is moved to the energy level k, the second largest weight p_1 to the second closest level to ϵ, and so forth. After part I, the density operator is still diagonal, but the probability weights on the diagonal are now either given by

$$\tilde{p}_n = \begin{cases} p_{2(k-n)} & \text{for } n = 0,\dots,k \\ p_{2(n-k)-1} & \text{for } n = k+1,\dots,\max\{1,2k\} \\ p_n & \text{for } n \geq \max\{2,2k+1\} \end{cases}, \tag{A.2}$$

if $k = \lfloor\epsilon\rfloor$, or, in case that $k = \lceil\epsilon\rceil$ by

$$\tilde{p}_n = \begin{cases} p_{2(k-n)-1} & \text{for } n = 0,\dots,k-1 \\ p_{2(n-k)} & \text{for } n = k,\dots,2k-1 \\ p_n & \text{for } n \geq 2k \end{cases}. \tag{A.3}$$

The resulting probability distribution, illustrated in Fig. A.1 (b) for the case $k = \lfloor \epsilon \rfloor$, has an average energy $\tilde{\epsilon}_{\mathrm{I}} = \sum_n \tilde{p}_n n$ and its average squared distance from the target ϵ is minimal, i.e., we have arrived at the unique state $\tilde{\rho}$ in the unitary orbit of the initial state τ that minimizes $\tilde{V}_{\mathrm{I}} = \sum_n \tilde{p}_n (n-\epsilon)^2$. However, as $\tilde{\epsilon}_{\mathrm{I}}$ in general does not match ϵ, which implies that $\tilde{V}_{\mathrm{I}}$ also is not equal to the energy variance, we are not yet done. Interestingly, for both $k = \lfloor \epsilon \rfloor$ and $k = \lceil \epsilon \rceil$ one may encounter combinations of T and $\Delta\epsilon$ such that $\tilde{\epsilon}_{\mathrm{I}} < \epsilon$ or $\tilde{\epsilon}_{\mathrm{I}} > \epsilon$.

A.1.II Part II of the protocol

In part II of the protocol we hence have to appropriately adjust the average energy. This can again be done by a sequence of two-level rotations. Each of this transformations will bring the average energy closer to ϵ, but since we start from a minimum of the average squared deviation from ϵ, the value of the latter will increase. We are hence interested in selecting the optimal sequence of these two-level rotations. To start, consider a rotation between the levels m and n with weights $\tilde{p}_m$ and $\tilde{p}_n$ by an angle θ. This corresponds to the map

$$(\tilde{p}_m, \tilde{p}_n) \mapsto (\cos^2\theta\, \tilde{p}_m + \sin^2\theta\, \tilde{p}_n, \cos^2\theta\, \tilde{p}_n + \sin^2\theta\, \tilde{p}_m), \tag{A.4}$$

and leads to a change in energy given by

$$\Delta\tilde{\epsilon} = \sin^2\theta\, (\tilde{p}_n - \tilde{p}_m)(m-n). \tag{A.5}$$

Meanwhile, the increase of the mean squared deviation from ϵ can be written as

$$\Delta\tilde{V} = \omega^2 \sin^2\theta\, (\tilde{p}_n - \tilde{p}_m)\big((m-\epsilon)^2 - (n-\epsilon)^2\big). \tag{A.6}$$

Now, let us pick two such values on either side of ϵ, i.e., $m = k - l$ and $n = k + l + j$, where $l \in \mathbb{N}_0$ determines the distance from the level k, and $j \in \mathbb{Z}$ quantifies the difference in distances to k (or equivalently to ϵ) between the levels m and n. More specifically, we have

$$d(n,\epsilon) - d(m,\epsilon) = (n-\epsilon) - (\epsilon - m) = 2(k-\epsilon) + j. \tag{A.7}$$

Moreover, this implies that the energy change from Eq. (A.5) is given by

$$\Delta\tilde{\epsilon} = \sin^2\theta\, (\tilde{p}_n - \tilde{p}_m)(2l + j). \tag{A.8}$$

With this we further find that the changes of the energy and of $\tilde{V}$ have the relation

$$\frac{1}{\omega^2}\frac{\Delta\tilde{V}}{\Delta\tilde{\epsilon}} = 2(k-\epsilon) + j. \tag{A.9}$$

With this knowledge, we come to a more detailed description of the protocol. First, we set $\tilde{\epsilon} = \tilde{\epsilon}_{\mathrm{I}}$ and

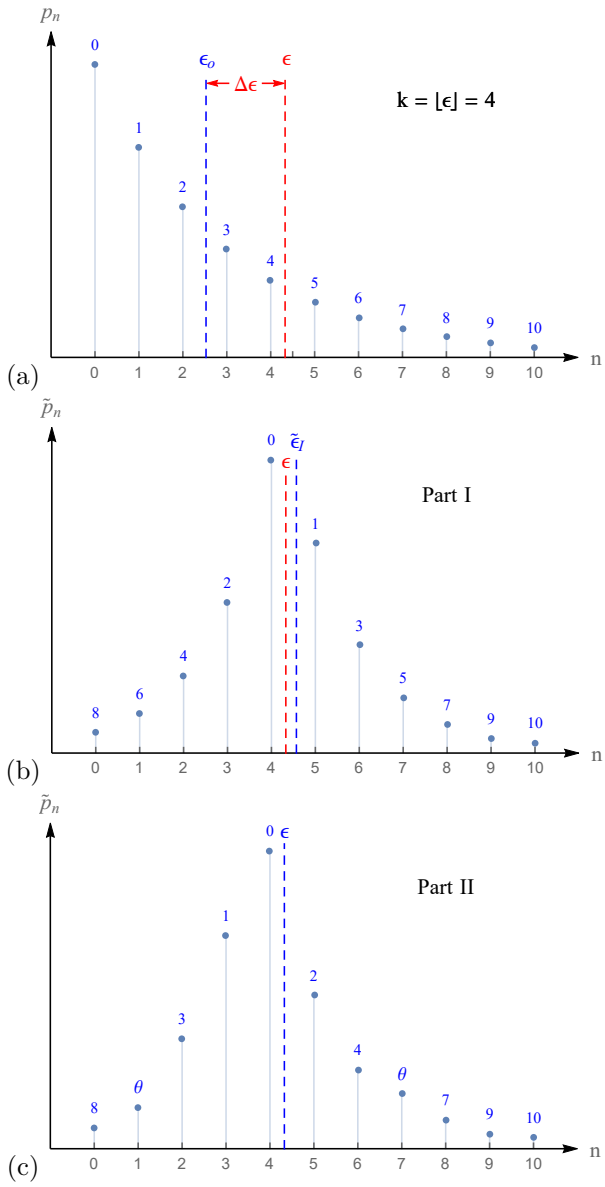

Figure A.1: **Optimal precision battery charging:** The protocol for optimal precision battery charging is illustrated for an initial thermal state of temperature $T = 3$ (in units of ω). (a) The probability weights p_n of the initial state decrease with increasing energy. The initial average energy ϵ_o (we use the dimensionless variables here for simplicity) is to be raised by $\Delta\epsilon$ to a value ϵ, that is closer to the energy level $k = \lfloor \epsilon \rfloor = 4$ rather than $\lceil \epsilon \rceil = 5$. (b) Part I: After rearranging the probability weights to place the largest weights closest to k, one obtains a distribution $\{\tilde{p}_n\}$ with an average energy of $\tilde{\epsilon}_{\mathrm{I}} > \epsilon$. The numbers $m = 0, \ldots, 10$ above the vertical lines at horizontal position n indicate that the corresponding weight $\tilde{p}_n$ corresponds to the value of the original weight p_m. (c) Part II: Additional two-level rotations adjust the energy to the target ϵ. The first two of these rotations (corresponding to $\varphi = 1$ with $j = 0$ and $l = 1, 2$, see Sec. A.1.II) have angles $\frac{\pi}{2}$ and hence completely exchange the populations of the levels $(m,n) = (3,5)$ and $(2,6)$. The third rotation between levels 1 and 7 requires only a smaller angle $0 < \theta < \frac{\pi}{2}$ until reaching the target energy.

$\tilde{V} = \tilde{V}_{\mathrm{I}}$, and we distinguish between the two situations $\tilde{\epsilon} < \epsilon$ and $\tilde{\epsilon} > \epsilon$. On the one hand, when $\tilde{\epsilon} < \epsilon$, we need to increase the energy, which means picking levels m and n such that $\tilde{p}_m > \tilde{p}_n$ and $2l + j > 0$, whilst choosing j as small as possible to minimize $\frac{\Delta\tilde{V}}{\Delta\tilde{\epsilon}}$. On the other hand, when $\tilde{\epsilon} > \epsilon$, we need to decrease the energy, suggesting that one should select levels m and n such that $\tilde{p}_m < \tilde{p}_n$ and $2l+j > 0$, whilst choosing j as large as possible to minimize $\frac{\Delta\tilde{V}}{\Delta\tilde{\epsilon}}$. When such levels are chosen and the potential energy change exceeds what is needed to reach the target, one appropriately fixes the rotation angle θ such that $\tilde{\epsilon} + \Delta\tilde{\epsilon} = \epsilon$. If the energy change achievable with a specific such rotation is not sufficient to reach the target, one rotates by $\theta = \frac{\pi}{2}$, updates $\tilde{\epsilon}$, $\tilde{V}$, and $\{\tilde{p}_n\}$ and continues with the next viable pair of levels minimizing $\frac{\Delta\tilde{V}}{\Delta\tilde{\epsilon}}$. Inspection of all cases then reveals that the (first phase) of part II consists of k or $k+1$ two-level rotations labelled by $l = l_{\min}, \ldots, k$, where

$$l_{\min} = \begin{cases} 0 & \text{if } \; k = \lfloor\epsilon\rfloor, \tilde{\epsilon} < \epsilon \\ 1 & \text{otherwise} \end{cases}, \tag{A.10}$$

and for each of these rotations we choose

$$j = \begin{cases} +1 & \text{if } \; k = \lfloor\epsilon\rfloor, \tilde{\epsilon} < \epsilon \\ 0 & \text{if } \; k = \lfloor\epsilon\rfloor, \tilde{\epsilon} > \epsilon \\ 0 & \text{if } \; k = \lceil\epsilon\rceil, \tilde{\epsilon} < \epsilon \\ -1 & \text{if } \; k = \lceil\epsilon\rceil, \tilde{\epsilon} > \epsilon \end{cases}. \tag{A.11}$$

Since $\frac{\Delta\tilde{V}}{\Delta\tilde{\epsilon}}$ does not depend on l and the rotations all commute (they pertain to different subspaces), the order of these operations within the first phase is irrelevant. However, not even all k (or $k+1$) rotations may generally be enough to sufficiently adjust the average energy. Consequently, part II may consist of an arbitrary number of phases labelled by $\varphi = 1, 2, \ldots$, where

$$\big(j(\varphi), l_{\min}(\varphi)\big) = \begin{cases} (\varphi, -\lceil\frac{\varphi}{2}\rceil + 1) & \text{if } k = \lfloor\epsilon\rfloor, \tilde{\epsilon} < \epsilon \\ (-\varphi + 1, \lceil\frac{\varphi}{2}\rceil) & \text{if } k = \lfloor\epsilon\rfloor, \tilde{\epsilon} > \epsilon \\ (\varphi - 1, -\lfloor\frac{\varphi}{2}\rfloor + 1) & \text{if } k = \lceil\epsilon\rceil, \tilde{\epsilon} < \epsilon \\ (-\varphi, \lfloor\frac{\varphi}{2}\rfloor + 1) & \text{if } k = \lceil\epsilon\rceil, \tilde{\epsilon} > \epsilon \end{cases}. \tag{A.12}$$

Let us now give a more compact description of part II. After part I, set $\{\tilde{p}\}$ as the initial distribution, and further set $\tilde{\epsilon} = \tilde{\epsilon}_{\mathrm{I}}$, $\tilde{V} = \tilde{V}_{\mathrm{I}}$, and $\varphi = 1$. Then perform the following steps:

(i) Set $j = j(\varphi)$, and $l = l_{\min}(\varphi)$.

(ii) If $\tilde{\epsilon} \neq \epsilon$ and $l \leq k$, set $m = k - l$, $n = k + l + j$, $\Delta\tilde{\epsilon}^{\mathrm{II}}_{\max} = (\tilde{p}_m - \tilde{p}_n)(2l + j)$, and continue with step (iii). If $\tilde{\epsilon} \neq \epsilon$ and $l > k$, increase φ by one, i.e., $\varphi \mapsto \varphi + 1$ and start again with step (i). If $\tilde{\epsilon} = \epsilon$ the protocol concludes.

(iii) If $\tilde{\epsilon} < \epsilon$, then $\Delta\tilde{\epsilon}^{\mathrm{II}}_{\max} > 0$ and θ_l is set to the value

$$\theta_l = \begin{cases} \frac{\pi}{2} & \text{if } \; \tilde{\epsilon} + \Delta\tilde{\epsilon}^{\mathrm{II}}_{\max} < \epsilon \\ \arcsin\sqrt{\frac{\epsilon - \tilde{\epsilon}}{\Delta\tilde{\epsilon}^{\mathrm{II}}_{\max}}} & \text{if } \; \tilde{\epsilon} + \Delta\tilde{\epsilon}^{\mathrm{II}}_{\max} \geq \epsilon \end{cases}. \tag{A.13}$$

If $\tilde{\epsilon} > \epsilon$, then $\Delta\tilde{\epsilon}^{\mathrm{II}}_{\max} < 0$ and θ_l is set to the value

$$\theta_l = \begin{cases} \frac{\pi}{2} & \text{if } \; \tilde{\epsilon} + \Delta\tilde{\epsilon}^{\mathrm{II}}_{\max} > \epsilon \\ \arcsin\sqrt{\frac{\epsilon - \tilde{\epsilon}}{\Delta\tilde{\epsilon}^{\mathrm{II}}_{\max}}} & \text{if } \; \tilde{\epsilon} + \Delta\tilde{\epsilon}^{\mathrm{II}}_{\max} \leq \epsilon \end{cases}. \tag{A.14}$$

Then continue with step (iv).

(iv) Perform the following updates:

$$\begin{aligned} \tilde{p}_m &\mapsto \cos^2\theta_l\, \tilde{p}_m + \sin^2\theta_l\, \tilde{p}_n, \\ \tilde{p}_n &\mapsto \cos^2\theta_l\, \tilde{p}_n + \sin^2\theta_l\, \tilde{p}_m, \\ \tilde{\epsilon} &\mapsto \tilde{\epsilon} + \sin^2\theta_l\, \Delta\tilde{\epsilon}^{\mathrm{II}}_{\max} = \Delta\tilde{\epsilon}^{\mathrm{II}}, \\ \tilde{V} &\mapsto \tilde{V} + \Delta\tilde{\epsilon}^{\mathrm{II}}(2(k - \epsilon) + j), \end{aligned}$$

Finally, increase l by one and start over from step (ii).

After the conclusion of part II, the target energy has been reached, $\tilde{\epsilon} = \epsilon$, and the average squared deviation from ϵ hence becomes the energy variance. The second part of the protocol is illustrated in Fig. A.1 (c) and the variances resulting from the protocol for different temperatures and input energies are shown in Fig. 3 of the main text.

A.2 Wigner representation of squared number operator

We do this by using the formulas of Eqs. (33) and (35). To this end, we start by rewriting $\hat{N}^2$ in terms of the local position and momentum operators as

$$\begin{aligned} \hat{N}^2 &= \tfrac{1}{4}\big(\hat{x}^2 + \hat{p}^2 - 1\big)^2 \\ &= \tfrac{1}{4}\big(\hat{x}^4 + \hat{p}^4 + \hat{x}^2\hat{p}^2 + \hat{p}^2\hat{x}^2 - 2(\hat{x}^2 + \hat{p}^2) - 1\big). \end{aligned} \tag{A.16}$$

We then insert term by term into Eq. (34) and calculate

$$\begin{aligned} &\langle\, x - \tfrac{y}{2} \,|\, f(\hat{x}) \,|\, x + \tfrac{y}{2} \,\rangle \\ &\quad = \langle\, x - \tfrac{y}{2} \,|\, f(x + \tfrac{y}{2}) \,|\, x + \tfrac{y}{2} \,\rangle \\ &\quad = f(x + \tfrac{y}{2})\, \langle\, x - \tfrac{y}{2} \,|\, x + \tfrac{y}{2} \,\rangle \\ &\quad = f(x + \tfrac{y}{2})\, \delta(y), \end{aligned} \tag{A.17}$$

$$\begin{aligned}
&\langle x - \tfrac{y}{2} \,|\, f(\hat{p}) \,|\, x + \tfrac{y}{2} \rangle \\
&\quad = \langle x - \tfrac{y}{2} \,|\, f(\hat{p}) \int dp' \,|\, p' \rangle \langle p' \,|\, x + \tfrac{y}{2} \rangle \\
&\quad = \int dp' \, f(p') \, \langle x - \tfrac{y}{2} \,|\, p' \rangle \langle p' \,|\, x + \tfrac{y}{2} \rangle \\
&\quad = \tfrac{1}{2\pi} \int dp' \, f(p') \, e^{-ip'y}, \qquad \text{(A.18)}
\end{aligned}$$

for functions f of the operators $\hat{x}$ and $\hat{p}$, where we have used that

$$\langle x \,|\, x' \rangle = \tfrac{1}{2\pi} \int dp \, e^{ip(x-x')} = \delta(x - x'), \qquad \text{(A.19)}$$

$$\langle x \,|\, p \rangle = \tfrac{1}{(2\pi)^{1/2}} \, e^{ipx}. \qquad \text{(A.20)}$$

After some algebra we then find the phase space representation of the operator $\hat{N}^2$ to be given by

$$\begin{aligned}
N^2(x,p) &= \tfrac{1}{4}\big(x^2 + p^2 - 1\big)^2 \\
&\quad + \tfrac{1}{16\pi} \int d\tilde{q} d\tilde{p} \tilde{q}^2 \tilde{p}^2 e^{i(p-\tilde{p})\tilde{q}}. \qquad \text{(A.21)}
\end{aligned}$$

The second term on the right-hand side can be understood in the distributional sense. That is, defining the distribution $\gamma[g(p)]$ via the function $\gamma(p)$ given by

$$\gamma(p) := \int dq \, q^2 \, e^{-2ipq} \qquad \text{(A.22)}$$

one finds that for any Schwartz function $g(p)$ we have

$$\begin{aligned}
\gamma[g(p)] &= \int dp \gamma(p) \, g(p) = -\tfrac{\pi}{4} \, \tfrac{\partial^2}{\partial p^2} g(p) \Big|_{p=0} \\
&= -\tfrac{\pi}{4} \, g''(0). \qquad \text{(A.23)}
\end{aligned}$$

Then note that the wave function $\psi(x) = \langle x \,|\, \psi \rangle$ of every single-mode pure state $|\psi\rangle$ can be expanded in terms of the Hermite polynomials $H_j(x)$ as

$$\psi(x) = \tfrac{1}{\pi^{1/4}} e^{-x^2/2} \sum_j \tfrac{c_j}{\sqrt{2^j j!}} H_j(x). \qquad \text{(A.24)}$$

with $\sum_j |c_j|^2 = 1$. Using Eq. (A.19) we can then write the Wigner function for an arbitrary single-mode pure state as

$$\mathcal{W}(x,p) = \tfrac{1}{\pi^{3/2}} e^{-x^2} \int dy \, e^{-y^2 - 2ipy} \, h(x,y) \qquad \text{(A.25)}$$

with the function

$$h(x,y) = \sum_{j,k} \tfrac{c_j c_k^*}{\sqrt{2^{j+k} j! k!}} H_j(x+y) H_k^*(x-y).$$

Finally, we can compute the integral of $\mathcal{W}(x,p)$ with the second term on the right-hand side of Eq. (A.21) and find

$$\begin{aligned}
&\tfrac{1}{16\pi} \int dx \, dp \, \mathcal{W}(x,p) \int d\tilde{q} \, d\tilde{p} \, \tilde{q}^2 \tilde{p}^2 e^{i(p-\tilde{p})\tilde{q}} \\
&= \tfrac{1}{16\pi^{5/2}} \int dx e^{-x^2} \int dy e^{-y^2} h(x,y) \\
&\quad \times \int d\tilde{p} \tilde{p}^2 \int d\tilde{q} \tilde{q}^2 e^{-i\tilde{p}\tilde{q}} \int dp e^{ip(\tilde{q}-2y)} \\
&= -\tfrac{1}{8\pi^{1/2}} \int dx e^{-x^2} \, \tfrac{\partial^2}{\partial y^2} \big(y^2 e^{-y^2} h(x,y)\big) \Big|_{y=0}.
\end{aligned}$$

where we have integrate over p using Eq. (A.19), followed by an integral over the delta function $\delta(\tilde{q} - 2y)$, and finally made use of Eqs. (A.22) and (A.23). It is then easy to see that only the term $2h(x,0)$ remains after taking the derivatives and evaluating at $y = 0$. Using the normalization of the wave function in Eq. (A.24) and arrive at

$$\tfrac{1}{4\pi^{1/2}} \int dx e^{-x^2} h(x,0) = \tfrac{1}{4} \int dx \, \psi(x) \psi^*(x) = \tfrac{1}{4}.$$

Due to the linearity of the Wigner function in ρ, this computation extends from $|\psi\rangle$ to arbitrary single-mode mixed states, and since the number operator of each mode is a local observable also to arbitrary N-mode states. We can hence rewrite Eq. (A.21) as

$$N^2(x,p) = \tfrac{1}{4}\big(x^2 + p^2 - 1\big)^2 - \tfrac{1}{4}. \qquad \text{(A.26)}$$

A.3 Extremal Gaussian precision and fluctuations

In this appendix, we give detailed proofs for the existence and uniqueness of the extremal values of the charging precision and fluctuations when restricting to single-mode Gaussian unitaries at fixed initial temperature and energy input. The corresponding results are presented and discussed in Sections 4.C and 4.E.

A.3.I Maximal variance

We begin with the maximally possible variance that single-mode Gaussian unitaries allow for, i.e., the worst-case scenario. Here, one is interested in determining the maximum of the function $V_+(r)$ from Eq. (54) over all $r \geq 0$. For brevity, we will (again) use the notation $\Delta\epsilon = \Delta E/\omega$ and $\nu = \coth\big(\frac{\beta\omega}{2}\big)$, as well as define $\mathcal{V}_+ := 4V_+/\omega^2$, such that the function that we wish to maximize can be written as

$$\begin{aligned}
\mathcal{V}_+ &= \nu^2 \cosh(4r) - 1 + 2\nu e^{2r} \big(2\Delta\epsilon - \nu\big[\cosh(2r) - 1\big]\big) \\
&= -\nu^2 \big[\sinh(4r) - 2e^{2r}(2\tfrac{\Delta\epsilon}{\nu} + 1)\big] - (\nu^2 + 1). \qquad \text{(A.27)}
\end{aligned}$$

To determine the extremal points, we calculate the first and second partial derivatives of $\mathcal{V}_+$ w.r.t. r, i.e.,

$$\frac{\partial \mathcal{V}_+}{\partial r} = -4\nu^2 \big[\cosh(4r) - (2\tfrac{\Delta\epsilon}{\nu} + 1) e^{2r}\big], \qquad \text{(A.28)}$$

$$\frac{\partial^2 \mathcal{V}_+}{\partial r^2} = -8\nu^2 \big[2\sinh(4r) - (2\tfrac{\Delta\epsilon}{\nu} + 1) e^{2r}\big]. \qquad \text{(A.29)}$$

The condition $\frac{\partial \mathcal{V}_+}{\partial r}|_{r=r_{\text{extr.}}} = 0$ at the extremal points $r = r_{\text{extr.}}$ yields

$$(2\tfrac{\Delta\epsilon}{\nu} + 1) = e^{-2r_{\text{extr.}}} \cosh(4r_{\text{extr.}}) \,. \tag{A.30}$$

Now note that the left-hand side is greater or equal than 1, while the function $f(r) = e^{-2r}\cosh(4r)$ appearing on the right-hand side satisfies $f(r=0) = 1$ and has a minimum at $r = \ln(3)/8 > 0$ (as can be seen by setting $\partial f(r)/\partial r = 0$), and $f(r) \overset{r\to\infty}{\longrightarrow} \infty$. Consequently, Eq. (A.30) has two solutions $r^{\pm}_{\text{extr.}}$, with $r^{+}_{\text{extr.}} > \ln(3)/8 > 0$ and $r^{-}_{\text{extr.}} < 0$. Inserting $(2\frac{\Delta\epsilon}{\nu}+1)$ from Eq. (A.30) back into the second partial derivative gives

$$\left.\frac{\partial^2 \mathcal{V}_+}{\partial r^2}\right|_{r=r_{\text{extr.}}} = -4\nu^2\big[e^{4r_{\text{extr.}}} - 3e^{-4r_{\text{extr.}}}\big], \tag{A.31}$$

which is negative for $r^{+}_{\text{extr.}}$ (which is $> \ln(3)/8$) and positive for $r^{-}_{\text{extr.}} < 0$. We thus have a local maximum at $r = r^{+}_{\text{extr.}}$, while $r^{-}_{\text{extr.}}$ is a local minimum. Then, recall that we are interested in non-negative solutions (a negative squeezing parameter would reverse the roles of ξ_1 and ξ_2 in our treatment, see Section 4.C), and can thus discard $r^{-}_{\text{extr.}}$. Moreover, this eliminates all values of $\mathcal{V}_+(r)$ for $r < r^{-}_{\text{extr.}}$, which can become larger than $\mathcal{V}_+(r^{+}_{\text{extr.}})$. In other words, in the relevant range $r \geq 0$, a (global) maximum of $\mathcal{V}_+(r)$ can be found at $r^{+}_{\text{extr.}}$. Nevertheless, this is not the sought-after maximum for the variance, as we shall explain next.

If $r^{+}_{\text{extr.}} > 0$, implicitly determined by Eq. (A.30), were the correct solution, we could express the total input energy as

$$\Delta\epsilon = \tfrac{\nu}{2}\big(e^{-2r^{+}_{\text{extr.}}} \cosh(4r^{+}_{\text{extr.}}) - 1\big) \,. \tag{A.32}$$

Since the energy (in units of ω) invested into squeezing as a function of the squeezing parameter is given by $\Delta\epsilon_{\text{sq}} = \frac{\nu}{2}\big(\cosh(2r) - 1\big)$, see Eq. (51), we could then write the energy invested into squeezing for $r = r^{+}_{\text{extr.}}$ as

$$\begin{aligned}\big(\Delta\epsilon_{\text{d}}\big)_{r=r^{+}_{\text{extr.}}} &= \big(\Delta\epsilon - \Delta\epsilon_{\text{sq}}\big)_{r=r^{+}_{\text{extr.}}} \\ &= \tfrac{\nu}{4}\big(e^{-6r^{+}_{\text{extr.}}} - e^{-2r^{+}_{\text{extr.}}}\big) < 0.\end{aligned} \tag{A.33}$$

In other words, the (local) maximum of the function $\mathcal{V}_+(r)$ at $r = r^{+}_{\text{extr.}}$ is not physically realizable, since we must require $\Delta\epsilon_{\text{d}} \geq 0$. Put simply, the maximum at $r = r^{+}_{\text{extr.}}$ would require more energy to be invested into squeezing than is available overall, $\Delta\epsilon_{\text{sq}} > \Delta\epsilon$. Since $\Delta\epsilon_{\text{sq}}$ is strictly increasing with increasing squeezing parameter, we are hence looking for a solution for some $r = r_+ < r^{+}_{\text{extr.}}$ that maximizes $\mathcal{V}_+$ within the physically allowed range. Our previous analysis informs us that such a solution exists uniquely. The function $\mathcal{V}_+(r)$ has one local minimum at a negative argument, and one local maximum for $r > 0$, and must hence be strictly increasing between r and $r^{+}_{\text{extr.}}$. The solution we are looking for is thus unique and found for the maximal value $r = r_+$ allowed by the global energy constraint, that is

$$\Delta\epsilon_{\text{sq}} = \Delta\epsilon = \tfrac{\nu}{2}\big(\cosh(2r_+) - 1\big). \tag{A.34}$$

Expressing r_+ as a function of $\Delta\epsilon$ and ν then yields the result presented in Eq. (55), i.e., the worst precision for Gaussian single-mode unitaries is achieved when all energy is invested into single-mode squeezing.

A.3.II Minimal variance

Next we are interested in determining the optimal strategy using Gaussian single-mode unitaries. To this end, we similarly define $\mathcal{V}_- := 4V_-/\omega^2$ with V_- as in Eq. (54), that is, we have to minimize

$$\begin{aligned}\mathcal{V}_- &= \nu^2\cosh(4r) - 1 + 2\nu e^{-2r}\big(2\Delta\epsilon - \nu\big[\cosh(2r) - 1\big]\big) \\ &= \nu^2\big[\sinh(4r) + 2e^{-2r}(2\tfrac{\Delta\epsilon}{\nu} + 1)\big] - (\nu^2 + 1).\end{aligned} \tag{A.35}$$

The partial derivatives w.r.t. to r yield

$$\frac{\partial \mathcal{V}_-}{\partial r} = 4\nu^2\big[\cosh(4r) - (2\tfrac{\Delta\epsilon}{\nu} + 1)e^{-2r}\big], \tag{A.36}$$

$$\frac{\partial^2 \mathcal{V}_-}{\partial r^2} = 8\nu^2\big[2\sinh(4r) + (2\tfrac{\Delta\epsilon}{\nu} + 1)e^{-2r}\big]. \tag{A.37}$$

The extremal condition $\frac{\partial \mathcal{V}_-}{\partial r}|_{r=r_{\text{extr.}}} = 0$ then yields

$$(2\tfrac{\Delta\epsilon}{\nu} + 1) = e^{2r_{\text{extr.}}} \cosh(4r_{\text{extr.}}) \,, \tag{A.38}$$

which has a unique solution $r_{\text{extr.}} = r_-$ for $r_{\text{extr.}} \geq 0$ since the function $e^{2r}\cosh(4r)$ is greater or equal than 1 and is strictly increasing for $r \geq 0$. Therefore, we have one and only one solution $r_- \geq 0$ for every value of $(2\frac{\Delta\epsilon}{\nu}+1)$. Moreover, inserting into the second derivative gives

$$\left.\frac{\partial^2 \mathcal{V}_-}{\partial r^2}\right|_{r=r_-} = 4\nu^2\big[3e^{4r_-} - e^{-4r_-}\big], \tag{A.39}$$

which is positive for $r_- > -\ln(3)/8$ and hence in particular when $r_- > 0$. Inserting $r_{\text{extr.}} = r_-$ into the condition of Eq. (A.38) and expressing $\Delta\epsilon$ one thus arrives at the result of Eq. (57), i.e.,

$$\Delta\epsilon = \tfrac{\nu}{2}\big(e^{2r_-}\cosh(4r_-) - 1\big) \,. \tag{A.40}$$

Moreover, for the minimum we find that the energy input splits into squeezing and displacement according to

$$\Delta\epsilon_{\text{sq}} = \tfrac{\nu}{2}\big(\cosh(2r_-) - 1\big) \,, \tag{A.41}$$

$$\Delta\epsilon_{\text{d}} = \Delta\epsilon - \Delta\epsilon_{\text{sq}} = \tfrac{\nu}{2}e^{4r_-}\sinh(2r_-) \geq 0 \,, \tag{A.42}$$

such that, unlike the maximum at $r^{+}_{\text{extr.}}$ discussed before, the desired minimum can be physically realized for all $\Delta\epsilon$ and ν.

A.3.III Maximal fluctuations

Let us now determine the maximal fluctuations that are possible during a charging process at fixed input energy via single-mode Gaussian unitaries. That is, we are interested in finding the maximum value of $(\Delta W_+(r)/\omega)^2$ from Eq. (67) over all r for fixed $\Delta\epsilon$ and ν. To simplify this task, we note that this is equivalent to the maximization problem for the function $\mathcal{W}_+(r)$, given by

$$\mathcal{W}_+(r) = \tfrac{1}{4}\mathcal{V}_+ - \tfrac{1}{2}(\nu-1)^2\cosh(2r), \tag{A.43}$$

which, up to terms independent of r, corresponds to $(\Delta W_+(r)/\omega)^2$ from Eq. (67). The first two partial derivatives w.r.t. r are

$$\begin{aligned}\frac{\partial\mathcal{W}_+}{\partial r} &= \frac{1}{4}\frac{\partial\mathcal{V}_+}{\partial r} - (\nu-1)^2\sinh(2r) \qquad (A.44)\\ &= \nu^2\Big[\big(2\tfrac{\Delta\epsilon}{\nu}+1\big)e^{2r} - \cosh(4r) - \big(\tfrac{\nu-1}{\nu}\big)^2\sinh(2r)\Big],\end{aligned}$$

$$\begin{aligned}\frac{\partial^2\mathcal{W}_+}{\partial r^2} &= \frac{1}{4}\frac{\partial^2\mathcal{V}_+}{\partial r^2} - 2(\nu-1)^2\cosh(2r) \qquad (A.45)\\ &= 2\nu^2\Big[\big(2\tfrac{\Delta\epsilon}{\nu}+1\big)e^{2r} - 2\sinh(4r) - \big(\tfrac{\nu-1}{\nu}\big)^2\cosh(2r)\Big],\end{aligned}$$

where we have inserted for the partial derivatives of $\mathcal{V}_+$ from Eqs. (A.28) and (A.29). For the purpose of solving the maximization problem, we introduce the notation $\chi := \big(2\frac{\Delta\epsilon}{\nu}+1\big) \geq 1$ and $\lambda := (\nu-1)^2/\nu^2$, with $0 \leq \lambda \leq 1$ since $\nu \geq 1$, such that the extremal condition $\big(\partial\mathcal{W}_+/\partial r\big)_{r=\tilde{r}_+} = 0$ at the extremal point $r = \tilde{r}_+$ reads

$$\lambda\tfrac{1}{2}(1-e^{-4\tilde{r}_+}) + \tfrac{1}{2}\big(e^{2\tilde{r}_+} + e^{-6\tilde{r}_+}\big) = \chi. \tag{A.46}$$

We then define $u := e^{-2r}$ along with a family of functions $f_\lambda(u)$ via

$$f_\lambda(u) := \lambda\tfrac{1}{2}\big(1-u^2\big) + \tfrac{1}{2}\big(\tfrac{1}{u}+u^3\big). \tag{A.47}$$

The maximization problem for $\mathcal{W}_+(r)$ can thus be formulated as the question: Does there exist a $u = u_\chi$, with $0 < u_\chi \leq 1$, for every pair of λ and χ, such that $f_\lambda(u = u_\chi) = \chi$? To answer this question, we first determine the extremal point u_λ of f_λ, i.e., such that

$$\left.\frac{\partial f_\lambda(u)}{\partial u}\right|_{u=u_\lambda} = -\lambda u_\lambda - \tfrac{1}{2}\big(\tfrac{1}{u_\lambda^2} - 3u_\lambda^2\big) = 0, \tag{A.48}$$

which implies $g(u_\lambda) := \frac{1}{2}\big(3u_\lambda - \frac{1}{u_\lambda^3}\big) = \lambda$ for $u_\lambda > 0$. Since $g(u_\lambda)$ is a continuous, strictly increasing function of u_λ that can take the values $g(u_\lambda = 3^{-1/4}) = 0$ and $g(u_\lambda = 1) = 1$, there is exactly one u_λ that satisfies $g(u_\lambda) = \lambda$ for any λ between 0 and 1, suggesting that $f_\lambda(u)$ always has a unique local extremal point within the interval $]0,1[$. Inspection of the second partial derivative, i.e.,

$$\begin{aligned}\left.\frac{\partial^2 f_\lambda(u)}{\partial u^2}\right|_{u=u_\lambda} &= \Big(-\lambda - \tfrac{1}{u_\lambda^3} + 3u_\lambda\Big)_{u=u_\lambda} \qquad (A.49)\\ &= \tfrac{1}{2}\big(3u_\lambda - \tfrac{1}{u_\lambda^3}\big) = \lambda \geq 0,\end{aligned}$$

then reveals that the local extremum is a local minimum. Moreover, since $f_\lambda(u=1) = 1$, this means that the minimal value is below one, $f_\lambda(u_\lambda) < 1$. In contrast, we have $f_\lambda(u) \overset{u\to 0}{\longrightarrow} \infty$, suggesting that $f_\lambda(u)$ is strictly decreasing on the interval $[0, u_\lambda[$ and can there take any value between $f_\lambda(u_\lambda) < 1$ and ∞ (in particular, any value χ). We have thus found that there is a unique $u_\chi < u_\lambda$ for every λ and χ such that $f_\lambda(u_\chi) = \chi$.

In other words, Eq. (A.46) has a unique solution $\tilde{r}_+$ for every valid λ and χ. To check that this solution is a maximum, we calculate

$$\begin{aligned}\left.\frac{\partial^2\mathcal{W}_+}{\partial r^2}\right|_{r=\tilde{r}_+} &= 2\nu^2\big[\chi e^{2\tilde{r}_+} - 2\sinh(4\tilde{r}_+) - \lambda\cosh(2\tilde{r}_+)\big]\\ &= 2\nu^2\big[\tfrac{\chi}{u_\chi} - \tfrac{1}{u_\chi^2} + u_\chi^2 - \tfrac{\lambda}{2}\big(\tfrac{1}{u_\chi} + u_\chi\big)\big], \qquad (A.50)\end{aligned}$$

where we have substituted $e^{-2\tilde{r}_+} = u_\chi$. Further inserting for $\chi = f_\lambda(u_\chi)$ from Eq. (A.47), and comparing with Eq. (A.48), we arrive at

$$\begin{aligned}\left.\frac{\partial^2\mathcal{W}_+}{\partial r^2}\right|_{r=\tilde{r}_+} &= 2\nu^2\Big[-\lambda u_\chi - \tfrac{1}{2}\big(\frac{1}{u_\chi^2} - 3u_\chi^2\big)\Big]\\ &= 2\nu^2\left.\frac{\partial f_\lambda(u)}{\partial u}\right|_{u=u_\chi} < 0, \qquad (A.51)\end{aligned}$$

which is negative since $u_\chi < u_\lambda$ is below the minimum u_λ of $f_\lambda(u)$. Consequently, the extremal value of $\mathcal{W}_+$ is a maximum, which we have thus shown exists and is unique for any fixed λ and χ corresponding to fixed values of ΔE and T.

A.3.IV Minimal fluctuations

Similarly, we now wish to determine the minimal fluctuations that are possible during a single-mode Gaussian unitary charging process at fixed input energy, i.e., we want to minimize $(\Delta W_-(r)/\omega)^2$ from Eq. (67) over all r for fixed $\Delta\epsilon$ and ν. As in the previous section, we simplify the problem by considering the equivalent minimization of the function $\mathcal{W}_-(r)$, given by

$$\mathcal{W}_-(r) = \tfrac{1}{4}\mathcal{V}_- - \tfrac{1}{2}(\nu-1)^2\cosh(2r). \tag{A.52}$$

Up to terms independent of r, this function corresponds to $(\Delta W_-(r)/\omega)^2$ from Eq. (67). The first two partial derivatives w.r.t. r are then

$$\begin{aligned}\frac{\partial\mathcal{W}_-}{\partial r} &= \frac{1}{4}\frac{\partial\mathcal{V}_-}{\partial r} - (\nu-1)^2\sinh(2r) \qquad (A.53)\\ &= \nu^2\Big[-\big(2\tfrac{\Delta\epsilon}{\nu}+1\big)e^{-2r} + \cosh(4r) - \big(\tfrac{\nu-1}{\nu}\big)^2\sinh(2r)\Big],\end{aligned}$$

$$\begin{aligned}\frac{\partial^2\mathcal{W}_-}{\partial r^2} &= \frac{1}{4}\frac{\partial^2\mathcal{V}_-}{\partial r^2} - 2(\nu-1)^2\cosh(2r) \qquad (A.54)\\ &= 2\nu^2\Big[\big(2\tfrac{\Delta\epsilon}{\nu}+1\big)e^{-2r} + 2\sinh(4r) - \big(\tfrac{\nu-1}{\nu}\big)^2\cosh(2r)\Big],\end{aligned}$$

where we have inserted for the partial derivatives of $\mathcal{V}_-$ from Eqs. (A.36) and (A.37). We then proceed

as in Appendix A.3.III, and formulate the extremal condition $\left(\partial\mathcal{W}_-/\partial r\right)_{r=\tilde{r}_+} = 0$ at the extremal point $r = \tilde{r}_-$ in terms of the constants $\chi := \left(2\frac{\Delta\epsilon}{\nu} + 1\right) \geq 1$ and $\lambda := (\nu - 1)^2/\nu^2$, with $0 \leq \lambda \leq 1$, as

$$\lambda\tfrac{1}{2}(1 - e^{4\tilde{r}_-}) + \tfrac{1}{2}\left(e^{-2\tilde{r}_-} + e^{6\tilde{r}_-}\right) = \chi. \tag{A.55}$$

To verify, that this condition can be met for all λ and χ, we again define a new variable $v := e^{-2r}$ with $0 \leq v \leq 1$ for $r \geq 0$, and define a family of functions $h_\lambda(v)$ via

$$h_\lambda(v) := \lambda\tfrac{1}{2}\left(1 - \tfrac{1}{v^2}\right) + \tfrac{1}{2}\left(v + \tfrac{1}{v^3}\right). \tag{A.56}$$

The minimization problem for $\mathcal{W}_-(r)$ can thus be formulated as: Does there exist a $v = v_\chi$, with $0 < v_\chi \leq 1$, for every pair of λ and χ, such that $h_\lambda(v = v_\chi) = \chi$? To provide an answer, we start again by determining if h_λ has any extremal points in the allowed range of v. At such an extremal point $v = v_\lambda$ we would have

$$\left.\frac{\partial h_\lambda(v)}{\partial v}\right|_{v=v_\lambda} = \lambda\tfrac{1}{v_\lambda^3} + \tfrac{1}{2}\left(1 - 3\tfrac{1}{v_\lambda^4}\right) = 0, \tag{A.57}$$

which would imply $\lambda = \frac{1}{2}\left(\frac{3}{v_\lambda} - v_\lambda^3\right) =: \tilde{g}(v_\lambda)$. However, the function $\tilde{g}(v_\lambda)$ is strictly decreasing for $v_\lambda \in [0, 1]$, with the minimal value $\tilde{g}(v_\lambda = 1) = 1$. Therefore, $h_\lambda(v)$ has no local minima (or maxima) on the open interval $]0, 1[$. Moreover, $h_\lambda(v)$ diverges as $v \to 0$, and takes its minimum within the allowed range of v for $v = 1$, i.e., $h_\lambda(v = 1) = 1$. We have thus shown that $h_\lambda(v)$ is a strictly decreasing function of $v \in [0, 1]$ that can take any value between 1 and ∞. There is thus a unique value v_χ such that $h_\lambda(v = v_\chi) = \chi$ for every χ and λ.

Finally, we check that we have indeed found a minimum of $\mathcal{W}_-$ by evaluating the second partial derivative at $r = \tilde{r}_-$ (corresponding to $v = v_\chi$), i.e.,

$$\begin{aligned}\left.\frac{\partial^2\mathcal{W}_-}{\partial r^2}\right|_{r=\tilde{r}_-} &= 2\nu^2\left[\chi e^{-2\tilde{r}_-} + 2\sinh(4\tilde{r}_-) - \lambda\cosh(2\tilde{r}_-)\right] \\ &= 2\nu^2\left[\chi v_\chi + \tfrac{1}{v_\chi^2} - v_\chi^2 - \tfrac{\lambda}{2}\left(\tfrac{1}{v_\chi} + v_\chi\right)\right], \end{aligned} \tag{A.58}$$

where we have substituted $e^{-2\tilde{r}_-} = v_\chi$. Inserting for $\chi = h_\lambda(v_\chi)$ from Eq. (A.56), and comparing with Eq. (A.57), we obtain

$$\begin{aligned}\left.\frac{\partial^2\mathcal{W}_-}{\partial r^2}\right|_{r=\tilde{r}_-} &= 2\nu^2\left[-\lambda\tfrac{1}{v_\chi} - \tfrac{1}{2}\left(v_\chi^2 - 3\frac{1}{v_\chi^2}\right)\right] \\ &= 2\nu^2 v_\chi^2 \left.\frac{\partial h_\lambda(v)}{\partial v}\right|_{v=v_\chi} \geq 0, \end{aligned} \tag{A.59}$$

which is nonnegative since $h_\lambda(v)$ is strictly decreasing for $0 \leq v \leq 1$, and hence has a negative first derivative on this interval. We can therefore finally conclude that the extremal value of $\mathcal{W}_-$ is a minimum that exists and is unique for any fixed λ and χ corresponding to fixed values of ΔE and T.

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II.5 Thermodynamically Optimal Creation of Correlations

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Thermodynamically optimal creation of correlations

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Overview: In this work, we consider the problem of optimal conversion of work into correlations via unitary (realized by cyclic Hamiltonian processes) operations on pairs of systems in initial thermal states. For symmetric situations where both systems have the same Hamiltonian, this question is tied to the existence of symmetrically thermalizing unitaries (STUs). We develop a variety of approaches for investigating the problem of the existence of STUs for arbitrary local dimensions and show how (and why) most of them successively fail. However, as a main result, we use what we informally term the 'geometric approach' to show that STUs exist in local dimensions $d = 3$ and $d = 4$ for arbitrary symmetric Hamiltonians. For asymmetric Hamiltonians, we show that STUs do not exist in general, but they are also not necessarily tied to the problem of optimal conversion of work into correlations in this case.

Contribution: I helped to develop the ideas, discussed them with my coauthors and supervised the overall project. I proved the results for $d = 3$ and $d = 4$ for the geometric method and helped writing the manuscript.

Thermodynamically optimal creation of correlations

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Correlations lie at the heart of almost all scientific predictions. It is therefore of interest to ask whether there exist general limitations to the amount of correlations that can be created at a finite amount of invested energy. Within quantum thermodynamics such limitations can be derived from first principles. In particular, it can be shown that establishing correlations between initially uncorrelated systems in a thermal background has an energetic cost. This cost, which depends on the system dimension and the details of the energy-level structure, can be bounded from below but whether these bounds are achievable is an open question. Here, we put forward a framework for studying the process of optimally correlating identical (thermal) quantum systems. The framework is based on decompositions into subspaces that each support only states with diagonal (classical) marginals. Using methods from stochastic majorisation theory, we show that the creation of correlations at minimal energy cost is possible for all pairs of three- and four-dimensional quantum systems. For higher dimensions we provide sufficient conditions for the existence of such optimally correlating operations, which we conjecture to exist in all dimensions.

I. INTRODUCTION

Correlations can be regarded as the fundamental means of obtaining information: From a physical perspective, obtaining knowledge requires correlation of physical variables in an observed system with physical variables in a measurement apparatus [1]. As long as one regards system and measurement apparatus as separate entities which are not persistently strongly interacting [2], this correlation requires an investment of energy. Conversely, all correlations imply extractable work [3]. Since correlations and energy (work) can be considered to be resources in (quantum) information theory and thermodynamics, respectively, these observations establish one of the fundamental connections between these theories. Indeed, both theories already share a common framework using statistical ensembles to capture knowledge about collections of physical systems. This knowledge can then be harnessed to facilitate the most efficient use of the available resources, e.g., energy, for the tasks at hand. The young field of quantum thermodynamics combines features from both fields and investigates their interplay in the quantum domain [4–6].

Here, we explore the specific quantitative relation between correlations and energy [7]. While general qualitative insights can help to understand some quandaries arising from Maxwell's demon or Szilard's engine [8–10], and correlations play various interesting roles in quantum thermodynamics (see, e.g., [2, 3, 11–18]), precise quantitative statements about the trade-off between work and correlations are generally complicated. For example, by allowing arbitrarily slow quasi-static operations and perfect control over arbitrarily many auxiliary systems, one may provide tight lower bounds on the work cost of creating bipartite correlations as measured by the mutual information [7, 11, 12, 19, 20]. However, for two identical systems, these bounds are tight only in the case when specific so-called symmetrically thermalizing unitaries (STUs) exist, i.e., unitaries that map initial thermal states to locally thermal states at higher local effective temperatures. Moreover, how well two systems can be correlated for a given energy in finite time and with limited control is generally not known. In particular, the possibility of optimal conversion of energy into correlations for the interesting special case of fully controlled, closed systems with two identical subsystems rests entirely on the assumed existence of STUs. In this sense, central open questions at the very foundations of quantum thermodynamics concern the quantitative correspondence between correlations and energy.

In this paper, we put forward a framework for investigating potential marginal spectra in the unitary orbit of any quantum state via decompositions into locally classical subspaces (LCSs) and majorisation relations. Here, LCSs are subspaces of a bipartite Hilbert space that support only states that are locally classical, i.e., states which have diagonal marginals with respect to a chosen basis. We use this approach to investigate the question of existence of STUs for arbitrary bipartite quantum systems. In particular, we show that STUs exist for two identical copies of arbitrary three- and four-dimensional systems, thereby extending previous results on the special case of Hamiltonians with equally-spaced energy gaps [11, 20]. In addition, we formulate sufficient conditions for the existence of STUs for identical subsystems with arbitrary local dimension and for multiple copies of the initial state. Indeed, we show that STUs exist in the limit of infinitely many copies.

This paper is structured as follows: In Sec. II, we describe the conceptual framework of our investigation, formulate our central question, and briefly review the state of the art. In Sec. II.1.1, we further discuss that, while STUs do not in general exist for asymmetric situations where the two systems have different Hamiltonians, this does not resolve the problem

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of maximizing correlations for asymmetric systems. We then focus on the open case of two identical subsystems in Sec. II.2 and discuss a family of unitary transformations that result in symmetric, diagonal marginals. These unitaries have a block-diagonal structure with respect to a specific choice of LCSs, and give rise to a rich structure of potential marginal transformations. In particular, they allow showing that STUs exist in the asymptotic limit of infinitely many copies. In Sec. III, we then formulate three alternative approaches to demonstrating the existence of STUs based on unitaries in these LCSs. As we show, all three approaches allow proving the existence of STUs in local dimension $d = 3$. However, only two of these approaches are successful (to different extent) in local dimension $d = 4$ and provide general sufficient conditions for the existence of STUs for higher dimensions. Finally, in Sec. IV we provide a summary and discussion and put forward a hypothesis for general dimensions.

II. FRAMEWORK

II.1. Main question

Let us start our investigation with a detailed description of the problem: We consider the process of correlating two previously uncorrelated quantum systems. More precisely, we study the controlled interaction of two quantum systems A and B with Hamiltonians H_A and H_B, respectively, which are initially in a tensor product state $\varrho_A \otimes \varrho_B$, with the goal of increasing the correlations between them. As we want to obtain optimal bounds, we consider optimal coherently controlled systems, i.e. the ability to externally engineer and time any interaction Hamiltonian between the two quantum systems, resulting in unitary dynamics on the system and we thus study the global unitary orbit of product states. Furthermore, we are interested in the energy needed to establish correlations between these initially uncorrelated quantum systems.

One could generalise the question from perfect system control and thus unitaries on the system, to correlating maps generated by unitaries on the system and arbitrary auxiliary systems, inducing completely positive and trace preserving (CPTP) maps on the system. However, some transformations (e.g., cooling to the ground state) may only be achievable asymptotically (i.e., using infinite time or energy [21–24]). Moreover, the implementation of CPTP maps generally requires access to and control over auxiliary systems, resulting in an implicit energy cost for preparation and manipulation of the auxiliaries that is obscured by the use of the CPTP maps instead of the explicit description of the corresponding unitaries on the total Hilbert space. Consequently, it is of interest to understand the limits of unitary correlating processes, which allow for a complete account of all the energetic changes within the system. Moreover, to give a fair account of the energy that needs to be supplied to the system, it is important to consider initial states that do not implicitly supply free energy. That is, we assume that the initial states of the subsystems are thermal states at the same temperature T, i.e., $\varrho_A = \tau_A(\beta)$ and $\varrho_B = \tau_B(\beta)$ with $\beta = 1/T$ (we use units where $k_B = 1$ throughout), where $\tau(\beta) := e^{-\beta H}/\mathcal{Z}$ and $\mathcal{Z} = \mathrm{Tr}(e^{-\beta H})$. In this way, the initial state $\tau_A(\beta) \otimes \tau_B(\beta)$ minimises the local energies for given entropies and is also completely passive [25], i.e., a state from which no energy can be extracted unitarily even when taking multiple copies.

Correlations can be quantified in different ways. Qualitatively, one may describe correlations between two subsystems as the feature that more information is available about properties of the joint system than about properties of the subsystems. If there is no preference as to the specific properties that are to be correlated, i.e., if one is interested in quantifying any correlations, it is most useful to consider entropic measures. The most paradigmatic among them is the mutual information $\mathcal{I}(\varrho_{AB}) = S(\varrho_A) + S(\varrho_B) - S(\varrho_{AB})$, based on the von Neumann entropy $S(\varrho) := -\mathrm{Tr}\big(\varrho \ln \varrho\big)$. Although we focus on the von Neumann entropy, in principle one can replace it with any other measure of local 'mixedness'. And indeed, our methods are framed in the context of majorisation relations, and thus in principle also open an avenue towards studying other entropic measures.

When focusing on the mutual information, the problem of unitarily creating correlations can be reduced to the task of maximising the sum of the marginal entropies under local energy constraints. That is, the invested energy $\Delta E := \mathrm{Tr}\big[H_{AB}\big(U_{AB}\tau_A(\beta) \otimes \tau_B(\beta)U_{AB}^\dagger - \tau_A(\beta) \otimes \tau_B(\beta)\big)\big]$, where $H_{AB} := H_A + H_B$, only depends on the local marginals, while the global entropy is invariant under unitary operations. The change in mutual information thus reduces to $\Delta\mathcal{I} = \Delta S_A + \Delta S_B$. We thus arrive at the following question:

Question 1: Maximal mutual information under energy constraint

For a given pair of local Hamiltonians H_A and H_B and initial temperature $T = 1/\beta$, what is the maximum value of

$$\Delta\mathcal{I} = \Delta S_A + \Delta S_B \tag{1}$$

for an invested energy of at most ΔE?

The fact that thermal states maximise the local entropies under local energy constraints suggests that the maximal value of $\Delta\mathcal{I}$ is always achieved when the final state marginals are thermal at a higher temperature $T' \geq T$ (or, equivalently, for $\beta' \leq \beta$), i.e., $\tilde{\varrho}_A = \tau_A(\beta')$ and $\tilde{\varrho}_B = \tau_B(\beta')$, provided that the corresponding STUs exist. This is indeed the case as shown in Appendix A.I and is what leads us to the question of the existence of STUs. While when $H_A = H_B$, one might still hope for STUs to exist for all dimensions, Hamiltonians and temperatures, such a statement cannot be made when $H_A \neq H_B$. Before introducing our framework for constructing STUs, let us therefore briefly discuss general bounds on the achievable local entropies.

II.1.1. Asymmetric case

Let us consider Question 1 in the case $H_A \neq H_B$. Although the problem is in general complex, note that nontrivial constraints on marginal spectra for given global spectra exist in the form of entropy inequalities. In particular, both the von Neumann entropy $S(\varrho)$ and the Rényi 0-entropy $S_0(\varrho) := -\log_2(\mathrm{rank}(\varrho))$ satisfy subadditivity [26, 27], while no other entropy satisfies (dimension independent) linear inequalities [28]. Since we are interested in thermodynamic questions, where the rank of the involved states is typically full, only subadditivity of the von Neumann entropy, which we can write as

$$S(\varrho_{AB}) \geq |S(\varrho_A) - S(\varrho_B)|, \tag{2}$$

provides a nontrivial constraint. Also observe that, given an initial state $\tau_A(\beta) \otimes \tau_B(\beta)$ subject to a unitary evolution, the entropy of the total system is fixed to $S(\varrho_{AB}) = S(\tau_A(\beta)) + S(\tau_B(\beta))$. If we assume that the final marginals are thermal states with equal higher temperature i.e., $\beta' \leq \beta$, inequality (2) can be rewritten as

$$|S(\tau_A(\beta')) - S(\tau_B(\beta'))| \leq S(\tau_A(\beta)) + S(\tau_B(\beta)). \tag{3}$$

This already implies a nontrivial constraint, and also provides a counterargument to the naive assumption that two thermal marginals at the same temperature are reachable in general.

While this makes the structure complicated to deal with in general, one can still solve the optimisation in some special cases. In particular, for a system initialised in the ground state, i.e., a product of two pure states $\tau_{AB}(\beta \to \infty) = |0_A\, 0_B\rangle\langle 0_A\, 0_B|$, the total state remains pure during the unitary evolution and hence has a Schmidt decomposition. The final state can therefore be written as

$$|\tilde{\psi}\rangle = \sum_{i=0}^{d} \sqrt{p_i}\, |\varphi_i^A, \varphi_i^B\rangle, \tag{4}$$

for some probabilities p_i, with $d = \min\{d_A, d_B\}$, and d_A and d_B the dimensions of the respective Hilbert spaces $\mathcal{H}_A$ and $\mathcal{H}_B$. And the final marginals are

$$\tilde{\varrho}_A = \sum_{i=0}^{d-1} p_i\, |\varphi_i^A\rangle\langle\varphi_i^A|, \quad \tilde{\varrho}_B = \sum_{i=0}^{d-1} p_i\, |\varphi_i^B\rangle\langle\varphi_i^B|. \tag{5}$$

This already means that the marginals (since they have equal rank) cannot both be thermal states if $d_A \neq d_B$.

However, in this case it is still possible to maximise the amount of correlation subject to a given maximal amount of energy consumption. In other words, one can find optimal points of the following optimisation problem:

$$\begin{aligned} &\max_{\tilde{\varrho}_A, \tilde{\varrho}_B} S(\tilde{\varrho}_A) + S(\tilde{\varrho}_B) \\ &\text{subject to } \mathrm{Tr}(\tilde{\varrho}_A H_A) + \mathrm{Tr}(\tilde{\varrho}_B H_B) \leq c \\ &\qquad \tilde{\varrho}_A \text{ and } \tilde{\varrho}_B \text{ as in Eq. (5).} \end{aligned} \tag{6}$$

To do so one first solves the (loosely speaking) inverse problem of minimising the energy consumption for a given amount of correlation. This problem can be turned into two instances of the well-known passivity problem [25], for which passive states are the solutions. Using this fact, the problem reduces to a simple problem that can be solved using Lagrange multipliers. One then shows that the solution found for that problem is strictly monotonically increasing in the constraint, allowing us to reverse the problem and show that it is a solution of the problem in (6). For more details see Appendix A.II. See also Ref. [29] for an alternative approach to the energy minimisation problem. The solutions obtained for (6) are thermal states at inverse temperature $\beta(c)$ (uniquely defined by the constant c) of the Hamiltonians $\tilde{H}_A$ and $\tilde{H}_B$, respectively, defined as

$$\begin{aligned} \tilde{H}_A &= \sum_{i=0}^{d-1} (E_i^A + E_i^B)\, |i\rangle\langle i|_A \\ \tilde{H}_B &= \sum_{i=0}^{d-1} (E_i^A + E_i^B)\, |i\rangle\langle i|_B, \end{aligned} \tag{7}$$

where

$$\begin{aligned} H_A &= \sum_{i=0}^{d_A-1} E_i^A\, |i\rangle\langle i|_A, \quad E_i^A \leq E_{i+1}^A, \\ H_B &= \sum_{i=0}^{d_B-1} E_i^B\, |i\rangle\langle i|_B, \quad E_i^B \leq E_{i+1}^B. \end{aligned} \tag{8}$$

The solutions hence take the form

$$\begin{aligned} \tilde{\varrho}_{\mathrm{opt},A}(\beta(c)) &= \frac{\tilde{\Pi}_A e^{-\beta(c)\tilde{H}_A} \tilde{\Pi}_A}{\mathrm{Tr}(\tilde{\Pi}_A e^{-\beta(c)\tilde{H}_A})}, \\ \tilde{\varrho}_{\mathrm{opt},B}(\beta(c)) &= \frac{\tilde{\Pi}_B e^{-\beta(c)\tilde{H}_B} \tilde{\Pi}_B}{\mathrm{Tr}(\tilde{\Pi}_B e^{-\beta(c)\tilde{H}_B})}, \end{aligned} \tag{9}$$

where $\tilde{\Pi}_{A/B} := \sum_{i=0}^{d-1} |i\rangle\langle i|_{A/B}$. The maximal amount of correlation achievable given a maximal amount of energy c is then

$$2S(\tilde{\varrho}_A) = 2\beta(c)c + 2\ln\left[\mathrm{Tr}(e^{-\beta(c)\tilde{H}_A})\right]. \tag{10}$$

II.1.2. Symmetric case

Let us now consider the symmetric case $H_A = H_B$. Following our considerations in the previous section and Appendix A.I, it is clear that an upper bound for $\Delta\mathcal{I}$ is in this case achieved when $\Delta S_A = \Delta S_B$, and since the states with maximal entropy given a fixed energy are thermal states, Question 1 can be substantially simplified to that of the existence of symmetrically thermalizing unitaries.

Question 2: Existence of STUs

Does there exist a unitary U_{AB} on $\mathcal{H}_{AB}$ such that

$$\begin{aligned} \tilde{\varrho}_A &= \mathrm{Tr}_B\left(U_{AB}\tau_{AB}(\beta)U_{AB}^\dagger\right) = \tau_A(\beta'), \\ \tilde{\varrho}_B &= \mathrm{Tr}_A\left(U_{AB}\tau_{AB}(\beta)U_{AB}^\dagger\right) = \tau_B(\beta'), \end{aligned} \tag{11}$$

for every pair of local Hamiltonians $H_A = H_B$, for all final temperatures $T' = 1/\beta'$ and all initial temperatures $T = 1/\beta \leq T'$?

If such STUs exist, then they are automatically the optimally correlating unitaries for a given system. It is already known that Question 1 can be answered affirmatively when the subsystem Hamiltonians are equally spaced, i.e., $H_A = H_B = \sum_{n=0}^{d-1} E_n |n\rangle\langle n|$ with $E_{n+1} - E_n = \omega \; \forall n \in \{0, 1, \ldots, d-1\}$ for some constant ω (with appropriate units) [11] (see also the more recent formulation of the derivation in Ref. [30]). In particular, this implies that such optimally correlating unitaries always exist for two qubits, i.e., when $d = 2$. However, there is (yet) no answer for the general situation of arbitrary Hamiltonians and dimensions. To fill this gap, we present a framework that generalises previous approaches [11] and allows investigating this general question in the symmetric cases for any dimension. This approach is based on unitary operations in LCSs, as we will explain in Sec. II.2, and enables us to show that STUs exist for the simplest nontrivial cases of two qutrits ($d = 3$) and two ququarts ($d = 4$).

II.2. Unitary operations on locally classical subspaces

Let us now discuss our general framework for marginal transformations. The central idea of this approach is to decompose the diagonal elements of the marginals into elements originating from different subspaces, with the property that any unitary that leaves the division into these subspaces invariant never creates local off-diagonal elements.

More precisely, we consider a pair of d-dimensional systems A and B with matching local Hamiltonians $H_A = H_B = \sum_{i=0}^{d-1} E_i |i\rangle\langle i|$, where we set $E_0 = 0$ without loss of generality. Moreover, let the energy eigenvalues, sorted in nondecreasing order (with $E_{i+1} \geq E_i \; \forall i$), be measured in units of E_1. The initial thermal states of A and B are

$$\tau_A(\beta) = \tau_B(\beta) = \sum_{i=0}^{d-1} p_i(\beta) \, |i\rangle\langle i| , \tag{16}$$

where $p_i(\beta) = e^{-\beta E_i}/\mathcal{Z}(\beta)$ and $\mathcal{Z}(\beta) = \sum_i e^{-\beta E_i}$. For convenience, we use the shorthand $p_i \equiv p_i(\beta)$. The joint initial state $\tau_{AB} = \tau_A(\beta) \otimes \tau_B(\beta)$ is also diagonal with respect to the energy eigenbasis and its diagonal entries are products of the diagonal entries of the reduced states, i.e., we write $p_{ij} := p_i p_j$ such that $\tau_{AB} = \sum_{i,j=0}^{d-1} p_{ij} |ij\rangle\langle ij|$. Since the unitaries that we use throughout the manuscript do not create coherence in the marginals, it is convenient to introduce a vectorised notation for the diagonal entries of the reduced states. That is, for arbitrary diagonal joint states $\mathbf{p}_{AB} = \mathrm{diag}\{p_{ij}\}_{i,j=0}^{d-1}$ where the p_{ij} need not factorise with respect to i and j, the reduced states $\varrho_A = \mathrm{Tr}_B(\varrho_{AB})$ and $\varrho_B = \mathrm{Tr}_A(\varrho_{AB})$ are diagonal, and the diagonal entries can be collected into vectors $\mathbf{p}_A, \mathbf{p}_B \in \mathbb{R}^d$ with nonnegative components and unit 1-norm, $\|\mathbf{p}_A\| = \mathrm{Tr}(\varrho_A) = 1 = \|\mathbf{p}_B\|$.

II.2.1. Locally classical subspaces

Having expressed the diagonals of the marginals in this vectorised form, we further choose particular subspace vector decompositions, as we illustrate for $d = 3$ in Box 1. For general local dimension $d \geq 3$, we write $\mathbf{p}_A$ and $\mathbf{p}_B$ as sums of d vectors according to

$$\mathbf{p}_A = \sum_{i=0}^{d-1} \mathbf{r}_i^A = \sum_{i=0}^{d-1} \mathbf{r}_i, \quad \mathbf{p}_B = \sum_{i=0}^{d-1} \mathbf{r}_i^B = \sum_{i=0}^{d-1} \Pi^i \mathbf{r}_i, \tag{17}$$

where $\mathbf{r}_i^A = \mathbf{r}_i = \sum_{j=0}^{d-1} p_{j\,j+i} \, \mathbf{e}_j$ and $\mathbf{r}_i^B = \sum_{j=0}^{d-1} p_{j-i\,j} \, \mathbf{e}_j$. Here, $\{\mathbf{e}_j\}_{j=0}^{d-1}$ denotes the chosen orthonormal basis of $\mathbb{R}^d$ and all indices are to be taken modulo d. In the second equality, we have further used the fact that the i-th vector $\mathbf{r}_i^B$ in the decomposition of $\mathbf{p}_B$ can be related to the i-th vector $\mathbf{r}_i^A$ in the decomposition of $\mathbf{p}_A$ via the i-th power of the cyclic permutation matrix $\Pi = (\Pi_{ij})$ with components $\Pi_{ij} = \delta_{i,j+1 \bmod d}$.

One further observes that the decomposition of $\mathbf{p}_A$ and $\mathbf{p}_B$ into these vectors corresponds to the selection of a total of d subspaces $\mathcal{H}_q$ and $\{\mathcal{H}_{r_i}\}_{i=1}^{d-1}$ of the joint Hilbert space $\mathcal{H}_{AB}$,

$$\mathcal{H}_q := \mathcal{H}_{r_0} = \mathrm{span}\{|j\,j\rangle\}_{j=0}^{d-1}, \quad \mathcal{H}_{r_i} = \mathrm{span}\{|j\,j+i\rangle\}_{j=0}^{d-1}, \tag{18}$$

with $\mathcal{H}_{AB} = \mathcal{H}_q \oplus_{i=1}^{d-1} \mathcal{H}_{r_i}$, such that arbitrary unitaries U_q and U_{r_i} applied in either of the d subspaces cannot lead to nonzero off-diagonal elements in the reduced states ϱ_A or ϱ_B, provided that none are present to begin with. In other words, any state on $\mathcal{H}_{AB}$ that has support on only one of these subspaces, or which has a block-diagonal structure with respect to this subspace decomposition, is locally classical, i.e., has diagonal marginals with respect to the local bases $\{|j\rangle\}_{j=0}^{d-1}$. We hence call these subspaces *locally classical*. However, note that the choice of LCSs we make here is not unique, as discussed below in Sec. II.2.2.

A unitary transformation $\varrho_{AB} \mapsto \tilde{\varrho}_{AB} = U \varrho_{AB} U^\dagger$ with $U = U_q \oplus_{i=1}^{d-1} U_{r_i}$ hence does not preclude off-diagonal elements from appearing in the joint state $\tilde{\varrho}_{AB}$ but leaves the reduced states diagonal, implying that we can directly read off the spectra of the marginals. In such a case, it is still useful to describe the marginals by d-component vectors $\tilde{\mathbf{p}}_A$ and $\tilde{\mathbf{p}}_B$ collecting their diagonal elements, and the transformation of these vectors can be represented as

$$\mathbf{p}_A \mapsto \tilde{\mathbf{p}}_A = M_q \mathbf{q} + \sum_{i=1}^{d-1} M_{r_i} \mathbf{r}_i, \tag{19}$$

$$\mathbf{p}_B \mapsto \tilde{\mathbf{p}}_B = M_q \mathbf{q} + \sum_{i=1}^{d-1} \Pi^i M_{r_i} \mathbf{r}_i, \tag{20}$$

where $\mathbf{q} = \mathbf{r}_0^A = \mathbf{r}_0^B$, and M_q and M_{r_i} are unistochastic $d \times d$ matrices, i.e., doubly stochastic (entries in rows and columns sum to 1) square matrices M_α (with $\alpha \in \{q, r_i\}$) whose components can be understood as the moduli squared of unitary matrices, $(M_\alpha)_{kl} = |(U_\alpha)_{kl}|^2$.

As a technical remark, note that the Schur-Horn theorem implies a weak convexity property for unistochastic matrices, namely that for any $\mathbf{v} \in \mathbb{R}^d$, the set of vectors obtained by applying the set of unistochastic $d \times d$ matrices to $\mathbf{v}$ is equivalent to the set of vectors obtained by applying the set of doubly stochastic $d \times d$ matrices to $\mathbf{v}$, see Ref. [31]. This means that

Box 1: Unitary operations on locally classical subspaces for two qutrits

For any tensor product of two identical states $\tau_{AB} = \sum_{i,j=0}^{2} p_{ij} |ij\rangle\langle ij|$, we can separate the local diagonal elements (i.e., the eigenvalues, since the matrix is diagonal) through an economic notation where we denote the diagonal entries of the marginals as entries in vectors

$$\mathbf{p}_A := \tau_A = \begin{pmatrix} p_{00}+p_{01}+p_{02} \\ p_{11}+p_{12}+p_{10} \\ p_{22}+p_{20}+p_{21} \end{pmatrix} = \mathbf{q} + \mathbf{r}_1 + \mathbf{r}_2, \quad \mathbf{p}_B := \tau_B = \begin{pmatrix} p_{00}+p_{10}+p_{20} \\ p_{11}+p_{21}+p_{01} \\ p_{22}+p_{02}+p_{12} \end{pmatrix} = \mathbf{q} + \Pi^2 \mathbf{r}_2 + \Pi \mathbf{r}_1, \tag{12}$$

where $\Pi = (\Pi_{ij})$ is a cyclic permutation matrix with components $\Pi_{ij} = \delta_{i,j+1 \bmod 3}$. Employing the unitary $U_{AB} = U_q \oplus U_{r_1} \oplus U_{r_2}$, where U_q, U_{r_1}, and U_{r_2} act unitarily on the subspaces

$$\mathcal{H}_q := \mathrm{span}\{|00\rangle, |11\rangle, |22\rangle\}, \quad \mathcal{H}_{r_1} := \mathrm{span}\{|01\rangle, |12\rangle, |20\rangle\}, \quad \mathcal{H}_{r_2} := \mathrm{span}\{|02\rangle, |10\rangle, |21\rangle\}, \tag{13}$$

respectively, the transformation of the marginals can be described by unistochastic matrices

$$\tilde{\mathbf{p}}_A = M_q \mathbf{q} + M_{r_1}\mathbf{r}_1 + M_{r_2}\mathbf{r}_2, \quad \tilde{\mathbf{p}}_B = M_q \mathbf{q} + \Pi^2 M_{r_2}\mathbf{r}_2 + \Pi M_{r_1}\mathbf{r}_1. \tag{14}$$

The components of the unistochastic matrices are determined by the moduli squared of corresponding unitary matrices, $(M_\alpha)_{kl} = |(U_\alpha)_{kl}|^2$ for $\alpha = q, r_1, r_2$. Due to the symmetry $p_{ij} = p_{ji}$, we can further identify $\mathbf{r}_2 = \Pi \mathbf{r}_1$ to obtain

$$\tilde{\mathbf{p}}_A = M_q \mathbf{q} + M_{r_1}\mathbf{r}_1 + M_{r_2}\Pi\mathbf{r}_1, \quad \tilde{\mathbf{p}}_B = M_q \mathbf{q} + \Pi^2 M_{r_2}\Pi \mathbf{r}_1 + \Pi M_{r_1}\mathbf{r}_1. \tag{15}$$

for any doubly stochastic matrix M and vector $\mathbf{v}$, there exists a unistochastic matrix $M_\mathbf{v}$ such that $M\mathbf{v} = M_\mathbf{v}\mathbf{v}$. Here, this property permits us to conclude that the vectorised marginals of any state reachable by unitaries of the form $U = U_q \oplus_{i=1}^{d-1} U_{r_i}$ can be written in terms of the action of doubly stochastic matrices M_q and M_{r_i} on $\mathbf{q}$ and $\mathbf{r}_i$, respectively.

In general, Eq. (19) can be rewritten as

$$\tilde{\mathbf{p}}_A = \begin{cases} M_q\mathbf{q} + \sum_{i=1}^{k} (M_{r_i}\mathbf{r}_i + M_{r_{d-i}}\mathbf{r}_{d-i}) & d \text{ odd} \\ M_q\mathbf{q} + \sum_{i=1}^{k-1} (M_{r_i}\mathbf{r}_i + M_{r_{d-i}}\mathbf{r}_{d-i}) + M_{r_{d/2}}\mathbf{r}_{d/2} & d \text{ even} \end{cases}, \tag{21}$$

where $k = \frac{d-1}{2}$ if d is odd and $k = \frac{d}{2}$ if d is even. Further taking into account the symmetry $p_{ij} = p_{ji}$, which implies $\mathbf{r}_{d-i} = \sum_{j=0}^{d-1} p_{j\,j+d-i}\Pi^i \mathbf{e}_{j-i} = \Pi^i \mathbf{r}_i$, the vectorised form of the marginal A can be written in a compact way as

$$\tilde{\mathbf{p}}_A = M_q\mathbf{q} + \sum_{i=1}^{k} (\lfloor \tfrac{2i}{d} \rfloor + 1)^{-1} (M_{r_i} + M_{r_{d-i}}\Pi^i)\mathbf{r}_i, \tag{22}$$

where $\lfloor x \rfloor$ denotes the floor function of x, and the prefactor $(\lfloor \frac{2i}{d} \rfloor + 1)^{-1}$ is equal to 1 unless d is even and $i = k$, in which case it is $\frac{1}{2}$. Using Eq. (20), one may obtain the transformed marginal B as

$$\tilde{\mathbf{p}}_B = M_q\mathbf{q} + \sum_{i=1}^{k} (\lfloor \tfrac{2i}{d} \rfloor + 1)^{-1} (\Pi^i M_{r_i} + \Pi^{-i} M_{r_{d-i}} \Pi^i)\, \mathbf{r}_i, \tag{23}$$

where we have used the property $\Pi^{-i} = \Pi^{d-i}$ of the d-dimensional cyclic permutation. To satisfy the conditions of Question 2, we need to further restrict ourselves to a subgroup of transformations which generate the same marginals, $\tilde{\mathbf{p}}_A = \tilde{\mathbf{p}}_B$. This requirement results in the condition

$$M_{r_{d-i}} = \Pi^i M_{r_i} \Pi^{-i} \quad \text{for} \quad i = 1, \ldots, k, \tag{24}$$

for the doubly stochastic matrices mentioned in Eq. (22). With this, we arrive at the form

$$\tilde{\mathbf{p}} = \tilde{\mathbf{p}}_A = \tilde{\mathbf{p}}_B = M_q\mathbf{q} + \sum_{i=1}^{k} (\lfloor \tfrac{2i}{d} \rfloor + 1)^{-1} (\mathbb{1} + \Pi^i) M_{r_i}\mathbf{r}_i \tag{25}$$

for both vectorised marginals. Now that we have ensured that both marginals transform in the same way, we may concentrate on one of them, say $\tilde{\mathbf{p}}_A$ (dropping the subscript A for ease of notation) and use the established framework to investigate the existence of STUs as specified in Question 2.

II.2.2. General locally classical subspaces

While we focus here on the decomposition into the specific LCSs from Eq. (18), we note that this is by far not the only option. Indeed, let $\{\mathcal{P}_i\}_{i=0,\ldots,d-1}$ be a set of permutations of d elements $j = 0, \ldots, d-1$. Then a sufficient condition for obtaining an LCS decomposition $\mathcal{H}_{AB} = \bigoplus_{i=0}^{d-1} \tilde{\mathcal{H}}_{\tilde{r}_i}$ into LCSs of the form $\tilde{\mathcal{H}}_{\tilde{r}_i} = \mathrm{span}\{|j\,\mathcal{P}_i(j)\rangle\}_{j=0}^{d-1}$ is that the $d \times d$ matrix Γ with components $\Gamma_{ij} = \mathcal{P}_i(j)$ is a Latin square, i.e., every entry $j = 0, \ldots, d-1$ appears exactly once in each row and in each column. For every such choice of LCS decomposition, one may then apply unitaries $\tilde{U} = \bigoplus_{i=0}^{d-1} U_{\tilde{r}_i}$ that leave the LCSs invariant, i.e., where $U_{\tilde{r}_i}$ acts nontrivially only on the subspace $\tilde{\mathcal{H}}_{\tilde{r}_i}$. Denoting the corresponding vector decomposition of the first vectorised marginal as

$\mathbf{p}_A = \sum_{i=0}^{d-1} \tilde{\mathbf{r}}_i$, with $(\tilde{\mathbf{r}}_i)_j = p_{j\mathcal{P}_i(j)}$, we have $\mathbf{p}_B = \sum_{i=0}^{d-1} \mathcal{P}_i^{-1}\tilde{\mathbf{r}}_i$ since $(\mathbf{r}_i^B)_j = p_{\mathcal{P}_i^{-1}(j)j} = (\mathcal{P}_i^{-1}\tilde{\mathbf{r}}_i)_j$. In this case, unitaries that leave the LCSs invariant transform the vectorised marginals according to

$$\mathbf{p}_A \mapsto \tilde{\mathbf{p}}_A = \sum_{i=0}^{d-1} M_{\tilde{r}_i}\tilde{\mathbf{r}}_i, \tag{26}$$

$$\mathbf{p}_B \mapsto \tilde{\mathbf{p}}_B = \sum_{i=0}^{d-1} \mathcal{P}_i^{-1} M_{\tilde{r}_i}\tilde{\mathbf{r}}_i, \tag{27}$$

where the matrices $M_{\tilde{r}_i}$ are the unistochastic matrices corresponding to the unitaries $U_{\tilde{r}_i}$. This implies the existence of symmetric marginal transformations, e.g., for unistochastic matrices M that commute with all the $\mathcal{P}_i$, such that the marginals take the form $M\mathbf{p}_A = M\mathbf{p}_B = \tilde{\mathbf{p}}$. For the remainder of this work we focus again on the specific special case we consider in Eq. (18), where $\mathcal{P}_i = (\Pi^{-1})^i$.

II.2.3. Asymptotic case

Before we go further into detail on trying to answer Question 2, let us briefly showcase that this is indeed a problem connected to the finite size of the system. More specifically, we can consider a scenario where one wishes to correlate multiple copies of the initial thermal states $\tau_A(\beta)$ and $\tau_B(\beta)$ via a joint unitary, such that the final state of n copies is $U\tau_{AB}(\beta)^{\otimes n}U^\dagger$, with $\tau_{AB}(\beta)^{\otimes n} = \tau_A(\beta)^{\otimes n} \otimes \tau_B(\beta)^{\otimes n}$. As we will show now, STUs such that $\tilde{\varrho}_A = \tilde{\varrho}_B = \tau(\beta')$ exist for all β', β such that $\beta' \leq \beta$, and for all local Hamiltonians in the limit of infinitely many copies, $n \to \infty$.

To see this, note that for any n we can find a unitary such that the marginals $\varrho_{A/B} = \mathrm{Tr}_{B/A}(U\tau_A(\beta)^{\otimes n} \otimes \tau_B(\beta)^{\otimes n}U^\dagger)$ are passive states whose entropy equals that of the thermal state with the target temperature, i.e., $S(\varrho_{A/B}) = S(\tau_{A/B}(\beta'))$. This is a consequence of Eq. (25) and the continuity of the von Neumann entropy. More specifically, note that a trivial way of obtaining marginals with the same final spectrum is to select M_q and all M_{r_i} in Eq. (25) to be circulant, i.e., convex combinations of cyclic permutations. Since these matrices commute with Π^i, one may reach any marginal whose vectorised marginal is any cyclic permutation of the original marginal vector, or indeed, any convex combination of cyclic permutations of the original marginal vector. In particular, the equally weighted convex combination of all cyclic permutations yields maximally mixed marginals, $\tilde{\mathbf{p}}_A = \tilde{\mathbf{p}}_B = \frac{1}{d}\sum_{i=0}^{d-1} \mathbf{e}_i$, corresponding to maximal local entropy $\beta' \to 0$. The convex structure of the set of vectors reachable from a given vector via application of unistochastic matrices then suggests that the points corresponding to $\mathbf{p}_A$ and $\tilde{\mathbf{p}}_A$ are connected by a continuous line of states reachable from $\mathbf{p}_A$ via circulant unistochastic matrices. The entropy must vary continuously along this line from the initial value $S\big(\tau_A(\beta)\big)$ to $S\big(\varrho_A\big) = S\big(\tau_A(\beta')\big)$. Thus, one may obtain marginals $\varrho_A = \varrho_B$ that are passive states with the desired entropy, but which might have a different spectra than $\tau_A(\beta') = \tau_B(\beta')$.

Given this fact, it is sufficient to note that for $n \to \infty$ we can convert passive states with a given entropy $S(\varrho)$ into thermal states with the same entropy only by using local operations [25]. Because the application of local operations to the subsystems leaves the mutual information $\mathcal{I}(\varrho_{AB})$ invariant, this consequently proves the existence of STUs for the asymptotic case $n \to \infty$. Further discussion of the case of finitely many copies is presented in Appendix A.III.

III. OPTIMALLY CORRELATING UNITARIES FOR BIPARTITE STATES WITH MATCHING HAMILTONIANS

In this section, we present three approaches to construct STUs for bipartite systems in an (uncorrelated) initial thermal state at temperature $T = 1/\beta$ of $H = H_A + H_B$ with matching Hamiltonians $H_A = H_B$. We focus on bipartite 3×3 and 4×4-dimensional systems, and show explicit constructions for such STUs, thereby proving that Question 2 can be answered affirmatively for $d = 3$ and $d = 4$. In the $d = 3$ case, we show the existence of STUs via all three alternative approaches as a basis for generalisations to higher dimensions. We then turn to the particular case of dimension $d = 4$, highlighting the challenges and strengths encountered for each of these methods, before proving the $d = 4$ case using a geometric argument.

Concretely, referring to Sec. II.2 as our framework, we start from Eq. (25), which provides a transformations of the reduced states that leaves both marginals equal and diagonal with respect to the chosen basis. In general, M_q and all the M_{r_i} are arbitrary doubly stochastic matrices, and therefore one can try to make use of majorisation conditions to prove the existence of STUs via the Hardy-Littlewood-Pólya (HLP) theorem [32, p. 75] or [33, p. 33]. The HLP theorem states that a necessary and sufficient condition that $y \succ x$ is that there exist a doubly stochastic matrix M such that $x = My$. This, in fact, is the first argument that we use below for $d = 3$, where it allows proving a statement that is actually even stronger than what is required for STUs. However, the theorem that we are going to prove holds only for the $d = 3$ case.

III.1. Majorised marginals approach

In the particular case of $d = 3$ the final expression for the (equal) marginals from Eq. (25) is

$$\tilde{\mathbf{p}} = \tilde{\mathbf{p}}_B = \tilde{\mathbf{p}}_A = M_q\mathbf{q} + (\mathbb{1} + \Pi)M_r\mathbf{r}, \tag{28}$$

where, as in Eq. (20), we have introduced the vectors $\mathbf{q}$ and $\mathbf{r}$, with components

$$\mathbf{q} = \big(p_{00},\, p_{11},\, p_{22}\big)^T, \quad \mathbf{r} = \big(p_{01},\, p_{12},\, p_{20}\big)^T. \tag{29}$$

Based on the HLP theorem and Eq. (28), we are going to prove the existence of STUs in $d = 3$ via the following lemma:

Lemma 1. *For any 3×3 doubly stochastic matrix M, there exists a 3×3 doubly stochastic matrix $\tilde{M}$ such that*

$$M(\mathbb{1} + \Pi) = (\mathbb{1} + \Pi)\tilde{M}. \tag{30}$$

The proof of Lemma 1 is presented in Appendix A.IV. We are now ready to state the first theorem for $d = 3$.

Theorem 1: Majorised marginals in $d = 3$

For every pair of states ϱ and $\tilde{\varrho}$ in a 3-dimensional Hilbert space which satisfy the condition $\lambda(\tilde{\varrho}) \prec \lambda(\varrho)$, where $\lambda(\varrho)$ is the vector of eigenvalues of ϱ, there exists a unitary U_{AB} on $\mathcal{H}_{AB}$ such that

$$\tilde{\varrho}_A = \mathrm{Tr}_B\left(U_{AB}\varrho \otimes \varrho U_{AB}^\dagger\right) = \tilde{\varrho},$$
$$\tilde{\varrho}_B = \mathrm{Tr}_A\left(U_{AB}\varrho \otimes \varrho U_{AB}^\dagger\right) = \tilde{\varrho}.$$

Proof. The unitary is given by $U_{AB} = U_{\mathrm{loc}}U_{\mathrm{ent}}$, i.e., by a product of an entangling unitary U_{ent} which acts on the global system preserving the equal marginals as discussed in Sec. II.2, and a local unitary of the form $U_{\mathrm{loc}} = U \otimes U$, such that the marginals are kept equal. From Lemma 1 we then see that, given any doubly stochastic matrix M_q, one can find M_r such that $M_q(\mathbb{1} + \Pi) = (\mathbb{1} + \Pi)M_r$ and consequently a mapping to each probability vector $\tilde{\mathbf{p}}$ such that

$$\tilde{\mathbf{p}} = M_q\mathbf{q} + (\mathbb{1} + \Pi)M_r\mathbf{r} = M_q\left[\mathbf{q} + (\mathbb{1} + \Pi)\mathbf{r}\right] = M_q\,\mathbf{p}. \tag{31}$$

From the HLP theorem, we thus know that it is possible to produce all the vectors $\tilde{\mathbf{p}}$ that are majorised by the vector $\mathbf{p}$ corresponding to the initial state. Thus, we proved that through the entangling unitary, all the marginals $\varrho' = \sum_{i=0}^{2}\tilde{p}_i\,|i\rangle\langle i|$ such that $\tilde{\mathbf{p}} \prec \mathbf{p}$ can be reached from the initial state. Then, if we can also arbitrarily change their eigenstates in a symmetric way, Theorem 1 is proven. For that, we use a local unitary $U \otimes U$ with $U = \sum_{i=0}^{2}|\phi_i\rangle\langle i|$ to change the eigenstates of the marginal to an arbitrary set of eigenstates $\{|\phi_i\rangle\}_{i=0}^{2}$, such that finally the marginals become $\tilde{\varrho} = \sum_{i=0}^{2}\tilde{p}_i\,|\phi_i\rangle\langle\phi_i|$ in which the set $\{|\phi_i\rangle\}$ can be any orthonormal basis in $d = 3$. □

As a corollary of Theorem 1, the existence of STUs is proven for the two-qutrit case for initial and final thermal states with inverse temperatures β and $\beta' \leq \beta$, respectively, since $\mathbf{p}(\beta') \prec \mathbf{p}(\beta)$ holds whenever $\beta' \leq \beta$.

Unfortunately, generalisations of Lemma 1 to higher dimensions fail, as we explain in more detail in Appendix A.V. Despite the general statement of Theorem 1, any attempts at generalisations must hence be based on a different approach, which does not require the existence of a doubly stochastic matrix $\tilde{M}$ as in Eq. (30) for all doubly stochastic matrices M.

III.2. Alternative approach: "passing on the norm"

Here, we explore another possible approach that makes use of the HLP theorem and tries to overcome the difficulties encountered for generalising the previous approach. Let us first discuss the method for two d-dimensional systems, before turning to the special cases $d = 3$ and $d = 4$.

To begin, we once again employ the transformation presented in Eq. (25) to generate equal marginals. Recalling that the marginals of our final target state are thermal states at inverse temperature $\beta' < \beta$, we decompose the final state vector into the form

$$\tilde{\mathbf{p}} = \mathbf{a} + \sum_{i=1}^{k}\left(\left\lfloor\tfrac{2i}{d}\right\rfloor + 1\right)^{-1}(\mathbb{1} + \Pi^i)\mathbf{b}_i, \tag{32}$$

with $\mathbf{a} := \mathbf{q}(\beta') = \sum_{j=0}^{d-1}p_{jj}(\beta')\,\mathbf{e}_j$ and $\mathbf{b}_i := \mathbf{r}_i(\beta') = \sum_{j=0}^{d-1}p_{jj+i}(\beta')\,\mathbf{e}_j$. Further, recall that the initial states have the same decomposition with inverse temperature β. One way of transforming $\mathbf{p}$ into $\tilde{\mathbf{p}}$ is to transform $\mathbf{q}$ into $\mathbf{a}$ and $\mathbf{r}_i$ into $\mathbf{b}_i$ for all $i = 1, \ldots, k$. Looking at Eq. (25), one realises that this is possible if

$$M_q\mathbf{q} = \mathbf{a}, \tag{33}$$
$$M_{r_i}\mathbf{r}_i = \mathbf{b}_i, \quad i = 1, \ldots, k. \tag{34}$$

Unfortunately, for any $\beta' < \beta$ we have $\|\mathbf{q}\| > \|\mathbf{a}\|$, where $\|\cdot\|$ denotes the 1-norm, see Appendix A.VII, and since doubly stochastic transformations conserve the norm of vectors, Eq. (33) cannot hold true. The norms of the $\mathbf{r}_i$ also generically differ from those of the $\mathbf{b}_i$, meaning Eq. (34) does not hold either. However, it is not clear whether $\|\mathbf{r}_i(\beta)\|$ is a monotonic function of β, and so $\|\mathbf{r}_i\|$ may in principle be smaller or larger than $\|\mathbf{b}_i\|$, depending on β, β' and i. Loosely speaking, the vector $\mathbf{q}$ has 'too much norm'. Intuitively, one may thus make use of the excessive norm of $\mathbf{q}$ by transforming the required 'amount' of $\mathbf{q}$ into $\mathbf{a}$, and redistributing its excessive norm to the rest of $\tilde{\mathbf{p}}$, or in other words by passing the excessive norm of $\mathbf{q}$ on to the rest of $\tilde{\mathbf{p}}$, giving rise to the name of the approach. Formally, one achieves this by splitting M_q into a convex combination of two doubly stochastic matrices, i.e.,

$$M_q = \alpha_0 M_{q\to a} + \alpha_1 M_{q\to b}, \tag{35}$$

where $\alpha_0, \alpha_1 \geq 0$, $\alpha_0 + \alpha_1 = 1$, and $M_{q\to a}$ and $M_{q\to b}$ are doubly stochastic matrices such that

$$M_{q\to a}\frac{\mathbf{q}}{\|\mathbf{q}\|} = \frac{\mathbf{a}}{\|\mathbf{a}\|}, \tag{36}$$
$$M_{q\to b}\frac{\mathbf{q}}{\|\mathbf{q}\|} = \mathbf{f}(\mathbf{b}_1, \ldots, \mathbf{b}_k), \tag{37}$$

where $\mathbf{f}$ depends on the vectors $\mathbf{b}_i$. The main idea of this approach is that if one chooses $\mathbf{f}$ and $M_{q\to b}$ in an appropriate way, the M_{r_i} may potentially be chosen such that

$$M_{r_i}\frac{\mathbf{r}_i}{\|\mathbf{r}_i\|} = \frac{\mathbf{b}_i}{\|\mathbf{b}_i\|}, \quad i = 1, \ldots, k, \tag{38}$$

in order for $\mathbf{p}$ to transform into $\tilde{\mathbf{p}}$ as desired. One way of 'passing on the norm' of $\mathbf{q}$ in this way, i.e. choosing $\mathbf{f}$ and $M_{q\to b}$, is to pass it to each $\mathbf{b}_i$ individually

$$M_{q\to b} = \sum_{i=1}^{k}\tilde{\alpha}_i(\mathbb{1} + \Pi^i)M_{q\to b_i}, \tag{39}$$

$$\text{with } M_{q\to b_i}\frac{\mathbf{q}}{\|\mathbf{q}\|} = \frac{\mathbf{b}_i}{\|\mathbf{b}_i\|}, \quad i = 1, \ldots, k, \tag{40}$$

which implies $\mathbf{f}(\mathbf{b}_1,\dots,\mathbf{b}_k) = \sum_{i=1}^k \tilde{\alpha}_i \frac{\mathbf{b}_i}{\|\mathbf{b}_i\|}$, where the $\{\tilde{\alpha}_i\}_i$ constitutes a probability distribution. However, this is not always possible as the condition $\mathbf{q}/\|\mathbf{q}\| \succ \mathbf{b}_i/\|\mathbf{b}_i\|$, necessary for the existence of $M_{q\to b_i}$, cannot be satisfied in general. See Appendix A.VI for details. One is therefore forced to pass the norm of $\mathbf{q}$ in a more clever way. A good candidate is

$$M_{q\to b} = \sum_{i=1}^k \tilde{\alpha}_i M_{q\to 2b_i}, \tag{41}$$

$$\text{with } M_{q\to 2b_i}\frac{\mathbf{q}}{\|\mathbf{q}\|} = \frac{(\mathbb{1}+\Pi^i)\mathbf{b}_i}{2\|\mathbf{b}_i\|}, \quad i=1,\dots,k. \tag{42}$$

However, this separation of M_q into one part "passing on the norm" from $\mathbf{q}$ to $\mathbf{a}$ and another part from $\mathbf{q}$ to the $\mathbf{b}_i$ is not strictly needed. One may instead only require the existence of a doubly stochastic matrix M_q such that

$$M_q\mathbf{q} = \mathbf{a} + \tilde{\mathbf{f}}(\mathbf{r}_1,\mathbf{b}_1,\dots,\mathbf{r}_k,\mathbf{b}_k), \tag{43}$$

with

$$\tilde{\mathbf{f}}(\mathbf{r}_1,\mathbf{b}_1,\dots,\mathbf{r}_k,\mathbf{b}_k) = \sum_{i=1}^k (\lfloor \tfrac{2i}{d}\rfloor + 1)^{-1}(1-\frac{\|\mathbf{r}_i\|}{\|\mathbf{b}_i\|})(\mathbb{1}+\Pi^i)\mathbf{b}_i. \tag{44}$$

Unfortunately, Eq. (43) is challenging to check, e.g., even confirming whether $\mathbf{a}+\tilde{\mathbf{f}}(\mathbf{r}_1,\mathbf{b}_1,\dots,\mathbf{r}_k,\mathbf{b}_k)$ is a vector of non-negative components is already complicated. In summary, the approach requires checking the two following conditions:

(i) For any pair of vectors $(\mathbf{r}_i,\mathbf{b}_i)$, there exists a doubly stochastic matrix M_{r_i} such that

$$\frac{\mathbf{b}_i}{\|\mathbf{b}_i\|} = M_{r_i}\frac{\mathbf{r}_i}{\|\mathbf{r}_i\|}, \; i=0,\dots,k, \tag{45}$$

while at the same time $\|\mathbf{q}\| \geq \|\mathbf{a}\|$, where $\mathbf{a} := \mathbf{q}(\beta')$.

(ii) There exists a doubly stochastic matrix M_q such that

$$M_q\mathbf{q} = \mathbf{a} + \sum_{i=1}^k (\lfloor \tfrac{2i}{d}\rfloor + 1)^{-1}(1-\frac{\|\mathbf{r}_i\|}{\|\mathbf{b}_i\|})(\mathbb{1}+\Pi^i)\mathbf{b}_i, \tag{46}$$

or the stronger version: $\|\mathbf{r}_i\| \leq \|\mathbf{b}_i\|$ for all $i=1,\dots,k$ and there exist doubly stochastic matrices $M_{q\to 2b_i}$, $i=1,\dots,k$ such that

$$M_{q\to 2b_i}\frac{\mathbf{q}}{\|\mathbf{q}\|} = \frac{(\mathbb{1}+\Pi^i)\mathbf{b_i}}{2\|\mathbf{b_i}\|} \quad i=1,\dots,k. \tag{47}$$

Condition (i) indeed holds, and we thus state it below as the following lemma, the proof of which is presented in Appendix A.VII.

Lemma 2. *For any d-dimensional system with arbitrary Hamiltonian, for any $\beta' < \beta$, and for all $i = 0,\dots,d-1$ we have*

$$\|\mathbf{q}\| \geq \|\mathbf{a}\|, \tag{48}$$

$$\frac{\mathbf{r}_i}{\|\mathbf{r}_i\|} \succ \frac{\mathbf{b}_i}{\|\mathbf{b}_i\|}. \tag{49}$$

Given the above lemma and assuming that condition (ii) also holds, one can prove the existence of STUs for all dimensions by using the following majorisation relations. The majorisation relation (49) from Lemma 2 ensures the existence of doubly stochastic matrices M_{r_i} which satisfy $M_{r_i}\mathbf{r}_i/\|\mathbf{r}_i\| = \mathbf{b}_i/\|\mathbf{b}_i\|$. To reach any thermal state with higher temperature, i.e., $\tilde{\mathbf{p}} = \mathbf{p}(\beta')$, we then need the existence of M_q such that

$$\mathbf{p}(\beta') = M_q\mathbf{q} + \sum_{i=1}^k (\lfloor \tfrac{2i}{d}\rfloor + 1)^{-1}\frac{\|\mathbf{r}_i\|}{\|\mathbf{b}_i\|}(\mathbb{1}+\Pi^i)\mathbf{b}_i, \tag{50}$$

therefore compensating for the required norm of the vectors associated to the subspace $\{\mathcal{H}_{r_i}\}_{i=1}^{d-1}$. This is precisely what is ensured by condition (ii).

However, since condition (ii) is in general cumbersome to prove, one can check whether Eq. (47) holds instead. See Appendix A.VIII for more details about how to prove the existence of STUs in general in this way. In the particular case of $d=3$, the strong version of condition (ii) can be reduced to a ("norm passing") requirement that we formulate in the following lemma:

Lemma 3. *In $d=3$, for every choice of E_1 and E_2 with $E_2 \geq E_1$ and for any $\beta' \leq \beta$, the following majorisation relation holds:*

$$\frac{\mathbf{q}}{\|\mathbf{q}\|} \succ \frac{(\mathbb{1}+\Pi)\mathbf{b}}{2\|\mathbf{b}\|} := \frac{\mathbf{c}}{\|\mathbf{c}\|}. \tag{51}$$

The proof of Lemma 3 is presented in Appendix A.IX. Together, Lemmas 2 and 3 then confirm the existence of STUs for the $d=3$ case.

Now, let us examine the case $d=4$. Consider a bipartite system with equal local Hamiltonians $H = \sum_{i=0}^3 E_i |i\rangle\langle i|$ in which the energy eigenvalues are ordered in increasing order and $E_0 = 0$. Following Eq. (17), the vectorised form of the marginals for the initial uncorrelated thermal state is given by $\mathbf{p} = \mathbf{q}+\mathbf{r}_1+\mathbf{r}_2+\mathbf{r}_3$ with

$$\mathbf{q} = \begin{pmatrix} p_{00}\\ p_{11}\\ p_{22}\\ p_{33}\end{pmatrix}, \; \mathbf{r}_1 = \begin{pmatrix} p_{01}\\ p_{12}\\ p_{23}\\ p_{30}\end{pmatrix}, \; \mathbf{r}_2 = \begin{pmatrix} p_{02}\\ p_{13}\\ p_{20}\\ p_{31}\end{pmatrix}, \; \mathbf{r}_3 = \begin{pmatrix} p_{03}\\ p_{10}\\ p_{21}\\ p_{32}\end{pmatrix} = \Pi\mathbf{r}_1, \tag{52}$$

where $\mathbf{r}_i$ is a shorthand for $\mathbf{r}_i(\beta)$. Furthermore, we again denote the vector decomposition of any thermal state with higher temperature $\beta' \leq \beta$ as $\mathbf{p}(\beta') = \mathbf{a} + \sum_i \mathbf{b}_i$ with $\mathbf{a} := \mathbf{q}(\beta')$ and $\mathbf{b}_i := \mathbf{r}_i(\beta')$. Unitaries on the LCSs that generate the same marginals, according to Eq. (25), lead to the transformation

$$\mathbf{p} \mapsto \tilde{\mathbf{p}} = M_q\mathbf{q} + (\mathbb{1}+\Pi)M_{r_1}\mathbf{r}_1 + \frac{1}{2}(\mathbb{1}+\Pi^2)M_{r_2}\mathbf{r}_2, \tag{53}$$

where the M_{r_i} are arbitrary doubly stochastic matrices. To achieve a thermal marginal with higher temperature, one can transform each $\mathbf{r}_i$ to $\mathbf{b}_i$ as prescribed by condition i and pass the extra norm of the vector $\mathbf{q}$ as dictated by the stronger version of condition (ii). This is possible at least under some restrictions on the energy level spacings $\delta_i := E_{i+1} - E_i$. Let us phrase this statement more precisely in the following Lemma 4, a detailed proof of which is presented in Appendix A.IX.

Lemma 4. *In $d = 4$, for every choice of the set $\{E_i\}_{i=0}^3$ with $E_{i+1} \geq E_i$ and $\delta_{i+1} \leq \delta_i$ and for any $\beta' \leq \beta$, the following relations hold for $i = 1, 2$:*

$$\frac{\mathbf{q}}{\|\mathbf{q}\|} \succ \frac{\mathbb{1} + \Pi^i}{2} \frac{\mathbf{b}_i}{\|\mathbf{b}_i\|}, \tag{54}$$

$$\|\mathbf{r}_i\| \leq \|\mathbf{b}_i\|. \tag{55}$$

With Lemma 4 at hand, we are ready to state the result achieved for the $d = 4$ with this method.

Theorem 2: Existence of STUs in $d = 4$

In any 4-dimensional system, for every set of energy eigenvalues $\{E_i\}_{i=0}^3$ with $E_{i+1} \geq E_i$ and $\delta_{i+1} \leq \delta_i$, and for any $\beta' \leq \beta$, there exists a set of doubly stochastic matrices $\{M_{r_i}\}_{i=0}^2$ such that

$$\mathbf{p}(\beta') = M_q\,\mathbf{q} + (\mathbb{1} + \Pi)\, M_{r_1}\mathbf{r}_1 + \frac{1}{2}(\mathbb{1} + \Pi^2) M_{r_2}\,\mathbf{r}_2, \tag{56}$$

which implies the existence of STUs in $d = 4$ for symmetric Hamiltonians with decreasing energy gaps.

Proof. The statement follows from Lemmas 2 and 4. □

We have thus seen that the approach discussed in this section does, at least partially, generalise to local dimension 4. However, the proof of Theorem 2, which exploits the stronger version of condition (ii) fails for $E_3 \gg 1$. Note that this failure is not an artifact of even dimensions, but rather continues to persist in subsequent higher dimensions. Still, the possibility remains that above results can be generalised by proving the weaker version of condition (ii).

III.3. Geometric approach

Our attempts to generalise the approaches of Secs. III.1 and III.2 to higher dimensions have shown that the problem can be recast in terms of different sets of conditions. However, checking these conditions has proven to be increasingly complex with growing dimension, and has thus only provided partial results even for the case $d = 4$. We therefore now turn to a third approach which at least provides a complete proof for $d = 4$. This approach is centred around the geometric structure generated by doubly stochastic matrices. More specifically, recall that the Schur-Horn theorem implies that for any $v \in \mathbb{R}^n$, the set of vectors obtained by applying the set of unistochastic $n \times n$ matrices to v is a convex polytope given by the convex hull of all permutations of the entries of v, see, e.g., Ref. [31]. Here, we can apply this idea to the vectors $\mathbf{q}$ and $\mathbf{r}_i$ and the matrices M_q and M_{r_i}, respectively, to generate matching diagonal marginals according to Eq. (25). In other words, the set of all possible symmetric marginal vectors reachable by unitaries that are block-diagonal with respect to the chosen LCS decomposition is the polytope with vertices given by the set of points

$$\tilde{\mathbf{p}}^{(i_0, i_1, \ldots, i_k)} = \Pi^{(i_0)}\mathbf{q} + \sum_{n=1}^{k} (\lfloor \tfrac{2n}{d} \rfloor + 1)^{-1} (\mathbb{1} + \Pi^n)\, \Pi^{(i_n)}\mathbf{r}_n, \tag{57}$$

with $i_j \in \{1, 2, \ldots, d!\}$ for all $j = 0, \ldots, k$ with k as in Eq. (21). Here, $\Pi^{(i)}$ for $i = 1, \ldots, d!$ are the possible permutations of d elements. The question about the existence of STUs is then equivalent to asking if the curve defined by the set of points $\{\mathbf{p}(\beta') | \beta \geq \beta' \geq 0\}$ is enclosed within the polytope corresponding to the convex hull of the $(d!)^{k+1}$ points $\tilde{\mathbf{p}}^{(i_0, i_1, \ldots, i_k)}$.

To answer this question, we then proceed in the following way. First, we note that the d-component vectors $\mathbf{p} = (p_i)$ only have $d - 1$ independent entries due to the normalisation condition $\sum_{i=0}^{d-1} p_i = 1$. The problem can thus be reduced to $d-1$ dimensions by choosing coordinates $\{x_i\}_{i=0,1,\ldots,d-2}$ with

$$x_n = (n+1)p_{n+1} - \sum_{i=0}^{n} p_i, \tag{58}$$

for $n = 0, 1, \ldots, d-2$, while the additional last coordinate $x_{d-1} = -\sum_{i=0}^{d-1} p_i = -1$ is fixed by normalisation and can thus be disregarded. In these coordinates, the point obtained for infinite temperature ($\beta \to 0$) is the origin, $p(\beta \to 0) = (0, 0, \ldots, 0, -1)$, the point for vanishing temperature is $p(\beta \to \infty) = (-1, -1, \ldots, -1)$ and all thermal states lie on a continuous curve connecting these points that is strictly confined to negative coordinate regions, $x_i \leq 0\ \forall i$. However, note that there are points corresponding to reachable marginals that have positive values for some of the new coordinates.

For any given initial temperature $1/\beta$ and dimension d, a sufficient set of conditions for the inclusion of the curve defined by the points $\{\mathbf{p}(\beta') | \beta \geq \beta' \geq 0\}$ in the reachable polytope is as follows:

(I) Inclusion of the vertices

$$\begin{aligned}
\mathbf{v}_0 &:= \mathbf{p}(\beta) = (x_0(\beta), x_1(\beta), \ldots, x_{d-3}(\beta), x_{d-2}(\beta), -1),\\
\mathbf{v}_1 &:= (0, x_1(\beta), \ldots, x_{d-3}(\beta), x_{d-2}(\beta), -1),\\
\mathbf{v}_2 &:= (0, 0, x_2(\beta) \ldots, x_{d-3}(\beta), x_{d-2}(\beta), -1),\\
&\vdots\\
\mathbf{v}_{d-2} &:= (0, 0, \ldots, 0, x_{d-2}(\beta), -1),\\
\mathbf{v}_{d-1} &:= \mathbf{p}(\beta \to 0) = (0, \ldots, 0, -1),
\end{aligned} \tag{59}$$

in the polytope of achievable marginals.

(II) Inclusion of all points $\mathbf{p}(\beta')$ with $\beta' \leq \beta$ in the simplex corresponding to convex hull of the set of vertices $\{\mathbf{v}_i | i = 0, \ldots, d-1\}$.

In general, confirming condition (II) is relatively straightforward, either by proving the positivity of the partial derivatives $\frac{\partial x_m}{\partial x_n} = \left(\frac{\partial x_n}{\partial \beta}\right)^{-1} \frac{\partial x_m}{\partial \beta} \geq 0$ and $\frac{\partial^2 x_m}{\partial x_n^2} = \frac{\partial^2 x_m}{\partial \beta^2} - \left(\frac{\partial x_m}{\partial \beta}\right)\left(\frac{\partial x_n}{\partial \beta}\right)^{-1} \frac{\partial^2 x_n}{\partial \beta^2} \geq 0$ (which we show explicitly for $d = 4$ in Appendix A.X), or by showing that all points $\mathbf{p}(\beta')$ can be written as a convex combination of the vertices (59), which we prove in general for dimension d in Appendix A.X. Let us therefore state condition (II) as the following lemma:

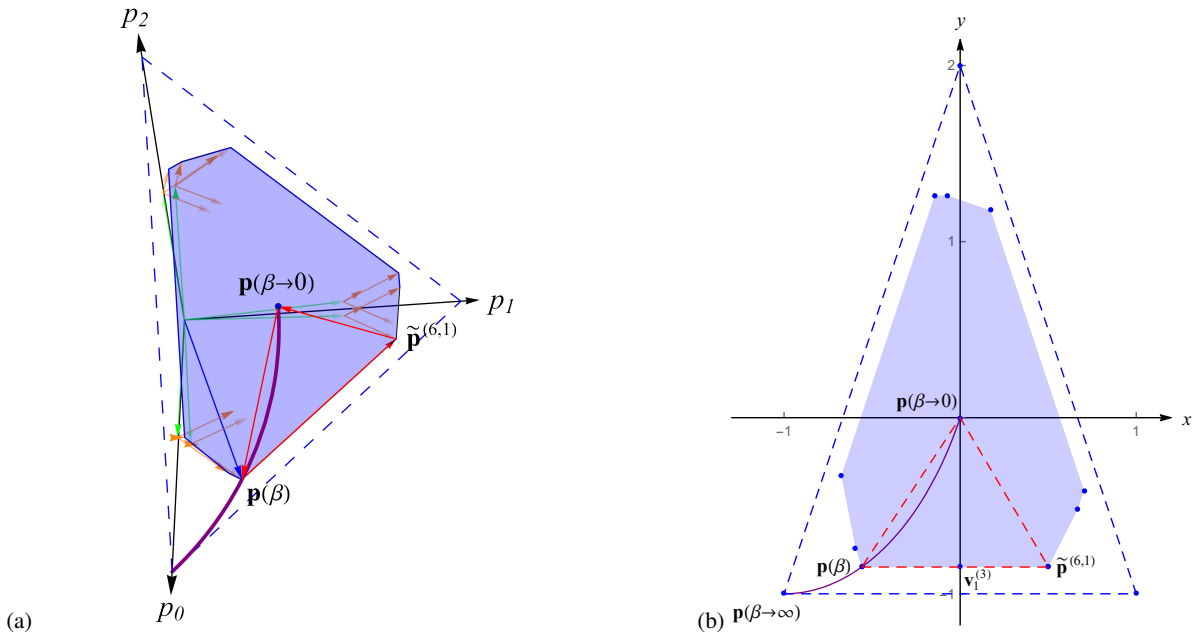

FIG. 1. **Polytope of reachable symmetric diagonal qutrit marginals**. (a) The axes show the components p_i (with $i = 0, 1, 2$) with respect to the original basis $\{\mathbf{e}_i\}_{i=0,1,2}$ and the simplex of all possible states (not necessarily reachable from a given state) is indicated by the dashed blue triangle. The parameter values chosen for the illustration are $\beta = 1.35E_1$ and $E_2 = 2E_1$. For these values, the family of thermal states is shown as a solid purple curve from $\mathbf{p}(\beta \to \infty) = (1, 0, 0)^T$ to $\mathbf{p}(\beta \to 0) = (\frac{1}{3}, \frac{1}{3}, \frac{1}{3})^T$. The initial state of the marginals is represented by the point $\mathbf{p}(\beta)$ indicated by a blue arrow. The shaded blue area shows the polytope of diagonal reduced states with $\tilde{\mathbf{p}}_A = \tilde{\mathbf{p}}_B$ that is reachable by the application of a unistochastic matrix M on $\mathbf{q}$ in combination with a circulant unistochastic matrix $\tilde{M}$ applied to $\mathbf{r}$. This polytope is the convex hull of the points obtained from combining any (out of 6 possible) permutations of q (green arrows) with cyclic permutations (out of 3 possible) of $\mathbf{r}$ (orange arrows). We have chosen to restrict to cyclic permutations on $\mathbf{r}$ here to illustrate that this is enough for $d = 3$, whereas this is no longer the case when $d = 4$. The red arrows between $\mathbf{p}(\beta)$, $\tilde{\mathbf{p}}^{(6,1)}$, and $\mathbf{p}(\beta \to 0)$ delineate a triangle, which we show contains all points $\mathbf{p}(\beta')$ that correspond to thermal states with temperatures higher than the original temperature, $\beta > \beta'$. (b) The polytope is shown in terms of the independent coordinates $x \equiv x_0 = -p_0 + p_1$ and $y \equiv x_1 = -p_0 - p_1 + 2p_2$ from Eq. (58). The vertex $\mathbf{v}_1^{(3)}$ is located at the intersection of the y axis with the line connecting $\mathbf{p}(\beta)$ and $\tilde{\mathbf{p}}^{(6,1)}$.

Lemma 5. *In the coordinates defined by Eq. (58), the curve of thermal states at inverse temperature $\beta' \leq \beta$ satisfies condition (II) for all β in all dimensions d.*

Besides satisfying condition (II), it is also easy to see that the vertices $\mathbf{v}_0$ and $\mathbf{v}_{d-1}$ from condition (I), which represent the initial thermal state and maximally mixed state in the new coordinates, are reachable for all dimensions. However, showing the inclusion of the rest of the points in (I) is increasingly difficult, due to the rapidly growing number of possible polytope vertices and the difficulty of visualizing the $(d-1)$-dimensional polytope beyond $d = 4$. In the following, we prove that condition (I) holds (at least) in the particular cases of $d = 3$ and $d = 4$. See also Fig. 1 for an illustration in dimension 3 and Appendices A.XI and A.XII for further details.

Theorem 3: Geometric approach in $d = 3$ and $d = 4$

In $d = 3$ and $d = 4$ systems, for every choice of Hamiltonians and initial inverse temperature β, the set of thermal states with $\beta' \leq \beta$ is contained within the polytope with vertices defined in Eq. (59), which proves the existence of STUs in the symmetric two-qutrit and two-ruqart cases.

Proof. For $d = 3$ we have to prove that the point $\mathbf{v}_1^{(3)} = (0, x_1(\beta), -1)$ can be reached with transformations of the type $\mathbf{p} \mapsto \tilde{\mathbf{p}} = M_q\mathbf{q} + (\mathbb{1} + \Pi)M_r\mathbf{r}$ for some doubly stochastic 3×3 matrices M_q and M_r, where $\mathbf{q}$ and $\mathbf{r}$ are as in Eq. (29). This is indeed the case, e.g., for $M_r = \mathbb{1}$ being the identity matrix and

$$M_q = \begin{pmatrix} m & 1-m & 0 \\ 1-m & m & 0 \\ 0 & 0 & 1 \end{pmatrix}, \tag{60}$$

with $m = 1 - 1/[2(p_0 + p_1)]$, which is a doubly stochastic matrix since $p_0 + p_1 \geq 1/2$ for $d = 3$. Note that, geometrically, this choice of M_q and M_r represents a convex combination of the points $\mathbf{p}(\beta)$ and $\tilde{\mathbf{p}}^{(6,1)}$ shown in Fig. (1). For this transformation, we have $\tilde{\mathbf{p}} = \sum_{i=0}^{2} \tilde{p}_i \mathbf{e}_i$, where the components with respect to the original basis $\{\mathbf{e}_j\}_{j=0,1,2}$ are $\tilde{p}_0 = \tilde{p}_1 = (p_0 + p_1)/2$ and $\tilde{p}_2 = p_2$, which means that the components of $\tilde{\mathbf{p}}$ with respect to to the new coordinates of Eq. (58) are $\tilde{\mathbf{p}} = (0, 2-3(p_0+p_1), -1)^T = (0, x_1(\beta), -1)^T = \mathbf{v}_1^{(3)}$. This concludes the proof for $d = 3$.

For $d = 4$, we have to show that the two points $\mathbf{v}_1^{(4)} = (0, x_1(\beta), x_2(\beta), -1)$ and $\mathbf{v}_2^{(4)} = (0, 0, x_2(\beta), -1)$ can be reached with transformations of the type $\mathbf{p} \mapsto \tilde{\mathbf{p}} = M_q\mathbf{q} +$

$(\mathbb{1}+\Pi)M_{r_1}\mathbf{r}_1+\frac{1}{2}(\mathbb{1}+\Pi^2)M_{r_2}\mathbf{r}_2$ for some doubly stochastic 4×4 matrices M_q, M_{r_1}, and M_{r_2}, where $\mathbf{q}$ and $\mathbf{r}_1$ are as in Eq. (52).

The point $\mathbf{v}_1^{(4)}$ can be reached with the equivalent of the transformation used to reach $\mathbf{v}_1^{(3)}$ above, that is, using $M_{r_1} = M_{r_2} = \mathbb{1}$ and

$$M_q = \begin{pmatrix} m & 1-m & 0 & 0 \\ 1-m & m & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \tag{61}$$

with $m = 1-1/[2(p_0+p_1)]$, which is again doubly stochastic since $p_0+p_1 \geq 1/2$ also holds for $d=4$.

To prove that $\mathbf{v}_2^{(4)}$ can be reached, one can see that it can be obtained as a convex combination of (at most) 5 of the vertices in Eq. (57), as we show in detail in Appendix A.XII. □

For higher dimensions, as we already mentioned, the problem becomes more and more complex, but one can try to build a recursive approach based on the above lower dimensional proofs and borrowing some ideas from the "passing on the norm" approach. In particular, we outline a possible route for such an approach for the case $d=5$ in Appendix A.XIII, where we can show the existence of STUs for $d=5$ for a subset of all possible Hamiltonians.

IV. CONCLUSIONS

We have investigated the generation of correlations in initially thermal, uncorrelated systems. For two identical d-dimensional systems, the conversion of energy into correlations as measured by the mutual information is optimal when the final state can be reached unitarily and both marginals of the final state are thermal at the same effective temperature. For any given system, the possibility of such an optimal conversion for all input energies (or desired amout of correlations) hence hinges upon the existence of symmetrically thermalizing unitaries for all initial temperatures and effective local final temperatures. This gives rise to the central question: *Is it possible to find unitaries (STUs) transforming thermal marginals to other thermal marginals with higher temperature for any local Hamiltonian?*

In asymmetric cases, where the two local Hamiltonians are different, this is generally not possible, as we have shown via constraints on the subsystem entropies. For the symmetric case (equal local Hamiltonians), we have provided a framework based on locally classical subspaces in $d\times d$-dimensional systems to address this question beyond previous partial results for equally gapped Hamiltonians [11]. In particular, we have shown that STUs exist for all (locally matching) Hamiltonians in local dimensions $d=3$ and $d=4$, and we conjecture that STUs exist in all local dimensions.

To showcase the complexity and interesting features of the problem, as well as to provide further guidance for proving (or disproving) our conjecture, we have discussed three approaches operating within our framework. Using the "majorised marginals" approach we showed for two qutrits ($d=3$) that, not only do STUs generically exist for any local Hamiltonian at any temperature, but also it is indeed possible to symmetrically reach any marginal that is majorised by the initial marginals. However, since this approach fails to be generalised to higher dimensions, we introduced two alternative approaches that we call "passing on the norm" and "geometric approach", respectively. Both allow proving the existence of STUs in the two-qutrit case. Using the "passings on the norm" approach, we were further able to show that STUs exist for $d=4$ when the local Hamiltonians satisfy specific conditions on their energy gaps, i.e., $\delta_{i+1} \leq \delta_i$. Finally, we have used the "geometric" method to prove the existence of STUs in local dimension $d=4$ for all symmetric Hamiltonians, and we formulate a set of conditions to extend this approach to higher dimensions.

Our work addresses a fundamental question in quantum thermodynamics, whether correlations can always be created energetically optimally, or not. Besides addressing a question about the conversion between thermodynamic and information-theoretic resources, the problem at hand can be considered a part of the quantum marginal problem. What kind of marginals can be unitarily reached from (are compatible with) a particular global state? The framework we put forward in terms of locally classical subspaces is more general than symmetric marginal transformations and also goes beyond mere majorisation relations of marginal eigenvalues. As such, it may also be relevant for other variants of this question, such as addressing the catalytic entropy conjecture of [34]. Just using one of the many possible such subspaces, we managed to resolve our main question for dimensions $d=3$ and $d=4$ and it would be interesting to know if all marginal eigenvalue distributions could potentially be reached by operating in locally classical subspaces only. Finally, a significant challenge lies in specializing from the creation of arbitrary correlations to generating entanglement [11, 12, 29, 35].

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APPENDICES

In these appendices, we provide detailed proofs of the lemmas supporting the main theorems, as well as additional detailed calculations and counterexamples mentioned in the main text. In Appendix A.II, we investigate the maximal amount of correlations unitarily achievable for a fixed amount of energy that can be created between two arbitrary asymmetric systems initialised in a pure state. In Appendix A.III, we propose a scheme to transform finitely many copies of bipartite thermal states with thermal marginals to states with symmetric thermal marginals at a higher temperature. In Appendix A.IV, we give a detailed proof of Lemma 1. We also discuss why Lemma 1 cannot be generalised to higher dimensions in Appendix A.V. In Appendix A.VI, we show via counterexample that it is in general not possible to map the normalised versions of the vectors $\mathbf{q}_i$ to the vectors $\mathbf{r}_i$ by doubly stochastic matrices. In Appendix A.VII, we present a detailed proof of Lemma 2 that confirms that condition (i), used in the "passing on the norm" approach, holds in general. In Appendix A.VIII, we discuss how one can show the existence of STUs via the stronger version of condition (ii). In Appendix A.IX, by proving Lemma 3 and Lemma 4, we complete the proof of the existence of STUs via this approach in $d = 3$ and under specific constraints on the energy gaps in $d = 4$. Then, we turn our attention to the geometric approach and show the monotonicity and convexity of the thermal curve in Appendix A.X. The detailed proofs of the existence of STUs using the geometric approach in dimensions 3 and 4 are presented in Appendix A.XI and Appendix A.XII, respectively. Finally, we discuss the possibility of generalising the geometric method to higher dimensions in Appendix A.XIII.

A.I. Upper bound on correlation

In this appendix, we show that if STUs exist in general, i.e., in particular for the desired temperature, they provide an upper bound for the amount of correlation that can be achieved unitarily. Using the same notation as in the main text, we are interested in solving the problem

$$\max_U S(\tilde{\varrho}_A)+S(\tilde{\varrho}_B) \quad \text{s.t. } \mathrm{Tr}(\tilde{\varrho}_A H_A)+\mathrm{Tr}(\tilde{\varrho}_B H_B) \le c. \tag{A.1}$$

This can be rewritten as

$$\begin{aligned} &\max_U S(\tilde{\varrho}_A \otimes \tilde{\varrho}_B) \\ &\text{s.t. } \mathrm{Tr}(\tilde{\varrho}_A \otimes \tilde{\varrho}_B (H_A \otimes \mathbb{1} + \mathbb{1} \otimes H_B)) \le c. \end{aligned} \tag{A.2}$$

According to Jaynes' principle, the maximum is obtained for

$$\tau_A(\bar{\beta}) \otimes \tau_B(\bar{\beta}), \tag{A.3}$$

where $\bar{\beta}$ is chosen such that

$$\mathrm{Tr}(\tau_A(\bar{\beta}) \otimes \tau_B(\bar{\beta})(H_A \otimes \mathbb{1} + \mathbb{1} \otimes H_B)) = c. \tag{A.4}$$

This solution can be found using Lagrange multipliers by considering the $n \times n$ matrix ϱ_{AB} as an vector with n^2 components. We therefore have as desired

$$\begin{aligned} &\max_U S(\tilde{\varrho}_A) + S(\tilde{\varrho}_B) \text{ s.t. } \mathrm{Tr}(\tilde{\varrho}_A H_A) + \mathrm{Tr}(\tilde{\varrho}_B H_B) \le c. \\ &\le S(\tau_A(\bar{\beta})) + S(\tau_B(\bar{\beta})), \end{aligned} \tag{A.5}$$

with $\mathrm{Tr}(\tau_A(\bar{\beta}) H_A) + \mathrm{Tr}(\tau_B(\bar{\beta}) H_B) = c$.

A.II. Maximal amount of correlation for a pure state in the asymmetric case

Here, we discuss and solve the problem of maximising the correlations under an energy constraint in the asymmetric case with an initial pure state. That is, we want to solve

$$\max_{\tilde{\varrho}_A, \tilde{\varrho}_B} S(\tilde{\varrho}_A) + S(\tilde{\varrho}_B), \tag{A.6}$$

subject to the constraint $\mathrm{Tr}(\tilde{\varrho}_A H_A) + \mathrm{Tr}(\tilde{\varrho}_B H_B) \le c$, where

$$\tilde{\varrho}_A = \sum_{i=0}^{d-1} p_i |\varphi_i^A\rangle\langle\varphi_i^A|, \quad \tilde{\varrho}_B = \sum_{i=0}^{d-1} p_i |\varphi_i^B\rangle\langle\varphi_i^B|, \tag{A.7}$$

with $p_i \ge 0$ for $i = 0, \dots, d-1$, $\sum_{i=0}^{d-1} p_i = 1$, $d = \min\{d_A, d_B\}$, and where $\{|\varphi_i^A\rangle\}_{i=0}^{d_A-1}$ and $\{|\varphi_i^B\rangle\}_{i=0}^{d_B-1}$ are orthonormal bases of $\mathcal{H}_A$ and $\mathcal{H}_B$, respectively. Without loss of generality we then assume $d = d_A$. Note that $S(\tilde{\varrho}_A) = S(\tilde{\varrho}_B)$ and $\tilde{\varrho}_B = U \tilde{\varrho}_A U^\dagger$, where we write $U = \sum_{i=0}^{d-1} |\varphi_i^B\rangle\langle\varphi_i^A|$ (in a slight abuse of notation) such that the problem may be rewritten as

$$\max_{\varrho, U} S(\varrho) \quad \text{s.t.} \quad \mathrm{Tr}[\varrho(H_A + U^\dagger H_B U)] \le c, \tag{A.8}$$

where we have dropped the tilde and subscript A on ϱ for ease of notation. To solve this problem we consider the converse problem

$$\min_{\varrho, U} \mathrm{Tr}[\varrho(H_A + U^\dagger H_B U)] \quad \text{s.t.} \quad S(\varrho) = \kappa, \tag{A.9}$$

and show that (at least a family of) optimal points of (A.9) are optimal points of (A.8). To simplify the notation further let us write

$$H_A = \sum_{i=0}^{d-1} E_i^A |i\rangle\langle i|_A \tag{A.10}$$

$$H_B = \sum_{i=0}^{d_B-1} E_i^B |i\rangle\langle i|_B, \tag{A.11}$$

with $E_i^A \le E_{i+1}^A$ and $E_i^B \le E_{i+1}^B$.

Proposition 1. *The pair* $(\varrho_{\mathrm{opt}}(\kappa), U_{\mathrm{opt}})$, *given by*

$$U_{\mathrm{opt}} := \sum_{i=0}^{d-1} |i\rangle_B \langle i|_A, \tag{A.12}$$

$$\varrho_{\mathrm{opt}}(\beta(\kappa)) := \frac{e^{-\beta(\kappa)\tilde{H}}}{\tilde{Z}}, \tag{A.13}$$

where $\tilde{Z} = \mathrm{Tr}(e^{-\beta(\kappa)\tilde{H}})$ *and* $\tilde{H} = \sum_{i=0}^{d-1}(E_i^A + E_i^B)|i\rangle\langle i|_A$, *is a solution of the minimisation in* (A.9).

Proof. Denoting the spectrum of ϱ as $\lambda_\varrho = (\lambda_0, \dots, \lambda_{d-1})$, we first show that $(\varrho_{\rm opt}(\beta(\kappa)), U_{\rm opt})$ is a solution of the following minimisation problem

$$\min_\lambda \left(\min_{\varrho \text{ s.t. } \lambda_\varrho = \lambda} \mathrm{Tr}(\varrho H_A) + \min_{U,\varrho \text{ s.t. } \lambda_\varrho = \lambda} \mathrm{Tr}(U\varrho U^\dagger H_B) \right), \tag{A.14}$$

subject to the constraint $H(\lambda) = \kappa$, where $H(\lambda)$ denotes the Shannon entropy of the probability distribution λ. Since all density matrices of a given spectrum are unitarily related we have

$$\min_{\varrho \text{ s.t. } \lambda_\varrho = \lambda} \mathrm{Tr}(\varrho H_A) = \min_V \mathrm{Tr}(V \,\mathrm{diag}\{\lambda_\varrho\} V^\dagger H_A), \tag{A.15}$$

where the minimisation on the right-hand side is over all unitaries V. The passive state with spectrum λ is well-known to solve this minimisation. Adopting the notation $v^\downarrow = (v_i^\downarrow)$ to denote the vector obtained by arranging the components of the vector $v = (v_1, \dots, v_n)$ in decreasing order, i.e., such that $v_1^\downarrow \geq v_2^\downarrow \geq \dots \geq v_n^\downarrow$, we thus have

$$\min_{\varrho \text{ s.t. } \lambda_\varrho = \lambda} \mathrm{Tr}(\varrho H_A) = \sum_{i=0}^{d-1} \lambda_i^\downarrow E_i^A. \tag{A.16}$$

For the second minimisation problem in (A.14), note that since $\{|\varphi_i^A\rangle\}_{i=0}^{d_A-1}$ and $\{|\varphi_i^B\rangle\}_{i=0}^{d_B-1}$ are orthonormal bases, the matrix representation of U can be extended to a unitary $d_B \times d_B$ matrix V. Similarly, the matrix representation of ϱ can be extended to a positive $d_B \times d_B$ matrix $\bar\varrho$ by padding it with zeroes. With this we have

$$\min_{U,\varrho \text{ s.t. } \lambda_\varrho = \lambda} \mathrm{Tr}(U\varrho U^\dagger H_B) \geq \min_{V,\bar\varrho \text{ s.t. } \lambda_{\bar\varrho} = (\lambda, 0, \dots, 0)} \mathrm{Tr}(V\bar\varrho V^\dagger H_B). \tag{A.17}$$

Again the passive state with spectrum $(\lambda, 0, \dots, 0)$ solves the right-hand side of (A.17) and we obtain

$$\min_{V,\bar\varrho \text{ s.t. } \lambda_{\bar\varrho} = (\lambda, 0, \dots, 0)} \mathrm{Tr}(V\bar\varrho V^\dagger H_B) = \sum_{i=0}^{d-1} \lambda_i^\downarrow E_i^B. \tag{A.18}$$

This solution is in fact also an attainable solution of the left-hand side of (A.17). Hence

$$\min_{U,\varrho \text{ s.t. } \lambda_\varrho = \lambda} \mathrm{Tr}(U\varrho U^\dagger H_B) = \sum_{i=0}^{d-1} \lambda_i^\downarrow E_i^B. \tag{A.19}$$

We have therefore reduced the minimisation problem of (A.14) to solving

$$\min_\lambda \sum_{i=0}^{d-1} \lambda_i^\downarrow (E_i^A + E_i^B), \text{ s.t. } H(\lambda) = \kappa, \ \sum_i \lambda_i^\downarrow = 1. \tag{A.20}$$

This problem, in turn, can be solved by means of Lagrange multipliers, which yields

$$\lambda_i^\downarrow = \frac{e^{-\beta(E_i^A + E_i^B)}}{\sum_{i=0}^{d-1} e^{-\beta(E_i^A + E_i^B)}}, \tag{A.21}$$

which is precisely what is delivered by the solution $(\varrho_{\rm opt}(\beta(\kappa)), U_{\rm opt})$.

We can further check that for every $\kappa \in [0, \ln(d)]$ there exists a unique $\beta \in [0, \infty]$ such that $H(\lambda_{\varrho_{\rm opt}}) = S(\varrho_{\rm opt}(\beta)) = \kappa$, i.e., that the notation $\beta(\kappa)$ is well defined and can be understood as a function. This can be seen from

$$S(\varrho_{\rm opt}(0)) = \ln(d), \tag{A.22}$$

$$S(\varrho_{\rm opt}(\infty)) = 0, \tag{A.23}$$

$$\frac{d}{d\beta} S(\varrho_{\rm opt}(\beta)) = \beta(\mathrm{Tr}(\varrho\tilde H)^2 - \mathrm{Tr}(\varrho\tilde H^2)) < 0, \ \forall \beta \in (0, \infty). \tag{A.24}$$

Strictly speaking, the last line is not valid when $\tilde H \propto \mathbb{1}$, but in that case one can straightforwardly check that $(U_{\rm opt}, \frac{\mathbb{1}}{d})$ solves our original problem (A.8) for any allowed c. We hence (tacitly) discard it from the start.

Having established this fact about $(\varrho_{\rm opt}(\beta(\kappa)), U_{\rm opt})$, let (ϱ, U) be such that $S(\varrho) = \kappa$. Then ϱ has some spectrum $\{\mu_0, \dots, \mu_{d-1}\}$. From the above we thus have

$$\begin{aligned} \mathrm{Tr}(\varrho H_A) + \mathrm{Tr}(U\varrho U^\dagger H_B) &\geq \sum_{i=0}^{d-1} \mu_i^\downarrow E_i^A + \sum_{i=0}^{d-1} \mu_i^\downarrow E_i^B \\ &\geq \sum_{i=0}^{d-1} \frac{e^{-\beta(\kappa)(E_i^A + E_i^B)}}{\sum_{i=0}^{d-1} e^{-\beta(\kappa)(E_i^A + E_i^B)}} (E_i^A + E_i^B) \\ &= \mathrm{Tr}(\varrho_{\rm opt}(\beta(\kappa)) H_A) + \mathrm{Tr}(U_{\rm opt}\varrho_{\rm opt}(\beta(\kappa)) U_{\rm opt}^\dagger H_B), \end{aligned} \tag{A.25}$$

which proves our claim, because $S(\varrho_{\rm opt}(\beta(\kappa))) = \kappa$. □

Now let us define the function

$$f(\kappa) = \mathrm{Tr}(\varrho_{\rm opt}(\beta(\kappa)) H_A) + \mathrm{Tr}(U_{\rm opt}\varrho_{\rm opt}(\beta(\kappa)) U_{\rm opt}^\dagger H_B). \tag{A.26}$$

We can then establish the following proposition.

Proposition 2. *If $f(\kappa_1) < f(\kappa_2)$ then $\kappa_1 < \kappa_2$.*

The above is saying that if f has an inverse then that inverse is strictly monotonically increasing. This is indeed how the proof proceeds.

Proof. First, note that we have

$$\frac{d}{d\kappa} f(\kappa) = \frac{d}{d\beta} f(\beta) \frac{d\beta}{d\kappa} = \frac{d}{d\beta} f(\beta) \left(\frac{d}{d\beta} S(\varrho_{\rm opt}(\beta) \right)^{-1}. \tag{A.27}$$

Further, we have already seen that

$$\frac{d}{d\beta} S(\varrho_{\rm opt}(\beta) = \beta(\mathrm{Tr}(\varrho\tilde H)^2 - \mathrm{Tr}(\varrho\tilde H^2)) < 0. \tag{A.28}$$

Similarly one calculates

$$\frac{d}{d\beta} f(\beta) = (\mathrm{Tr}(\varrho\tilde H)^2 - \mathrm{Tr}(\varrho\tilde H^2)) < 0. \tag{A.29}$$

So for $\kappa \in (0, \ln(d))$ we have

$$\frac{d}{d\kappa} f(\kappa) = \frac{1}{\beta} > 0. \tag{A.30}$$

This proves that f is strictly monotonically increasing. It therefore has an inverse f^{-1} that is also strictly monotonically increasing which proves our claim. □

We are now ready to solve the maximisation in (A.8).

Proposition 3. *There is a (unique) β such that $(\varrho_{\rm opt}(\beta), U_{\rm opt})$ is a solution of (A.8).*

Proof. First, if $c \geq \frac{1}{d}\sum_{i=0}^{d-1} E_i^A + E_i^B$, then $(\varrho_{\rm opt}(0), U_{\rm opt})$ is the sought after solution, because

$$H((\tfrac{1}{d},\ldots,\tfrac{1}{d})) = S(\varrho_{\rm opt}(0)) \geq S(\varrho)\ \forall\, \varrho\,, \tag{A.31}$$

and $\mathrm{Tr}(\varrho_{\rm opt}(0)\tilde{H}) \leq c$. Second, if $c \leq \frac{1}{d}\sum_{i=0}^{d-1} E_i^A + E_i^B$ then

$$\frac{d}{d\beta} f(\beta) < 0 \tag{A.32}$$

ensures that there exists a (unique) β, say $\bar{\beta}$, such that

$$f(\bar{\beta}) = \mathrm{Tr}(\varrho_{\rm opt}(\bar{\beta})\tilde{H}) = c. \tag{A.33}$$

Let us now prove that $(\varrho_{\rm opt}(\bar{\beta}), U_{\rm opt})$ is the desired solution in this case. Consider a pair (ϱ_1, U_1) such that

$$\mathrm{Tr}(\varrho_1 H_A) + \mathrm{Tr}(U_1\varrho_1 U_1^\dagger H_B) \leq c = \mathrm{Tr}(\varrho_{\rm opt}(\bar{\beta})\tilde{H}). \tag{A.34}$$

Let us further consider $\varrho_{\rm opt}(\beta(\kappa))$, the solution of (A.9) and choose $\kappa = S(\varrho_1)$. Then it holds that

$$\begin{aligned}\mathrm{Tr}(\varrho_{\rm opt}(\beta(\kappa))\tilde{H}) &\leq \mathrm{Tr}(\varrho_1 H_A) + \mathrm{Tr}(U_1\varrho U_1^\dagger H_B)\\ &\leq \mathrm{Tr}(\varrho_{\rm opt}(\bar{\beta})\tilde{H}),\end{aligned} \tag{A.35}$$

which, using Proposition (2), implies

$$S(\varrho_{\rm opt}(\beta(\kappa)) \leq S(\varrho_{\rm opt}(\bar{\beta})). \tag{A.36}$$

But $S(\varrho_{\rm opt}(\beta(\kappa)) = S(\varrho_1)$ such that

$$S(\varrho_1) \leq S(\varrho_{\rm opt}(\bar{\beta})). \tag{A.37}$$

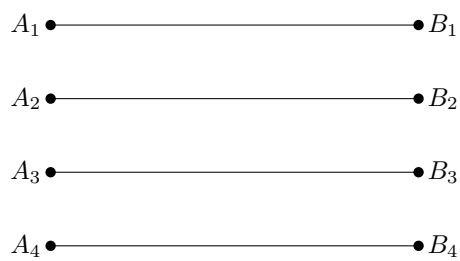
□

A.III. Correlating finitely many copies

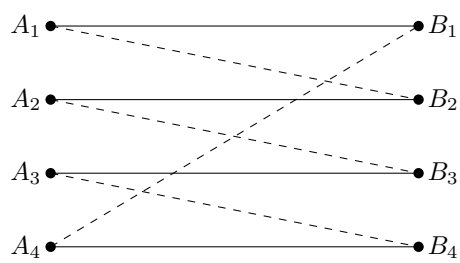
Here we discuss a protocol that can be applied to n copies of the initial state, i.e., $\tau_A(\beta)^{\otimes n} \otimes \tau_B(\beta)^{\otimes n}$, to increase the temperature of the marginals in cases where STUs for single copies exist for small temperature differences, but are not attainable for larger temperature differences. For n copies, the protocol consists of n consecutive steps. In each step, a fixed STU achieving some finite temperature increase is applied to different LCSs that correspond to particular pairs of subsystems, such that for $i, j = 1,\ldots,n$, each subsystem A_i interacts with one and only one subsystem B_j, and no subsystem interacts again with a subsystem it has previously interacted with. This ensures that all marginals are left diagonal and thermal after each step, and leads to a step-wise increase in the temperature of the marginals. In particular, in the jth step the unitary is applied to the subsystems A_i and $B_{i+j-1 \bmod(n)}$. This construction guarantees that the tensor product structure of thermal states is preserved for each pair, i.e., $\tau_A(\beta_j) \otimes \tau_B(\beta_j)$ where β_j denotes the effective local inverse temperature after the jth step. Furthermore, this means that in each step the STU can be applied, as the marginals are in a thermal state with the same temperature and moreover, product to each other. While this protocol ensures that the desired structures are preserved, the necessary conditions for the protocol to achieve arbitrary final temperatures are still part of ongoing research. In general the question remains, whether it is possible to reach any arbitrary temperature difference, $\beta' < \beta$, within finitely many steps. This discussion contrasts the proof of the existence of STUs in the asymptotic case, discussed in Sec. II.2.3.

To illustrate the protocol, let us illustrate the case of $n = 4$ copies here. In the following, each dot reprsents a subsystem and the application of STUs is denoted by lines connnecting pairs of subsystems. Initially one is confronted with the following situation.

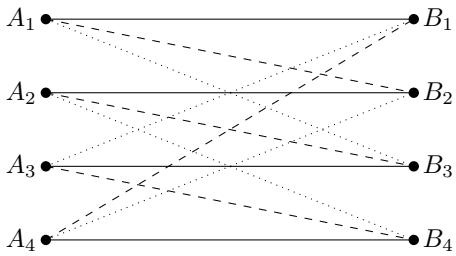
In the first step, we connect subsystems corresponding to the same copy, i.e., A_i and B_i.



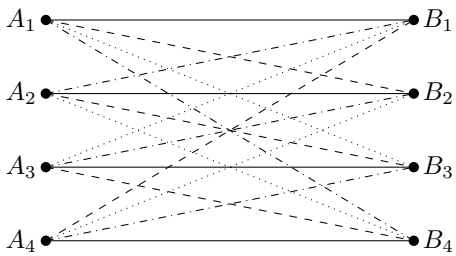
As specified, we are now in the situation where we have slightly decreased the inverse temperature of the marginals to $\tau(\beta_1)$. We now apply a unitary to the subsystems A_i and $B_{i+1 \bmod(n)}$, further decreasing the inverse temperature of the marginals.



In the third step, the subsystems A_i and $B_{i+2\,\mathrm{mod}(n)}$ are connected.



Lastly, we connect A_i and $B_{i+3\,\mathrm{mod}(n)}$ as prescribed.



We have now used the four copies of the initial state, to increase the correlations between the two sides step by step, while retaining the product structure of the thermal marginals, arriving at the final reduced states $\rho_{A/B} = \tau(\beta_4)^{\otimes 4}$ for the subsystems A and B.

A.IV. Proof of Lemma 1

In this section, we will first show that for any 3×3 doubly stochastic matrix M, there exists another 3×3 doubly stochastic matrix $\tilde{M}$ such that $M(\mathbb{1}+\Pi) = (\mathbb{1}+\Pi)\tilde{M}$, a statement we called Lemma 1 in the main text.

Proof of Lemma 1. In general, all doubly stochastic matrices can be written as convex combinations of permutation matrices as the following

$$M = \sum_{i=1}^{d!} \alpha_i \Pi^{(i)}, \tag{A.38}$$

where $\Pi^{(i)}$ indicate permutation matrices in dimension d, and $\sum_i \alpha_i = 1$. Since we need to show that the statement is true for any doubly stochastic matrix M, it is sufficient to prove that the statement is true for M being a permutation matrix. In the following we will just focus on dimension 3. In this dimension there are $3! = 6$ permutation matrices where we collect the cyclic permutations $\Pi_{\mathrm{C}}^{(1)} = \mathbb{1}$, $\Pi_{\mathrm{C}}^{(2)} = \Pi$, $\Pi_{\mathrm{C}}^{(3)} = \Pi^2$, and anticyclic permutations

$$\Pi_{\mathrm{AC}}^{(1)} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \Pi_{\mathrm{AC}}^{(2)} = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \Pi_{\mathrm{AC}}^{(3)} = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 1 & 0 \\ 1 & 0 & 0 \end{pmatrix}, \tag{A.39}$$

in a particular order, i.e.,

$$\begin{aligned} &\Pi^{(1)} = \Pi_{\mathrm{C}}^{(1)},\ \Pi^{(2)} = \Pi_{\mathrm{AC}}^{(1)},\ \Pi^{(3)} = \Pi_{\mathrm{C}}^{(3)}, \\ &\Pi^{(4)} = \Pi_{\mathrm{AC}}^{(3)},\ \Pi^{(5)} = \Pi_{\mathrm{C}}^{(2)},\ \Pi^{(6)} = \Pi_{\mathrm{AC}}^{(2)}. \end{aligned} \tag{A.40}$$

where $\Pi^{(1)}$, $\Pi^{(5)}$, and $\Pi^{(3)}$ trivially commute with $\Pi^{(5)} = \Pi$. It is further straightforward to show that

$$\begin{aligned} \Pi^{(6)}(\mathbb{1}+\Pi^{(5)}) &= (\mathbb{1}+\Pi^{(5)})\Pi^{(2)} \\ \Pi^{(2)}(\mathbb{1}+\Pi^{(5)}) &= (\mathbb{1}+\Pi^{(5)})\Pi^{(4)} \\ \Pi^{(4)}(\mathbb{1}+\Pi^{(5)}) &= (\mathbb{1}+\Pi^{(5)})\Pi^{(6)}. \end{aligned} \tag{A.41}$$

For any 3×3 doubly stochastic matrix M given in the form of Eq. (A.38), by using Eq. (A.41), one may thus find a 3×3 doubly stochastic matrix $\tilde{M}$ of the form

$$\begin{aligned} \tilde{M} = \alpha_1 \Pi^{(1)} + \alpha_3 \Pi^{(3)} + \alpha_5 \Pi^{(5)} \\ + \alpha_2 \Pi^{(4)} + \alpha_4 \Pi^{(6)} + \alpha_6 \Pi^{(2)}, \end{aligned} \tag{A.42}$$

which satisfies the required condition. □

A.V. Generalisation of the majorised marginals approach

One of the approaches to investigating the existence of the unitaries mentioned in Question 1 is to generalise the 'majorised marginals approach' of Sec. III.1. This approach appears to be promising because of its simplicity and utility. For such a generalisation to higher dimensions to be successful, Eq. (57) would demand to prove the following claim.

Claim 1. *For any $d\times d$ doubly stochastic matrix M, with $d\geq 4$, there exists a doubly stochastic matrix $\tilde{M}$ such that*

$$M(\mathbb{1}+\Pi^i) = (\mathbb{1}+\Pi^i)\tilde{M}_i \quad \forall i \in \{1,\ldots,\lfloor \tfrac{d}{2} \rfloor\}. \tag{A.43}$$

Note that by permuting the chosen basis vectors in a particular manner, $(\mathbb{1}+\Pi^i)$ can always be written in the form of $(\mathbb{1}+\Pi)$. Additionally, we know that such a transformation of the basis transforms any doubly stochastic matrix to another doubly stochastic matrix. Since Eq. (A.43) should be true for any doubly stochastic matrix M, these two facts imply that Eq. (A.43) can be reduced to showing that for any doubly stochastic $d\times d$ matrix M there exists a doubly stochastic matrix $\tilde{M}$ such that

$$M(\mathbb{1}+\Pi) = (\mathbb{1}+\Pi)\tilde{M}. \tag{A.44}$$

Using a counterexample, we show that Claim 1 does not hold in general for dimensions $d\geq 4$. In particular, we construct a counterexample in dimension 4.

Counterexample to Claim 1. Let

$$M = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0. \end{pmatrix}. \tag{A.45}$$

As we will now show, there exists no doubly stochastic $\tilde{M}$ such that $M(\mathbb{1}+\Pi) = (\mathbb{1}+\Pi)\tilde{M}$. To do so, we first calculate

$$M(\mathbb{1}+\Pi) = \begin{pmatrix} 1 & 0 & 0 & 1 \\ 0 & 0 & 1 & 1 \\ 1 & 1 & 0 & 0 \\ 0 & 1 & 1 & 0 \end{pmatrix}. \tag{A.46}$$

We are then interested in determining whether this can be equal to $(\mathbb{1}+\Pi)\tilde{M}$ for some $\tilde{M}$, with elements $m_{ij} \geq 0$, for $i,j = 0,\dots,3$ such that each column and row sums to 1. From the first row of $(\mathbb{1}+\Pi)\tilde{M}$, we obtain $m_{00}+m_{30} = 1$, which implies $m_{10} = m_{20} = 0$, $m_{01} = m_{31} = 0$, and $m_{02} = m_{32} = 0$, and further we obtain $m_{03}+m_{33} = 1$, which implies $m_{13} = m_{23} = 0$. Thus, $\tilde{M}$ must be of the form

$$\tilde{M} = \begin{pmatrix} m_{00} & 0 & 0 & m_{03} \\ 0 & m_{11} & m_{12} & 0 \\ 0 & m_{21} & m_{22} & 0 \\ m_{30} & 0 & 0 & m_{33} \end{pmatrix}. \tag{A.47}$$

Then, from the second row of $(\mathbb{1}+\Pi)\tilde{M}$ we obtain $m_{00} = 0$, implying $m_{30} = 1$, as well as $m_{11} = 0$, which implies $m_{21} = 1$. Moreover, we have $m_{12} = 1$, which means $m_{22} = 0$, while $m_{03} = 1$ suggests $m_{33} = 0$. We hence have

$$\tilde{M} = \begin{pmatrix} 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{pmatrix}. \tag{A.48}$$

But since $((\mathbb{1}+\Pi)\tilde{M})_{31} = 0 \neq (M(\mathbb{1}+\Pi))_{31} = 0$ we arrive at a contradiction.

Since Lemma 1 does not hold in general, one can try to relax it to a weaker statement which is still suitable for our purposes. One way to do so is to demand that an equivalent of Eq. (A.44) holds only when applied to (certain) vectors, in the spirit of the observation that although the set of doubly stochastic matrices does not coincide with that of unistochastic ones, given a vector $\mathbf{v}$ with nonnegative components and a doubly stochastic matrix M, there is yet a unistochastic M_U such that $M\mathbf{v} = M_U\mathbf{v}$. Thus, we investigate whether the following statement is true or not:

Claim 2. *Given a vector $\mathbf{v}$ of nonnegative components and a doubly stochastic matrix M, there exists a doubly stochastic matrix $\tilde{M}_v$, which may depend on $\mathbf{v}$, such that*

$$M(\mathbb{1}+\Pi)\mathbf{v} = (\mathbb{1}+\Pi)\tilde{M}_v\mathbf{v}. \tag{A.49}$$

The relaxation being clearly that now $\tilde{M}$ is allowed to depend on $\mathbf{v}$. Unfortunately, the previous counterexample carries over to Claim 2, as we shall see.

Counterexample to Claim 2. Let us consider M as in Eq. (A.45) and let $\mathbf{v} = (1,0,0,0)^T$. Then we have

$$M(\mathbb{1}+\Pi)\mathbf{v} = (1,\ 0,\ 1,\ 0)^T. \tag{A.50}$$

We then want to know if this equals $(\mathbb{1}+\Pi)\tilde{M}_v\mathbf{v}$ for some $\tilde{M}_v$ with elements $m_{ij} \geq 0$, $i,j = 0,\dots,3$ such that each column and row sums to 1. We obtain

$$(\mathbb{1}+\Pi)\tilde{M}_v\mathbf{v} = \begin{pmatrix} m_{00}+m_{30} \\ m_{00}+m_{10} \\ m_{10}+m_{20} \\ m_{20}+m_{30} \end{pmatrix}. \tag{A.51}$$

Comparing this with Eq. (A.50), we have $m_{00}+m_{30} = m_{10}+m_{20}$ and $m_{20}+m_{30} = m_{00}+m_{10}$, which yields $m_{30} = m_{10}$, and concurrently $m_{00}+m_{30} = 0$ and $m_{00}+m_{30} = 1$, which means we arrive at a contradiction.

A.VI. Counterexample for majorisation relations

Here, via counterexample, we show that the following claim (discussed in Sec. III.2) does not hold in general:

Claim 3. *The relation $\frac{\mathbf{q}}{\|\mathbf{q}\|} \succ \frac{\mathbf{b}_i}{\|\mathbf{b}_i\|}$ holds $\forall\ d \geq 2$ and $\beta' \leq \beta$.*

Counterexample to Claim 3. Consider the case where the last eigenvalue of the local Hamiltonian is infinitely large, $E_{d-1} \to \infty$. In this case, we know that for a thermal state with any finite temperature, the corresponding probability weight vanishes, $p_{d-1}(\beta) \to 0$. Due to the definitions of the vectors $\mathbf{q}$ and $\mathbf{r}_i$ [Eq. (17)], it is clear that $p_{d-1}(\beta)$ contributes to one element of the vector $\mathbf{q}$, i.e., to $(\mathbf{q})_{d-1} = p_{d-1}^2$, and two elements of the vector $\mathbf{r}_i$, $(\mathbf{r}_i)_{d-1-i} = p_{d-1-i}\,p_{d-1}$ and $(\mathbf{r}_i)_{d-1} = p_{d-1}p_{d-1+i}$. Hence, the vectors $\mathbf{q}$ and $\mathbf{b}_i = \mathbf{r_i}(\beta')$ have $d-1$ and $d-2$ nonzero elements, respectively. Now recalling from majorisation theory that no vector can be majorised by a vector with higher rank one can see that Claim 3 does not hold.

A.VII. Proof of Lemma 2

Here, we present the proof of Lemma 2 for any d-dimensional system, which ensures that condition (i) holds in general.

Proof of Eq. (48) in Lemma 2. To prove that Eq. (48) holds, we need to show that for any $\beta > \beta'$, $g(\beta) := \|\mathbf{q}\|$ is greater than $g(\beta') = \|\mathbf{a}\|$ in d-dimensional systems. To achieve this, one may use the positivity of the first derivative of the function $g(\beta)$. We therefore calculate

$$\begin{aligned} \partial_\beta\|\mathbf{q}\| &= \partial_\beta\Big(\frac{\sum_i e^{-2\beta E_i}}{\sum_{m,n} e^{-\beta(E_m+E_n)}}\Big) \\ &= \tfrac{1}{z(\beta)^4}\sum_{i,m,n}(E_m+E_n-2E_i)e^{-\beta(2E_i+E_m+E_n)} \\ &= \tfrac{2}{z(\beta)^3}\sum_{i,m}(E_m-E_i)e^{-\beta(2E_i+E_m)} \\ &= \tfrac{2}{z(\beta)^3}\sum_{m>i}\big[(E_m-E_i)e^{-\beta(2E_i+E_m)} \\ &\quad + (E_i-E_m)e^{-\beta(E_i+2E_m)}\big] \\ &= \tfrac{2}{z(\beta)^3}\sum_{m>i}(E_m-E_i)(e^{-\beta(2E_i+E_m)} - e^{-\beta(E_i+2E_m)}). \end{aligned} \tag{A.52}$$

Without loss of generality, we have ordered the energy eigenvalues in increasing order, $E_{i+1} \geq E_i$. Then, for any pair (m,i) with $m > i$, we know that $(e^{-\beta(2E_i+E_m)} - e^{-\beta(E_i+2E_m)})$ is nonnegative, implying $\partial_\beta \|\mathbf{q}\| \geq 0$. Also note that the last inequality is strict unless $E_i = E_m$ for all $m > i$ implying $H \propto \mathbb{1}$ for which the problem is already solved. For all practical purposes, the inequality is therefore strict. □

Proof of Eq. (49) *in Lemma* 2. Using our convention of LCSs, see Sec. II.2.1, we have

$$\mathbf{r}_i := \sum_{j=0}^{d-1} p_{j\,j+i}\,\mathbf{e}_j \quad \text{and} \quad \mathbf{b}_i := \sum_{j=0}^{d-1} p'_{j\,j+i}\,\mathbf{e}_j, \tag{A.53}$$

where $p_{j\,j+i}(\beta) := e^{-\beta(E_j+E_{j+i})}/\mathcal{Z}(\beta)^2$ and $p'_{j\,j+i} := p_{j\,j+i}(\beta')$. We then calculate

$$\begin{aligned}\frac{\mathbf{r}_i}{\|\mathbf{r}_i\|} &= \sum_{j=0}^{d-1} \frac{p_{j,j+i}}{\sum_{k=0}^{d-1} p_{k\,k+i}}\,\mathbf{e}_j = \sum_{j=0}^{d-1} \frac{e^{-\beta(E_j+E_{j+i})}}{\sum_{k=0}^{d-1} e^{-\beta(E_k+E_{k+i})}}\,\mathbf{e}_j \\ &= \sum_{j=0}^{d-1} \frac{e^{-\beta A_j}}{\sum_{k=0}^{d-1} e^{-\beta A_k}}\,\mathbf{e}_j,\end{aligned} \tag{A.54}$$

where $A_j := E_j + E_{j+i}$ for all $j = 0,\ldots,d-1$. The last expression is nothing else than the vectorised form of a thermal state at inverse temperature β with respect to the Hamiltonian $\sum_{j=0}^{d-1} A_j\,|j\rangle\langle j|$. Since the vector of diagonal entries of any thermal state majorises the vectorised diagonal of any thermal state (with respect to the same Hamiltonian) with lower inverse temperature $\beta' < \beta$, which is nothing else than $\mathbf{b}_i/\|\mathbf{b}_i\|$, Eq. (49) holds, which concludes the proof. □

A.VIII. The existence of STUs with the stronger version of condition (ii)

In this section, we will show how one can prove the existence of the STUs if condition (i) and the stronger version of condition (ii) from Sec. III.2 hold.

condition (i) ensures that there exist doubly stochastic matrices M_{r_i} which transform $\mathbf{r}_i/\|\mathbf{r}_i\|$ to $\mathbf{b}_i/\|\mathbf{b}_i\|$ and also $\mathbf{q}/\|\mathbf{q}\|$ to $\mathbf{a}/\|\mathbf{a}\|$, i.e.,

$$M_{r_i}\frac{\mathbf{r}_i}{\|\mathbf{r}_i\|} = \frac{\mathbf{b}_i}{\|\mathbf{b}_i\|} \quad \text{for} \quad i = 0,\ldots,k. \tag{A.55}$$

Using Eqs. (25) and (A.55), the marginals then become

$$\tilde{\mathbf{p}} = M_q\mathbf{q} + \sum_{i=1}^{k}(\lfloor \tfrac{2i}{d} \rfloor + 1)^{-1}(\mathbb{1} + \Pi^i)\frac{\|\mathbf{r}_i\|}{\|\mathbf{b}_i\|}\mathbf{b}_i. \tag{A.56}$$

To reach a thermal state with higher temperature, one can compensate the required norm of $(\mathbb{1} + \Pi^i)\,\mathbf{b}_i$ by extra norm from the vector $\mathbf{q}$. To achieve that, we need to have $\|\mathbf{q}\| \geq \|\mathbf{a}\|$, which is ensured by condition (i). In addition, another requirement for shifting the norm is the existence of doubly stochastic matrices $M_{q\to 2b_i}$ to transform $\mathbf{q}/\|\mathbf{q}\|$ to $(\mathbb{1}+\Pi^i)\,\mathbf{b}_i/2\|\mathbf{b}_i\|$. Due to the HLP theorem, condition (ii) ensures that such matrices exist. Therefore, M_q can be written as

$$M_q = \alpha_0\, M_{q\to a} + \sum_{i=1}^{k} \alpha_i\, M_{q\to 2b_i}, \tag{A.57}$$

where $M_{q\to a}$ and the $M_{q\to b_i}$ are doubly stochastic matrices that map $\mathbf{q}$ to $\mathbf{a}$ and $\mathbf{q}$ to each of the $(\mathbb{1} + \Pi^i)\,\mathbf{b}_i/2\|\mathbf{b}_i\|$, respectively. Applying M_q to the vector $\mathbf{q}$ in Eq. (A.56), we have

$$\tilde{\mathbf{p}} = \alpha_0\frac{\|\mathbf{q}\|}{\|\mathbf{a}\|}\mathbf{a} + \sum_{i=1}^{k}(\alpha_i\frac{\|\mathbf{q}\|}{2\|\mathbf{b}_i\|} + (\lfloor \tfrac{2i}{d} \rfloor + 1)^{-1}\frac{\|\mathbf{r}_i\|}{\|\mathbf{b}_i\|})(\mathbb{1} + \Pi)\mathbf{b}_i. \tag{A.58}$$

In order to obtain a thermal state at inverse temperature $\beta' \leq \beta$, one must choose the coefficients α_i as

$$\alpha_0 = \frac{\|\mathbf{a}\|}{\|\mathbf{q}\|}, \quad \alpha_i = 2(\lfloor \tfrac{2i}{d} \rfloor + 1)^{-1}\frac{(\|\mathbf{b}_i\| - \|\mathbf{r}_i\|)}{\|\mathbf{q}\|}. \tag{A.59}$$

Using the fact that convex combinations of doubly stochastic matrices are doubly stochastic matrices, to ensure that M_q is indeed a doubly stochastic matrix, one needs to show that the coefficients α_i as given by Eq. (A.59) are positive and sum to one. The inequality $\|\mathbf{b}_i\| \geq \|\mathbf{r}_i\|$ in condition (ii) ensures their positivity. Furthermore by using that $\|\mathbf{p}\| = \|\tilde{\mathbf{p}}\|$ we have that

$$\|\mathbf{p}\| = \|\mathbf{q}\| + 2\sum_{i=1}^{k}(\lfloor \tfrac{2i}{d} \rfloor + 1)^{-1}\|\mathbf{r}_i\| = \|\mathbf{a}\| + 2\sum_{i=1}^{k}(\lfloor \tfrac{2i}{d} \rfloor + 1)^{-1}\|\mathbf{b}_i\|, \tag{A.60}$$

which shows that $\alpha_0 + \sum_{i=1}^{k}\alpha_i = 1$, concluding the proof. □

A.IX. Proofs of Lemma 3 and Lemma 4

Here we present detailed proofs of Lemma 3 and Lemma 4.

Proof of Lemma 3. To prove $\frac{\mathbf{q}}{\|\mathbf{q}\|} \succ \frac{(\mathbb{1}+\Pi)b}{2\|\mathbf{b}\|}$, we can employ the following two majorisation relations:

$$\frac{\mathbf{q}}{\|\mathbf{q}\|} \succ \frac{(\mathbb{1} + \Pi)\mathbf{r}}{2\|\mathbf{r}\|}, \tag{A.61}$$

$$\frac{(\mathbb{1} + \Pi)\mathbf{r}}{2\|\mathbf{r}\|} \succ \frac{(\mathbb{1} + \Pi)\mathbf{b}}{2\|\mathbf{b}\|}. \tag{A.62}$$

If we show that these relations are true for any $\beta' \leq \beta$, we can say that our statement is proven.

To prove the relation in (A.61), we need to show that the greatest/smallest entry of $\mathbf{q}/\|\mathbf{q}\|$ is greater/smaller than the greatest/smallest entry of $\frac{\mathbb{1}+\Pi}{2}\frac{\mathbf{r}}{\|\mathbf{r}\|}$. That is, we first need to check that

$$\frac{p_{00}}{p_{00} + p_{11} + p_{22}} \geq \frac{p_{01} + p_{20}}{2(p_{01} + p_{12} + p_{20})}. \tag{A.63}$$

Disregarding the trivial case where $p_0 = 1$, and $p_1 = p_2 = 0$, this inequality can be transformed to

$$\begin{aligned}&e^{-\beta E_1} + 2e^{-\beta E_1}e^{-\beta E_2} + e^{-\beta E_2} \\ &\quad \geq (e^{-\beta E_1} + e^{-\beta E_2})(e^{-2\beta E_1} + e^{-2\beta E_2})\end{aligned} \tag{A.64}$$

This inequality indeed holds, since the left-hand side is larger than (or equal to) $(e^{-\beta E_1}+e^{-\beta E_2})^2$, which, in turn, is larger or equal to the righ-hand side of (A.64).

Then, second, we need to check the inequality

$$\frac{p_{22}}{p_{00}+p_{11}+p_{22}} \leq \frac{p_{20}+p_{12}}{2(p_{01}+p_{12}+p_{20})}, \tag{A.65}$$

where the relevant case (i.e., for $p_1 \neq 1$ such that at least $p_1 \neq 0$) can be rewritten as

$$\begin{aligned} &e^{-2\beta E_2}(e^{-\beta(E_1+E_2)}+2e^{-\beta E_1}+e^{-\beta E_2}) \\ &\leq (e^{-\beta E_2}+e^{-\beta(E_1+E_2)})(1+e^{-2\beta E_1}). \end{aligned} \tag{A.66}$$

This second inequality holds since the right-hand side of (A.66) is larger than (or equal to) $(e^{-\beta E_2}+e^{-\beta(E_1+E_2)})^2$, which, in turn, is larger or equal to the righ-hand side of (A.66).

To prove Eq. (A.62), we show that the greatest entry of $\frac{(\mathbb{1}+\Pi)\mathbf{r}}{2\|\mathbf{r}\|}$ is monotonically increasing with β, and that its smallest entry is monotonically decreasing with β. For the former we calculate

$$\begin{aligned} \partial_\beta \frac{p_{01}+p_{20}}{2(p_{01}+p_{12}+p_{20})} &= \partial_\beta \frac{e^{-\beta E_1}+e^{-\beta E_2}}{2(e^{-\beta E_1}+e^{-\beta(E_1+E_2)}+e^{-\beta E_2})} \\ &= \frac{E_2 e^{-\beta(2E_1+E_2)}+E_1 e^{-\beta(E_1+2E_2)}}{2(e^{-\beta E_1}+e^{-\beta(E_1+E_2)}+e^{-\beta E_2})^2} \geq 0. \end{aligned} \tag{A.67}$$

And for the latter

$$\begin{aligned} \partial_\beta \frac{p_{20}+p_{21}}{2(p_{01}+p_{12}+p_{20})} &= \partial_\beta \frac{e^{-\beta E_2}+e^{-\beta(E_1+E_2)}}{2(e^{-\beta E_1}+e^{-\beta(E_1+E_2)}+e^{-\beta E_2})} \\ &= \frac{(E_1-E_2)e^{-\beta(E_1+E_2)}-E_2 e^{-\beta(2E_1+E_2)}}{2(e^{-\beta E_1}+e^{-\beta(E_1+E_2)}+e^{-\beta E_2})^2} \leq 0. \end{aligned} \tag{A.68}$$

□

Proof of Eq. (54) *in Lemma* 4. We prove this lemma under the stated condition on the gap structure of the local Hamiltonians, i.e., $\delta_i \geq \delta_{i+1}$. Here we need to show that

$$\frac{\mathbf{q}}{\|\mathbf{q}\|} \succ \frac{\mathbb{1}+\Pi}{2}\frac{\mathbf{b}_1}{\|\mathbf{b}_1\|}, \tag{A.69}$$

$$\frac{\mathbf{q}}{\|\mathbf{q}\|} \succ \frac{\mathbb{1}+\Pi^2}{2}\frac{\mathbf{b}_2}{\|\mathbf{b}_2\|} = \frac{\mathbf{b}_2}{\|\mathbf{b}_2\|}. \tag{A.70}$$

To do so, we argue that the majorisation relations $\frac{\mathbf{q}}{\|\mathbf{q}\|} \succ \frac{\mathbb{1}+\Pi^i}{2}\frac{\mathbf{r}_i}{\|\mathbf{r}_i\|}$ and $\frac{\mathbb{1}+\Pi^i}{2}\frac{\mathbf{r}_i}{\|\mathbf{r}_i\|} \succ \frac{\mathbb{1}+\Pi^i}{2}\frac{\mathbf{b}_i}{\|\mathbf{b}_i\|}$, for $i=1,2$, hold.

Let us start with Eq. (A.69). The condition $\delta_i \geq \delta_{i+1}$ sets the ordering of $\frac{\mathbb{1}+\Pi}{2}\frac{\mathbf{r}_1}{\|\mathbf{r}_1\|}$. Still, some ambiguity remains as

$$p_{01}+p_{30} \geq p_{12}+p_{01} \geq p_{30}+p_{23} \geq p_{23}+p_{12}. \tag{A.71}$$

Thus, we need to prove the following three inequalities:

$$\frac{p_{00}}{p_{00}+p_{11}+p_{22}+p_{33}} \geq \frac{p_{01}+p_{30}}{2(p_{01}+p_{12}+p_{23}+p_{30})}, \tag{A.72a}$$

$$\frac{p_{00}+p_{11}}{p_{00}+p_{11}+p_{22}+p_{33}} \geq \frac{p_{01}+p_{30}+p_{12}+p_{01}}{2(p_{01}+p_{12}+p_{23}+p_{30})}, \tag{A.72b}$$

$$\frac{p_{33}}{p_{00}+p_{11}+p_{22}+p_{33}} \leq \frac{p_{32}+p_{12}}{2(p_{01}+p_{12}+p_{23}+p_{30})}. \tag{A.72c}$$

Inequality (A.72a) can be rewritten as

$$\frac{1}{1+e^{-2\beta E_1}+e^{-2\beta E_2}+e^{-2\beta E_3}} \geq \frac{1}{2\left(1+\frac{p_{12}+p_{23}}{p_{01}+p_{30}}\right)}, \tag{A.73}$$

which can further be turned into the inequality

$$1+1+e^{-\beta E_2}+e^{-\beta E_2} \geq 1+e^{-2\beta E_1}+e^{-2\beta E_2}+e^{-2\beta E_3}, \tag{A.74}$$

which holds because $E_1 \geq 0$ and $E_3 \geq E_2$.

Similarly, inequality (A.72b) can be recast as

$$\frac{1}{1+\frac{p_{22}+p_{33}}{p_{00}+p_{11}}} \geq \frac{1}{2}\left(\frac{1}{1+\frac{p_2}{p_0}}+\frac{1}{1+\frac{p_3}{p_1}}\right) \tag{A.75}$$

which implies the inequality

$$f\left(\frac{p_2^2+p_3^2}{p_0^2+p_1^2}\right) \geq \frac{1}{2}\left[f\left(\frac{p_2}{p_0}\right)+f\left(\frac{p_3}{p_1}\right)\right], \tag{A.76}$$

where we have introduced $f(x) := 1/(1+x)$. We can see that $f'(x) \leq 0$ and $\frac{p_2^2+p_3^2}{p_0^2+p_1^2} \leq \frac{p_2}{p_0}$, and in the case of $E_3 \leq E_1+E_2$ also $\frac{p_2}{p_0} \leq \frac{p_3}{p_1}$. Thus,

$$f\left(\frac{p_3}{p_1}\right) \leq f\left(\frac{p_2}{p_0}\right) \leq f\left(\frac{p_2^2+p_3^2}{p_0^2+p_1^2}\right), \tag{A.77}$$

from which

$$\frac{1}{2}\left[f\left(\frac{p_3}{p_1}\right)+f\left(\frac{p_2}{p_0}\right)\right] \leq f\left(\frac{p_2^2+p_3^2}{p_0^2+p_1^2}\right) \tag{A.78}$$

follows. This proves inequality (A.72b).

Inequality (A.72c) is equivalent to

$$\frac{1}{e^{2\beta E_3}+e^{2\beta(E_3-E_1)}+e^{2\beta(E_3-E_2)}+1} \leq \frac{1}{2\left(1+\frac{p_{01}+p_{30}}{p_{32}+p_{12}}\right)}, \tag{A.79}$$

which can be rewritten as

$$1+2\frac{p_0}{p_2} \leq e^{2\beta E_3}+e^{2\beta(E_3-E_1)}+e^{2\beta(E_3-E_2)}, \tag{A.80}$$

which is true since the right-hand side is larger or equal $e^{2\beta E_2}+2$, which, in turn, is larger than (or equal to) the left-hand side, because $(1-e^{\beta E_2})^2 \geq 0$. This proves Inequality (A.72c).

Thus far, we have shown that $\frac{\mathbf{q}}{\|\mathbf{q}\|} \succ \frac{\mathbb{1}+\Pi}{2}\frac{\mathbf{r}_1}{\|\mathbf{r}_1\|}$. To complete the first part of the proof, we also need to show that

$$\frac{\mathbb{1}+\Pi}{2}\frac{\mathbf{r}_1}{\|\mathbf{r}_1\|} \succ \frac{\mathbb{1}+\Pi}{2}\frac{\mathbf{b}_1}{\|\mathbf{b}_1\|}. \tag{A.81}$$

To do so, we first prove that the derivatives with respect to β of the two largest elements of $\frac{\mathbb{1}+\Pi}{2}\frac{\mathbf{r}_1}{\|\mathbf{r}_1\|}$ are positive and those

of the two smallest elements are negative. Let us represent the elements of $\frac{\mathbb{1}+\Pi}{2}\frac{\mathbf{r}_1}{\|\mathbf{r}_1\|}$ as

$$\frac{\mathbb{1}+\Pi}{2}\frac{\mathbf{r}_1}{\|\mathbf{r}_1\|} = \big(f_0(\beta), f_1(\beta), f_2(\beta), f_3(\beta)\big)^T. \tag{A.82}$$

Note that

$$\begin{aligned} f_0(\beta) &= \frac{p_{01}+p_{30}}{2(p_{01}+p_{12}+p_{23}+p_{30})} = \frac{1}{2}\frac{p_0(p_1+p_3)}{(p_0+p_2)(p_1+p_3)} \\ &= \frac{1}{2}\frac{1}{1+e^{-\beta E_2}}, \end{aligned} \tag{A.83}$$

which yields

$$f_0'(\beta) = \frac{1}{2}\frac{E_2\, e^{-\beta E_2}}{(1+e^{-\beta E_2})^2} \geq 0. \tag{A.84}$$

Similarly, we obtain

$$\begin{aligned} f_1(\beta) &= \frac{1}{2}\frac{1}{1+e^{-\beta(E_3-E_1)}},\ f_1'(\beta) = \frac{(E_3-E_1)e^{-\beta(E_3-E_1)}}{2(1+e^{-\beta(E_3-E_1)})^2} \geq 0, \\ f_2(\beta) &= \frac{1}{2}\frac{1}{1+e^{\beta E_2}},\ f_2'(\beta) = \frac{-E_2\, e^{\beta E_2}}{2(1+e^{\beta E_2})^2} \leq 0, \\ f_3(\beta) &= \frac{1}{2}\frac{1}{1+e^{\beta(E_3-E_1)}},\ f_3'(\beta) = \frac{-(E_3-E_1)e^{\beta(E_3-E_1)}}{2(1+e^{\beta(E_3-E_1)})^2} \leq 0. \end{aligned} \tag{A.85}$$

In the regime $E_3 \leq E_1 + E_2$, the components are ordered as

$$f_0(\beta) \geq f_4(\beta) \geq f_3(\beta) \geq f_2(\beta), \tag{A.86}$$

and thus one needs to show for $\beta' \leq \beta$ that

$$\begin{aligned} f_0(\beta) &\geq f_0(\beta'), \\ f_0(\beta)+f_1(\beta) &\geq f_0(\beta')+f_1(\beta'), \\ f_2(\beta) &\leq f_2(\beta'), \\ f_2(\beta)+f_3(\beta) &\leq f_2(\beta')+f_3(\beta'). \end{aligned} \tag{A.87}$$

These relations follow straightforwardly from Eqs. (A.84) and (A.85), noting that a function $f_i(x)$ that fulfils $f_i'(x) \leq 0$ ($f_i'(x) \geq 0$) the relation $f_i(x_1) \leq f_i(x_2)$ ($f_i(x_1) \geq f_i(x_2)$) is also satisfied for $x_1 \geq x_2$.

In a similar fashion, in the regime $E_3 \geq E_1+E_2$ the ordering of the components is

$$f_1(\beta) \geq f_0(\beta) \geq f_2(\beta) \geq f_3(\beta), \tag{A.88}$$

and thus one needs to show for $\beta' \leq \beta$ that

$$\begin{aligned} f_1(\beta) &\geq f_1(\beta'), \\ f_0(\beta)+f_1(\beta) &\geq f_0(\beta')+f_1(\beta'), \\ f_3(\beta) &\leq f_3(\beta'), \\ f_2(\beta)+f_3(\beta) &\leq f_2(\beta')+f_3(\beta'). \end{aligned} \tag{A.89}$$

These relations also follow straightforwardly from Eqs. (A.84) and (A.85).

We next turn our attention to Eq. (A.70). Because $p_{02} \geq p_{13}$, we need to prove the following inequalities:

$$\frac{p_{00}}{p_{00}+p_{11}+p_{22}+p_{33}} \geq \frac{p_{02}}{2(p_{02}+p_{13})}, \tag{A.90a}$$

$$\frac{p_{00}+p_{11}}{p_{00}+p_{11}+p_{22}+p_{33}} \geq \frac{2p_{02}}{2(p_{02}+p_{13})}, \tag{A.90b}$$

$$\frac{p_{33}}{p_{00}+p_{11}+p_{22}+p_{33}} \leq \frac{p_{13}}{2(p_{02}+p_{13})}. \tag{A.90c}$$

Inequality (A.90a) can be simplified to

$$\frac{p_0}{p_{00}+p_{11}+p_{22}+p_{33}} \geq \frac{p_2}{2(p_{02}+p_{13})}, \tag{A.91}$$

where we can use the inequalities $p_0p_{02} \geq p_2p_{11}$ and $p_0p_{13} \geq p_2p_{33}$ to arrive at the condition $p_0p_{13} \geq p_2p_{22}$. Due to our assumption on the energy eigenvalues, we have $p_{03} \geq p_{22}$, which proves inequality (A.90a).

Inequality (A.90b) can be rewritten as

$$\frac{p_{00}+p_{11}}{p_{00}+p_{11}+p_{22}+p_{33}} \geq \frac{2p_{02}}{2(p_{02}+p_{13})}, \tag{A.92}$$

which implies $p_{13}(p_{00}+p_{11}) \geq p_{02}(p_{22}+p_{33})$. Moreover, because $p_{00}+p_{11} \geq 2p_{01}$, we have

$$\begin{aligned} p_{13}(p_{00}+p_{11}) &\geq 2p_{01}p_{13} \geq p_{01}p_{13}+p_{02}p_{33} \\ &\geq p_{02}p_{22}+p_{02}p_{33}, \end{aligned} \tag{A.93}$$

where in the last step the condition $\delta_i \geq \delta_{i+1}$ was used. This proves inequality (A.90b).

Inequality (A.90c) can be recast as

$$\frac{p_3}{p_{00}+p_{11}+p_{22}+p_{33}} \leq \frac{p_1}{2(p_{02}+p_{13})}, \tag{A.94}$$

from which we arrive at

$$\begin{aligned} 2p_3p_{02}+p_3p_{13} &\leq p_1(p_{01}+p_{10}+p_{22}) \\ &\leq p_1(p_{00}+p_{11}+p_{22}), \end{aligned} \tag{A.95}$$

which is true for any energy spectrum. □

Unfortunately, Eq. (54) in Lemma 4 fails when $\delta_i \ngeq \delta_{i+1}$.

Counterexample to Eq. (54) for $\delta_i < \delta_{i+1}$. The relation $\frac{\mathbf{q}}{\|\mathbf{q}\|} > \frac{\mathbb{1}+\Pi}{2}\frac{\mathbf{r}_1}{\|\mathbf{r}_1\|}$ fails in the regime $\delta_i < \delta_{i+1}$. In that regime, the greatest element of $\frac{\mathbb{1}+\Pi}{2}\frac{\mathbf{r}_1}{\|\mathbf{r}_1\|}$ is $\frac{p_{12}+p_{01}}{2(p_{01}+p_{12}+p_{23}+p_{30})}$ and so

$$\frac{p_{00}}{p_{00}+p_{11}+p_{22}+p_{33}} \geq \frac{p_{12}+p_{01}}{2(p_{01}+p_{12}+p_{23}+p_{30})} \tag{A.96}$$

must hold in order for the relation to be true. But in the limit $E_3 \to \infty, E_1 \to 0, E_2 \to 0$ the above inequality becomes

$$\frac{p_{00}}{3p_{00}} \geq \frac{2p_{00}}{2(p_{00}+p_{00})} \Leftrightarrow \frac{1}{3} \geq \frac{1}{2}, \tag{A.97}$$

which is obviously invalid. The relation $\frac{\mathbf{q}}{\|\mathbf{q}\|} > \frac{\mathbf{r}_1}{\|\mathbf{r}_1\|}$ also fails sometimes. Looking at the limit $E_3 \to \infty$,

$\frac{\mathbf{q}}{\|\mathbf{q}\|} \to (p_{00}, p_{11}, p_{22}, 0)$ and $\frac{\mathbf{r}_1}{\|\mathbf{r}_1\|} \to (p_{02}, 0, p_{20}, 0)$. For the majorisation relation to hold, we would need $p_{22} \leq 0$, which is clearly not true in general.

Proof of Eq. (55) in Lemma 4. We need to prove that $\|\mathbf{r}_i\| \leq \|\mathbf{b}_i\|$, for $i = 1, 2$, for $d = 4$. To do so, we again use the first derivative of $\|\mathbf{r}_i\|$ and show that it is always negative. From

$$\|\mathbf{r}_i\| = \sum_j p_j p_{j+i} = \frac{\sum_j e^{-\beta(E_j+E_{j+i})}}{\sum_{m,n} e^{-\beta(E_m+E_n)}}, \tag{A.98}$$

the partial derivative with respect to β reads

$$\begin{aligned}\partial_\beta \|\mathbf{r}_i\| &= \sum_{j,m,n} (E_m + E_n - E_j - E_{j+i}) \frac{e^{-\beta(E_m+E_n+E_j+E_{j+i})}}{z(\beta)^4} \\ &= \frac{1}{z(\beta)^3} \sum_{j,m} (2E_m - E_j - E_{j+i}) e^{-\beta(E_m+E_j+E_{j+i})}.\end{aligned} \tag{A.99}$$

Now we need to show that for any $i \neq 0$, $\partial_\beta \|\mathbf{r}_i\|$ is negative. For $i = 1$ we find

$$\begin{aligned}\partial_\beta \|\mathbf{r}_1\| = \frac{-1}{z(\beta)^3} \big[& E_1 (e^{-\beta E_1} + e^{-\beta(E_1+E_2)} - e^{-2\beta E_1}) + (E_2 - E_1)(-e^{-\beta(E_1+E_2)} + e^{-\beta(2E_1+E_2)} \\ & + e^{-\beta(E_1+E_2+E_3)} - e^{-\beta(E_1+2E_2)}) + (E_3 - E_2)(e^{-\beta(E_2+E_3)} - e^{-\beta(E_1+E_2+E_3)} + e^{-\beta(2E_2+E_3)} - e^{-\beta(E_2+2E_3)}) \\ & + E_3 (e^{-\beta(E_2+E_3)} + e^{-\beta E_3} - e^{-\beta(E_1+E_3)} - e^{-2\beta E_3}) \big] \leq 0.\end{aligned} \tag{A.100}$$

Since we have ordered the energy eigenvalues in the increasing order, it is straightforward to show that $\partial_\beta \|\mathbf{r}_1\|$ is always nonpositive for any set of energy eigenvalues in dimension 4.

Now similarly, we can show that $\|\mathbf{r}_2\| \leq \|\mathbf{b}_2\|$. By employing Eq. (A.99), $\partial_\beta \|\mathbf{r}_2\|$ is obtained as

$$\begin{aligned}\partial_\beta \|\mathbf{r}_2\| = \frac{-2}{z(\beta)^3} \big[& E_1 (e^{-\beta E_2} + 2e^{-\beta(E_1+E_3)} - e^{-\beta(E_1+E_2)} - e^{-2\beta E_2} - e^{-\beta(E_2+E_3)}) + (E_2 - E_1)(e^{-\beta E_2} + e^{-\beta(E_1+E_3)} \\ & + e^{-\beta(E_1+E_2)} + e^{-\beta(2E_1+E_3)} - e^{-2\beta E_2} - e^{-\beta(E_1+E_2+E_3)} - e^{-\beta(E_2+E_3)} - e^{-\beta(E_1+2E_3)}) \\ & + (E_3 - E_2)(e^{-\beta(E_1+E_3)} + e^{-\beta(2E_1+E_3)} + e^{-\beta(E_1+E_2+E_3)} - 2e^{-\beta(E_2+E_3)} - e^{-\beta(E_1+2E_3)}) \big].\end{aligned} \tag{A.101}$$

The above expression is also nonpositive under the assumption $\delta_i \geq \delta_{i+1}$. □

A.X. Proof of monotonicity and convexity of the thermal curve

In this section, we will show that the condition (II) mentioned in Sec. III.3 is true for every choice of the set $\{E_i\}_{i=0}^{d-1}$ and any initial inverse temperature.

Expressing a general point in the curve $\mathbf{p}(\beta') = (x_0(\beta'), \ldots, x_{d-2}(\beta'), -1)$ as a linear combination of the vertices $\mathbf{v}_i$ in condition (I) we have

$$\begin{pmatrix} x_0(\beta') \\ x_1(\beta') \\ \vdots \\ x_{d-2}(\beta') \\ -1 \end{pmatrix} = \begin{pmatrix} x_0(\beta) & 0 & 0 & \ldots & 0 \\ x_1(\beta) & x_1(\beta) & 0 & \ldots & 0 \\ \vdots & \ddots & x_2(\beta) & \ddots & 0 \\ x_{d-2}(\beta) & \ldots & \ddots & \ddots & 0 \\ -1 & -1 & -1 & \ldots & -1 \end{pmatrix} \begin{pmatrix} a_0 \\ a_1 \\ \vdots \\ a_{d-2} \\ a_{d-1} \end{pmatrix}, \tag{A.102}$$

where the $(a_0, \ldots, a_{d-1})$ are coefficients given by

$$a_i = \frac{x_i(\beta')}{x_i(\beta)} - \frac{x_{i-1}(\beta')}{x_{i-1}(\beta)} = \frac{x_i(\beta')}{x_{i-1}(\beta)} \left(\frac{x_{i-1}(\beta)}{x_i(\beta)} - \frac{x_{i-1}(\beta')}{x_i(\beta')} \right), \tag{A.103}$$

which are positive for all i and satisfy $\sum_i a_i = 1$ (and thus below 1 for all i) if the following condition holds

$$\frac{\mathrm{d}}{\mathrm{d}\beta} \left(\frac{x_m}{x_{m+1}}(\beta) \right) = \frac{1}{x_{m+1}} \left(\frac{\partial x_m}{\partial \beta} - \frac{x_m}{x_{m+1}} \frac{\partial x_{m+1}}{\partial \beta} \right) \geq 0 \ \forall m, \tag{A.104}$$

which means that the function $x_i/x_{i+1}(\beta)$ is monotonically decreasing with β. This in turn means that the final point can be reached as a convex combination of the vertices.

With a convenient relabeling of the index we can write $x_{m+1}/x_m(\beta)$ in the form

$$\begin{aligned}\frac{x_m}{x_{m-1}} &= \frac{(m+1)e^{-\beta E_{m+1}} - \sum_{i=0}^{m} e^{-\beta E_i}}{m e^{-\beta E_m} - \sum_{i=0}^{m-1} e^{-\beta E_i}} \\ &= \frac{(m+1)e^{-\beta E_{m+1}} - m e^{-\beta E_m} + m e^{-\beta E_m} - \sum_{i=0}^{m-1} e^{-\beta E_i} - e^{-\beta E_m}}{m e^{-\beta E_m} - \sum_{i=0}^{m-1} e^{-\beta E_i}} \\ &= 1 + \frac{(m+1)(e^{-\beta E_{m+1}} - e^{-\beta E_m})}{m e^{-\beta E_m} - \sum_{i=0}^{m-1} e^{-\beta E_i}}.\end{aligned} \tag{A.105}$$

To prove our claim [condition (II)], we need to show that for any $\beta \geq 0$ the function x_m/x_{m-1} is monotonically decreasing with β. That is, we have to show that

$$R(\beta) = \frac{\mathrm{d}}{\mathrm{d}\beta} \left(\frac{x_m}{x_{m-1}}(\beta) \right) \leq 0. \tag{A.106}$$

The derivative takes the form

$$
\begin{aligned}
R(\beta) &= \tfrac{m+1}{x_{m-1}^2}\Big[-(E_{m+1}e^{-\beta E_{m+1}} - E_m e^{-\beta E_m})(m e^{-\beta E_m} - \sum_{i=0}^{m-1} e^{-\beta E_i}) + (e^{-\beta E_{m+1}} - e^{-\beta E_m})(m\, E_m e^{-\beta E_m} - \sum_{i=0}^{m-1} E_i e^{-\beta E_i})\Big] \\
&= \tfrac{m+1}{x_{m-1}^2}\Big(\sum_{i=0}^{m-1}[(E_{m+1}-E_i)e^{-\beta(E_{m+1}+E_i)} - (E_m - E_i)e^{-\beta(E_m+E_i)}] - m(E_{m+1}-E_m)e^{-\beta(E_{m+1}+E_m)}\Big) \\
&= \tfrac{m+1}{x_{m-1}^2}\Big(\sum_{i=0}^{m-1}[(E_{m+1}-E_i)e^{-\beta(E_{m+1}+E_i)} - (E_m - E_i)e^{-\beta(E_m+E_i)}] - m(E_{m+1}-E_i+E_i-E_m)e^{-\beta(E_{m+1}+E_m)}\Big) \\
&= \tfrac{m+1}{x_{m-1}^2}\sum_{i=0}^{m-1}\big[(E_{m+1}-E_i)(e^{-\beta(E_{m+1}+E_i)} - e^{-\beta(E_{m+1}+E_m)}) - (E_m-E_i)(e^{-\beta(E_m+E_i)} - e^{-\beta(E_{m+1}+E_m)})\big] \\
&= \tfrac{m+1}{x_{m-1}^2}e^{-\beta(E_{m+1}+E_m)}\sum_{i=0}^{m-1}\big[(E_{m+1}-E_i)(e^{\beta(E_m-E_i)}-1) - (E_m-E_i)(e^{\beta(E_{m+1}-E_i)}-1)\big]. \qquad \text{(A.107)}
\end{aligned}
$$

To prove the nonpositivity of $R(\beta)$ we need to show that

$$
(E_{m+1}-E_i)(e^{\beta(E_m-E_i)}-1) - (E_m-E_i)(e^{\beta(E_{m+1}-E_i)}-1) \le 0 \quad \text{for} \quad E_i \le E_m \le E_{m+1}. \qquad \text{(A.108)}
$$

To do so, we define $y_m := \beta(E_m - E_i)$, and write inequality (A.108) in the form

$$
h(y_{m+1}) = \frac{y_{m+1}}{e^{y_{m+1}}-1} \le \frac{y_m}{e^{y_m}-1}. \qquad \text{(A.109)}
$$

This inequality is satisfied for all $y_{m+1} \ge y_m$ if the function $h(y)$ is monotonically decreasing. To see that this is the case, we calculate the derivative

$$
\frac{\partial h(y)}{\partial y} = \frac{(1-y)e^y - 1}{(e^y-1)^2}, \qquad \text{(A.110)}
$$

which can be seen to be smaller or equal to zero since $(1-y)e^y$ is a monotonically decreasing function for $y \ge 0$, i.e., $\frac{\partial}{\partial y}(1-y)e^y = -ye^y \le 0$ and hence has its maximum value of 1 at $y = 0$, confirming that $(1-y)e^y - 1 \le 0$ and that $h(y)$ is monotonically decreasing.

A.X.a. Explicit partial derivatives in dimension 3

To show explicitly that the curve $x(\beta)$ is monotonically increasing in $d = 3$, we first calculate

$$
\frac{\partial x}{\partial \beta} = -\mathcal{Z}^{-2} f_x(\beta), \qquad \text{(A.111a)}
$$

$$
\frac{\partial y}{\partial \beta} = -3\mathcal{Z}^{-2} f_y(\beta), \qquad \text{(A.111b)}
$$

where the functions $f_x(\beta)$ and $f_y(\beta)$ are given by

$$
f_x(\beta) = E_1 e^{-\beta E_1}(2 + e^{-\beta E_2}) + E_2 e^{-\beta E_2}(1 - e^{-\beta E_1}) \ge 0, \qquad \text{(A.112a)}
$$

$$
f_y(\beta) = (E_2 - E_1)e^{-\beta(E_1+E_2)} + E_2 e^{-\beta E_2} \ge 0. \qquad \text{(A.112b)}
$$

With this, we can then evaluate

$$
\frac{\partial y}{\partial x} = \left(\frac{\partial x}{\partial \beta}\right)^{-1}\frac{\partial y}{\partial \beta} = \frac{3 f_y(\beta)}{f_x(\beta)} \ge 0, \qquad \text{(A.113)}
$$

which allows us to confirm that the curve segment lies above (with respect to the coordinates x and y in the plane of the polytope) the line connecting $\big(x(\beta), y(\beta)\big)$ and $(\tilde{x}, \tilde{y})$.

To show that this curve is a convex function, we need to show that $\partial^2 y/\partial x^2 \ge 0$, and calculate

$$
\begin{aligned}
\frac{\partial^2 y}{\partial x^2} &= \left(\frac{\partial x}{\partial \beta}\right)^{-1}\frac{\partial}{\partial \beta}\left[\left(\frac{\partial x}{\partial \beta}\right)^{-1}\frac{\partial y}{\partial \beta}\right] \\
&= \left(\frac{\partial x}{\partial \beta}\right)^{-2}\left\{\frac{\partial^2 y}{\partial \beta^2} - \left(\frac{\partial y}{\partial \beta}\right)\left(\frac{\partial x}{\partial \beta}\right)^{-1}\left(\frac{\partial^2 x}{\partial \beta^2}\right)\right\}. \qquad \text{(A.114)}
\end{aligned}
$$

The second partial derivatives with respect to β are found to be

$$
\frac{\partial x^2}{\partial \beta^2} = \mathcal{Z}^{-3}\big[2\tfrac{\partial \mathcal{Z}}{\partial \beta} f_x - \mathcal{Z}\tfrac{\partial f_x}{\partial \beta}\big], \qquad \text{(A.115a)}
$$

$$
\frac{\partial y^2}{\partial \beta^2} = 3\mathcal{Z}^{-3}\big[2\tfrac{\partial \mathcal{Z}}{\partial \beta} f_y - \mathcal{Z}\tfrac{\partial f_y}{\partial \beta}\big]. \qquad \text{(A.115b)}
$$

Combining this with Eq. (A.114) we find

$$
\frac{\partial^2 y}{\partial x^2} = 3\,\mathcal{Z}^2 f_x^{-3}\big(f_y \frac{\partial f_x}{\partial \beta} - f_x \tfrac{\partial f_y}{\partial \beta}\big). \qquad \text{(A.116)}
$$

The derivatives of $f_x(\beta)$ and $f_y(\beta)$ are

$$
\frac{\partial f_x}{\partial \beta} = \big(E_2^2 - E_1^2\big)e^{-\beta(E_1+E_2)} - 2E_1^2 e^{-\beta E_1} - E_2^2 e^{-\beta E_2}, \qquad \text{(A.117a)}
$$

$$
\frac{\partial f_y}{\partial \beta} = -\big(E_2^2 - E_1^2\big)e^{-\beta(E_1+E_2)} - E_2^2 e^{-\beta E_2}, \qquad \text{(A.117b)}
$$

which we insert into Eq. (A.116) to arrive at

$$\frac{\partial^2 y}{\partial x^2} = 6\,\mathcal{Z}^3 f_x^{-3} E_1 E_2 \big(E_2 - E_1\big) e^{-\beta(E_1+E_2)} \geq 0, \quad \text{(A.118)}$$

where we have made use of the fact that $f_x \geq 0$ (since $1 \geq e^{-\beta E_1}$), along with $E_1, E_2, \mathcal{Z} \geq 0$ and $E_2 \geq E_1$.

A.X.b. *Explicit partial derivatives in dimension 4*

In this appendix, we present the explicit expressions for the first and second partial derivatives. The derivatives of the coordinates $x(\beta)$, $y(\beta)$ and $z(\beta)$ along the curve of thermal states can be written as

$$\frac{\partial x}{\partial \beta} = -\mathcal{Z}^{-2} f_x(\beta), \quad \text{(A.119a)}$$

$$\frac{\partial y}{\partial \beta} = -\mathcal{Z}^{-2} f_y(\beta), \quad \text{(A.119b)}$$

$$\frac{\partial z}{\partial \beta} = -\mathcal{Z}^{-2} f_z(\beta), \quad \text{(A.119c)}$$

where the functions $f_x(\beta)$, $f_y(\beta)$, and $f_z(\beta)$ are given by

$$\begin{aligned} f_x(\beta) &= E_1 e^{-\beta E_1}(2 + e^{-\beta E_2} + e^{-\beta E_3}) \\ &\quad + (E_2 e^{-\beta E_2} + E_3 e^{-\beta E_3})(1 - e^{-\beta E_1}) \geq 0, \quad \text{(A.120a)} \\ f_y(\beta) &= 3(E_2 - E_1) e^{-\beta(E_1+E_2)} + 3E_2 e^{-\beta E_2} \\ &\quad + E_3 e^{-\beta E_3}(1 + e^{-\beta E_1} - 2e^{-\beta E_2}) \\ &\quad + e^{-\beta E_3}(2E_2 e^{-\beta E_2} - E_1 e^{-\beta E_1}) \geq 0, \quad \text{(A.120b)} \\ f_z(\beta) &= 4e^{-\beta E_3}\big[E_3 + e^{-\beta E_1}(E_3 - E_1) \\ &\quad + e^{-\beta E_2}(E_3 - E_2)\big] \geq 0, \quad \text{(A.120c)} \end{aligned}$$

where $f_y \geq 0$ follows, since $E_3 \geq E_2$ and $(1 + e^{-\beta E_1} - 2e^{-\beta E_2}) \geq 0$ which implies that the terms in f_y proportional to $e^{-\beta E_3}$ can be bounded by $E_3 e^{-\beta E_3}\big(E_2 + e^{-\beta E_1}(E_2 - E_1)\big) \geq 0$. Since f_x, f_y, and f_z are nonnegative, all partial derivatives with respect to β are nonpositive, but

$$\frac{\partial y}{\partial x} = \left(\frac{\partial x}{\partial \beta}\right)^{-1} \frac{\partial y}{\partial \beta} = \frac{f_y(\beta)}{f_x(\beta)} \geq 0, \quad \text{(A.121)}$$

$$\frac{\partial z}{\partial x} = \left(\frac{\partial x}{\partial \beta}\right)^{-1} \frac{\partial z}{\partial \beta} = \frac{f_z(\beta)}{f_x(\beta)} \geq 0, \quad \text{(A.122)}$$

$$\frac{\partial z}{\partial y} = \left(\frac{\partial y}{\partial \beta}\right)^{-1} \frac{\partial z}{\partial \beta} = \frac{f_z(\beta)}{f_y(\beta)} \geq 0. \quad \text{(A.123)}$$

For the second partial derivatives, we have

$$\begin{aligned} \frac{\partial^2 y}{\partial x^2} &= \left(\frac{\partial x}{\partial \beta}\right)^{-1} \frac{\partial}{\partial \beta}\left[\left(\frac{\partial x}{\partial \beta}\right)^{-1} \frac{\partial y}{\partial \beta}\right] \\ &= \left(\frac{\partial x}{\partial \beta}\right)^{-2} \left\{\frac{\partial^2 y}{\partial \beta^2} - \left(\frac{\partial y}{\partial \beta}\right)\left(\frac{\partial x}{\partial \beta}\right)^{-1}\left(\frac{\partial^2 x}{\partial \beta^2}\right)\right\} \\ &= \mathcal{Z}^2 f_x^{-3}\big(f_y \frac{\partial f_x}{\partial \beta} - f_x \frac{\partial f_y}{\partial \beta}\big), \quad \text{(A.124)} \end{aligned}$$

$$\frac{\partial^2 z}{\partial x^2} = \mathcal{Z}^2 f_x^{-3}\big(f_z \frac{\partial f_x}{\partial \beta} - f_x \frac{\partial f_z}{\partial \beta}\big), \quad \text{(A.125)}$$

$$\frac{\partial^2 z}{\partial y^2} = \mathcal{Z}^2 f_y^{-3}\big(f_z \frac{\partial f_y}{\partial \beta} - f_y \frac{\partial f_z}{\partial \beta}\big). \quad \text{(A.126)}$$

We thus have to calculate the derivatives of the functions f_x, f_y, and f_z with respect to β, which evaluate to

$$\begin{aligned} \frac{\partial f_x}{\partial \beta} &= -E_1^2 e^{-\beta E_1}\big(2 + e^{-\beta E_2} + e^{-\beta E_3}\big) \\ &\quad - \big(E_2^2 e^{-\beta E_2} + E_3^2 e^{-\beta E_3}\big)\big(1 - e^{-\beta E_1}\big), \quad \text{(A.127)} \\ \frac{\partial f_y}{\partial \beta} &= E_1^2 e^{-\beta E_1}\big(3e^{-\beta E_2} + e^{-\beta E_3}\big) \\ &\quad - E_2^2 e^{-\beta E_2}\big(3 + 3e^{-\beta E_1} + 2e^{-\beta E_3}\big) \\ &\quad - E_3^2 e^{-\beta E_3}\big(1 + e^{-\beta E_1} - 2e^{-\beta E_2}\big), \quad \text{(A.128)} \\ \frac{\partial f_z}{\partial \beta} &= 4e^{-\beta E_3}\big[E_1^2 e^{-\beta E_1} + E_2^2 e^{-\beta E_2} \\ &\quad - E_3^2\big(1 + e^{-\beta E_1} + e^{-\beta E_2}\big)\big]. \quad \text{(A.129)} \end{aligned}$$

Then, since $\mathcal{Z}^2 \geq 0$ and $f_x, f_y, f_z \geq 0$, we just have to evaluate

$$\begin{aligned} f_y \frac{\partial f_x}{\partial \beta} - f_x \frac{\partial f_y}{\partial \beta} &= 2\big[E_1 E_2 e^{-\beta(E_1+E_2)}(E_2 - E_1) + E_1 E_3 e^{-\beta(E_1+E_3)}(E_3 - E_1)\big]\big(3 + e^{-\beta E_3}\big)\mathcal{Z} \\ &\quad + 2E_2 E_3 e^{-\beta(E_2+E_3)}(E_3 - E_2)\big(1 + e^{-\beta E_1}\big)\big(2 + 2e^{-\beta E_1} - e^{-\beta E_2} + e^{-\beta E_3}\big) \geq 0, \quad \text{(A.130a)} \end{aligned}$$

$$\begin{aligned} f_z \frac{\partial f_x}{\partial \beta} - f_x \frac{\partial f_z}{\partial \beta} &= 4\mathcal{Z} e^{-\beta E_3}\big[E_1 e^{-\beta E_1}(E_2 - E_1)\big(e^{-\beta E_2}(E_3 - E - 2) + 2E_3\big) \\ &\quad + (E_3 - E_2)E_3\big(E_1 e^{-\beta(E_1+E_2)} + 2E_1 e^{-\beta E_1} + E_2 e^{-\beta E_2}(1 - e^{-\beta E_1})\big)\big] \geq 0, \quad \text{(A.130b)} \end{aligned}$$

$$f_z \frac{\partial f_y}{\partial \beta} - f_y \frac{\partial f_z}{\partial \beta} = 12\mathcal{Z} e^{-\beta(E_2+E_3)}\big(E_3 - E_2\big)\big[E_1^2 e^{-\beta E_1} + E_3\big(E_2 - E_1\big)e^{-\beta E_1} + E_2\big(E_3 - e^{-\beta E_1} E_1\big)\big] \geq 0. \quad \text{(A.130c)}$$

A.XI. Geometry of equal thermal marginals for two qutrits

For local dimension $d = 3$, the transformations we consider here to ensure symmetric marginals imply that the vectors collecting the diagonal entries of the final state marginals are of the form

$$\tilde{\mathbf{p}} = \tilde{\mathbf{p}}_A = \tilde{\mathbf{p}}_B = M_q\mathbf{q} + (\mathbb{1} + \Pi)M_r\mathbf{r}\,, \tag{A.131}$$

where $\mathbf{q} = (q_i)$ with $q_i = p_i^2$ and $(\mathbb{1} + \Pi)\mathbf{r} = \big(p_i(1 - p_i)\big)$. This means that the set of reachable states corresponds to the convex hull of the points $\tilde{\mathbf{p}}^{(i,j)} := \Pi^{(i)}\mathbf{q} + (\mathbb{1} + \Pi)\Pi^{(j)}\mathbf{r}$ for $i, j \in \{1, \ldots, 3!\}$. However, in our case the problem can be further reduced to that of showing that the points $\mathbf{p}(\beta' < \beta)$ lie within a restricted part of the polytope, i.e., a triangle between the points $\mathbf{p}(\beta)$, $\mathbf{p}(\beta \to 0)$, and $\tilde{\mathbf{p}}^{(6,1)} = \Pi^{(6)}\mathbf{q} + (\mathbb{1} + \Pi)\Pi^{(1)}\mathbf{r}$, where

$$\Pi^{(1)} = \mathbb{1},\ \ \Pi^{(6)} = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix},\ \ \Pi = \begin{pmatrix} 0 & 0 & 1 \\ 1 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}.. \tag{A.132}$$

Note that the point $\mathbf{p}(\beta \to 0)$ trivially lies in the polytope, since it can be obtained from an equally weighted convex combination of all cyclic permutations obtained for $\Pi^{(i)} = \Pi^{(j)} = \mathbb{1}, \Pi, \Pi^2$. Then we switch from the coordinates $\{p_0, p_1, p_2\}$ to the coordinates $\{x, y, z\}$, given by

$$x = -p_0 + p_1, \tag{A.133a}$$
$$y = -p_0 - p_1 + 2p_2, \tag{A.133b}$$
$$z = -p_0 - p_1 - p_2 = -1. \tag{A.133c}$$

The plane containing the polytope is defined by $z = -1$ such that $p_2 = 1 - p_0 - p_1$, and we therefore have $y = 2 - 3p_0 - 3p_1$. The corners $(p_0, p_1, p_2) = (1, 0, 0)$, $(0, 1, 0)$, and $(0, 0, 1)$ of the outer triangle containing all (diagonal) qutrit states are mapped to $(x, y) = (-1, -1)$, $(1, -1)$, and $(0, 2)$, respectively, see Fig. 1. Moreover, the curve of thermal states is parameterised as $\big(x(\beta), y(\beta)\big)$ with $x(\beta) = -p_0(\beta) + p_1(\beta)$ and $y(\beta) = 2 - 3p_0(\beta) - 3p_1(\beta)$, and connects the points $(x, y) = (-1, -1)$ corresponding to $\mathbf{p}(\beta \to \infty)$ and $(x, y) = (0, 0)$ corresponding to $\mathbf{p}(\beta \to 0)$.

Next, we note that for any given thermal state with coordinates $\big(x(\beta), y(\beta)\big)$, the coordinates $\big(x = \tilde{x}, y = \tilde{y}\big)$ of the corresponding point $\tilde{\mathbf{p}}^{(6,1)}$ are given by

$$\tilde{x} = (p_0 - p_1)\big[2(p_0 + p_1) - 1\big] \geq 0, \tag{A.134a}$$
$$\tilde{y} = 2 - 3p_0 - 3p_1 = y(\beta), \tag{A.134b}$$

and consequently this point always lies on the same height $\tilde{y} = y(\beta)$ as the point corresponding to $\mathbf{p}(\beta)$, but to the right, i.e., $\tilde{x} \geq 0 \geq x(\beta)$ of any of the points corresponding to thermal states (in particular $\mathbf{p}(\beta \to 0)$). This shows that the point with coordinates $(x_0, x_1) = (0, y(\beta))$ is included in the polytope, since it can be obtained as a convex combination of $\mathbf{p}(\beta)$ and $\tilde{\mathbf{p}}^{(6,1)}$.

Moreover, evaluating the partial derivatives, we have found $(\partial y(\beta)/\partial x(\beta) \geq 0)$ and $(\partial^2 y/\partial x^2 \geq 0)$, see Appendix A.X.b, i.e., the curve described by $\big(x(\beta), y(\beta)\big)$ is a monotonically increasing $(\partial y(\beta)/\partial x(\beta) \geq 0)$ and convex $(\partial^2 y/\partial x^2 \geq 0)$ function, implying that the curve segment corresponding to all points $\big(x(\beta'), y(\beta')\big)$ with $\beta' \leq \beta$ is contained within the triangle $\mathbf{p}(\beta)$, $\tilde{\mathbf{p}}^{(6,1)}$, and $\mathbf{p}(\beta \to 0)$.

A.XII. Geometry of equal thermal marginals for two ququarts

For local dimension $d = 4$, we have marginal vectors of the form

$$\tilde{\mathbf{p}} = \tilde{\mathbf{p}}_A = \tilde{\mathbf{p}}_B = M_q\mathbf{q} + (\mathbb{1} + \Pi)M_{r_1}\mathbf{r}_1 + \tfrac{1}{2}(\mathbb{1} + \Pi^2)M_{r_2}\mathbf{r}_2. \tag{A.135}$$

The set of reachable states corresponds to the convex hull of the points $\tilde{\mathbf{p}}^{(i,j,k)} := \Pi^{(i)}\mathbf{q} + (\mathbb{1} + \Pi)\Pi^{(j)}\mathbf{r}_1 + \frac{1}{2}(\mathbb{1} + \Pi^2)\Pi^{(k)}\mathbf{r}_2$ for $i, j, k \in \{1, \ldots, 4!\}$. Switching from the coordinates $\{p_0, p_1, p_2, p_3\}$ to the new coordinates $\{x, y, z, w\}$, given by

$$x = -p_0 + p_1, \tag{A.136a}$$
$$y = -p_0 - p_1 + 2p_2, \tag{A.136b}$$
$$z = -p_0 - p_1 - p_2 + 3p_3, \tag{A.136c}$$
$$w = -p_0 - p_1 - p_2 - p_3 = -1\,. \tag{A.136d}$$

We here show that the points $(x, y, z) = (0, y(\beta), z(\beta))$ and $(0, 0, z(\beta))$ are included. To achieve this, we identify a set of 5 points that all lie in the plane of constant $z = z(\beta)$, and show that the two-dimensional polytope spanned by these points encloses both $(0, y(\beta), z(\beta))$ and $(0, 0, z(\beta))$, see Fig. 2. These five points are $A = \tilde{\mathbf{p}}^{(1,1,1)}$, $B = \tilde{\mathbf{p}}^{(7,1,1)}$, $C = \tilde{\mathbf{p}}^{(13,7,1)}$, $D = \tilde{\mathbf{p}}^{(9,7,1)}$, and $E = \tilde{\mathbf{p}}^{(9,1,1)}$, where

$$\Pi = \begin{pmatrix} 0 & 0 & 0 & 1 \\ 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \end{pmatrix},\ \Pi^{(7)} = \begin{pmatrix} 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix},$$
$$\Pi^{(9)} = \begin{pmatrix} 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix},\ \Pi^{(13)} = \begin{pmatrix} 0 & 0 & 1 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \tag{A.137}$$

The point B can be seen to have the same y-coordinate as the starting point A, $y(B) = y(A) = y(\beta)$, while

$$x(B) = \big(p_0(\beta) - p_1(\beta)\big)\big(2p_0(\beta) + 2p_1(\beta) - 1\big) \geq 0, \tag{A.138}$$

since $p_0(\beta) \geq p_1(\beta)$ and $p_0(\beta) + p_1(\beta) \geq \frac{1}{2}$ for any Hamiltonian and any temperature. This means the point $(0, y(\beta), z(\beta))$ is contained in polytope since it can be obtained as a convex combination of A and B.

Finally, to show that the point $\tilde{O} = (0, 0, z(\beta))$ is contained within the polytope, we show that the points C, D and E satisfy, $x(C) \geq 0$, $y(D) \geq 0$ and $x(E) \leq 0$, while $(A\tilde{O} \times AE)_z \geq 0$, $(E\tilde{O} \times ED)_z \geq 0$ and $(D\tilde{O} \times DC)_z \geq 0$, where for any two points X and Y we use the notation $XY = Y - X$. In other words, the closed path connecting the five points A, B, C, D and E encircles the point $\tilde{O}$, meaning that $\tilde{O}$ can be written as a convex combination of these points.

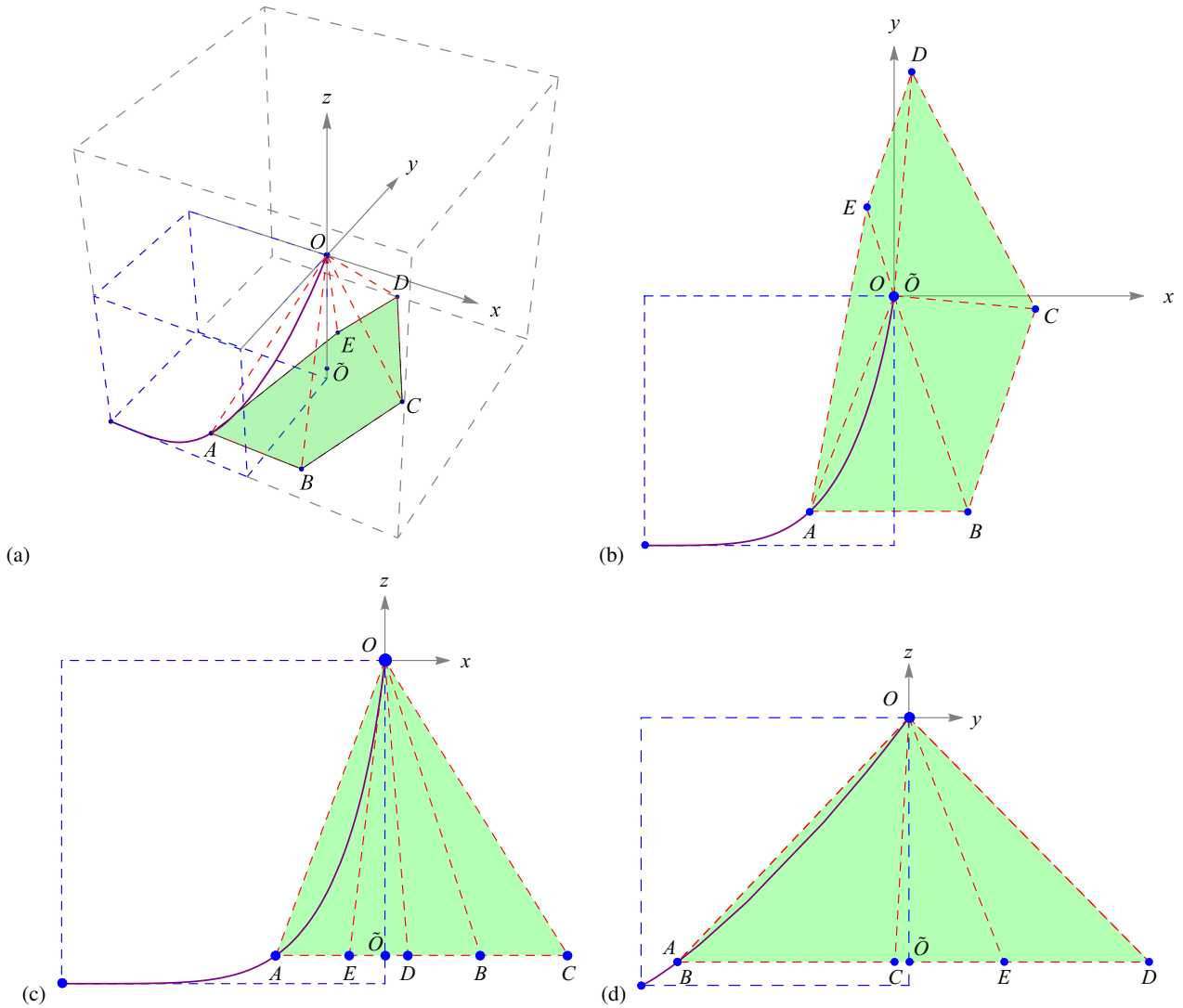


FIG. 2. **Illustration of geometric method for** $d = 4$. (a) In local dimension $d = 4$, the space of marginal probability distributions can be mapped to the hyperplane of constant coordinate $w = -1$, leaving a three-dimensional space with coordinates x, y, and z. For any fixed Hamiltonian, the thermal states form a one-parameter family of states along a line connecting $(x, y, z) = (-1, -1, -1)^T$ with $(0, 0, 0)^T$, where the curve is strictly confined to the region $x, y, z \leq 0$ (dashed blue). The dashed red lines delineate the minimal polytope spanned by the points A, B, C, D, E (which all share the same z-coordinate $z = z(\beta)$) and the origin O. (b), (c), (d) show the projections onto the x-y, x-z and y-z plane, respectively.

In a slight abuse of notation, we now use the shorthand $p_i \equiv p_i(\beta)$ for the thermal state diagonal components to express the relevant coordinates for the points C, D and E as

$$x(C) = \big(p_0 - p_1\big)\big(2p_0 + 2p_1 + p_2 - 1\big) + \big(p_1 - p_2\big)\big(p_0 + p_1 + p_2\big) \geq 0, \tag{A.139a}$$

$$y(D) = 3\big(p_0 - p_1\big)\big(p_0 + p_1 + p_2 - \tfrac{1}{3}\big) + 3\big(p_1 - p_2\big)\big(p_0 + p_1 + p_2 - \tfrac{2}{3}\big) \geq 0, \tag{A.139b}$$

$$x(E) = -\big[\big(p_0 - p_1\big)\big(1 - p_0 - p_1\big) + \big(p_1 - p_2\big)\big(p_1 + p_2\big)\big] \leq 0, \tag{A.139c}$$

while the z-components of the relevant cross products are given explicitly below. In Appendix A.X.b we have further shown that the partial derivatives $\partial y/\partial x$, $\partial z/\partial x$, and $\partial z/\partial y$ as well as second derivatives $\partial^2 y/\partial x^2$, $\partial^2 z/\partial x^2$, and $\partial^2 z/\partial y^2$ are nonnegative along the curve of thermal states, meaning that conditions (I) and (II) are satisfied for $d = 4$.

A.XII.a. Relevant cross products

Finally, the z-components of the relevant cross products are given

$$(A\tilde{O}\times AE)_z = \big(p_0-p_1\big)^2\big(2p_0-2p_1+3p_2\big) + \big(p_0-p_1\big)\big(p_1-p_2\big)\big(p_0-p_1+4p_2\big) + 2\big(p_1-p_2\big)^2\big(p_1+p_2\big) \geq 0, \quad \text{(A.140a)}$$

$$(E\tilde{O}\times ED)_z = 2p_1\big(p_0-p_2\big)\Big[\big(p_0-p_1\big)\big(1-p_1+p_2\big) + \big(p_1-p_2\big)\big(p_1+2p_2-p_3\big)\Big] \geq 0, \quad \text{(A.140b)}$$

$$\begin{aligned}(D\tilde{O}\times DC)_z = {} & \big(p_0-p_1\big)^2\big(p_0+p_2\big)\big((3p_0+3p_1-1)+3(p_1-p_3)\big) \\ & + \big(p_0-p_1\big)\big(p_1-p_2\big)\big(3(p_0-p_1)(p_0+p_1)+6p_0(p_2-p_3)+2p_2(2p_0+5p_1+2p_2-p_3)\big) \\ & + \big(p_1-p_2\big)^2\big(\big[p_0-3p_3(1-p_3)+6p_1p_2\big]+(p_1-p_2)+3p_2^2+2(p_0-p_1)(1+3p_0)\big) \geq 0, \end{aligned} \quad \text{(A.140c)}$$

where the last inequality follows since the term in angled brackets in the last line can be shown to nonnegative using $6p_1p_2 \geq 6p_3^2$ and $3p_3(1-3p_3) \leq \frac{1}{4} \leq p_0$.

A.XIII. Outlook on the geometric method in higher dimensions

Here we present a possible recursive generalisation of the geometric approach, based on re-expressing condition (I) in terms of majorisation relations rather than trying to prove it in a full geometric way.

To illustrate this method we refer back to the proof of Theorem 3 for the case $d = 4$. We have seen that the point $\mathbf{v}_1 = (0, x_1(\beta), x_2(\beta), -1)$ can be reached with a transformation of the type $\mathbf{v}_1 = M_{q_1}\mathbf{q} + (\mathbb{1}+\Pi)\mathbb{1}\mathbf{r}_1 + \frac{1}{2}(\mathbb{1}+\Pi^2)\mathbb{1}\mathbf{r}_2$, where

$$M_{q_1} = \begin{pmatrix} m & 1-m & 0 & 0 \\ 1-m & m & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \quad \text{(A.141)}$$

with $m = 1 - 1/2(p_0+p_1)$, which is doubly stochastic since $p_0+p_1 \geq 1/2$ holds for $d = 4$.

To prove that $\mathbf{v}_2 = (0, 0, x_2(\beta), -1)$ can be reached, we also considered $M_{r_2} = \mathbb{1}$, which turns out to be a big simplification, due to the following reasoning. Assuming $M_{r_2} = \mathbb{1}$ we can rewrite the general transformation that (potentially) reaches $\mathbf{v}_2$ as

$$\mathbf{v}_2 + \mathbf{q} - \mathbf{p} - (\mathbb{1}+\Pi)(M_{r_1}-\mathbb{1})\mathbf{r}_1 = M_q\mathbf{q}, \quad \text{(A.142)}$$

where we used the fact that $\frac{1}{2}(\mathbb{1}+\Pi^2)\mathbf{r}_2 = \mathbf{p}-\mathbf{q}-(\mathbb{1}+\Pi)\mathbf{r}_1$. Thus, with this simplification of assuming $M_{r_2} = \mathbb{1}$, the problem reduces to showing that there exist some pair (M_q, M_{r_1}) such that Eq. (A.142) holds. In particular, we use the fact that $(1+\Pi)(M_{r_1}-\mathbb{1})$ has norm equal to zero, which in turn implies that the sum of vectors on the left hand side of Eq. (A.142) has the same norm as $\mathbf{q}$, since both $\mathbf{v}_2$ and $\mathbf{p}$ have norm 1 (i.e., they are probabilities). Note also that the vector $(\mathbb{1}+\Pi)(M_{r_1}-\mathbb{1})$ by itself need not be ordered with decreasing components, as well as the whole expression in the left hand side of Eq. (A.142). More precisely, in this case the greatest component of expression $\mathbf{v}_2+\mathbf{q}-\mathbf{p}$ is the third.

To prove the statement, then, we can show that $\mathbf{v}_2+\mathbf{q}-\mathbf{p}-(\mathbb{1}+\Pi)(M_{r_1}-\mathbb{1})\mathbf{r}_1$ is majorised by $\mathbf{q}$, or, more precisely, we have to find a proper stochastic matrix M_{r_1} such that the above majorisation holds. This is achieved again with a matrix of the form $M_{r_1} = M_{q_1}$ that leads to

$$(\mathbb{1}+\Pi)(M_{r_1}-\mathbb{1})\mathbf{r}_1 = (-a, 0, a, 0), \quad \text{(A.143)}$$

with $a = (m-1)p_1(p_0-p_2)$. Then, the first condition for the majorisation relation to hold is that the third component of the left hand side of Eq. (A.142) is smaller than the first component of $\mathbf{q}$, i.e., that

$$\frac{p_0+p_1-2p_2}{3} + p_2^2 + (m-1)p_1(p_0-p_2) \leq p_0^2, \quad \text{(A.144)}$$

which implies that we have to choose

$$1-m = \max\{0, \frac{p_0+p_1-2p_2-3(p_0^2-p_2^2)}{3p_1(p_0-p_2)}\}, \quad \text{(A.145)}$$

such that the majorisation relation is correctly satisfied, since the other conditions are also trivially satisfied.

The last step to check is that with this choice of the parameter $1-m$, M_{r_1} is indeed a doubly stochastic matrix. This is true because

$$1-m \leq 1 \iff p_0+p_1-2p_2-3(p_0^2-p_2^2) \leq 3p_1(p_0-p_2) \quad \text{(A.146)}$$

also holds, as well as $1-m \geq 0$, which holds by construction.

For higher dimensions, we can try a sort of recursive reasoning, that we work out here for the case $d = 5$. Let us hypotesise that the three vertices $(\mathbf{v}_1, \mathbf{v}_2, \mathbf{v}_3)$ can be found with a set of transformations $\{(M_{q_i}, (M_{r_1})_i, \mathbb{1})\}_{i=1,2,3}$, i.e., that are such that the last stochastic matrix is always the identity $(M_{r_2})_i = \mathbb{1}$ for all $i = 1, 2, 3$. This allows us to decompose the last vector as

$$(\mathbb{1}+\Pi^2)\mathbf{r}_2 = \mathbf{p}-\mathbf{q}-(\mathbb{1}+\Pi)\mathbf{r}_1, \quad \text{(A.147)}$$

that is $\mathbf{p}-\mathbf{q}$ minus a sum of terms that have zero norm. Then, we rewrite the condition for a vertex v_i to be reached as

$$\mathbf{v}_i - \mathbf{p} + \mathbf{q} - (\mathbb{1}+\Pi)((M_{r_1})_i - \mathbb{1})\mathbf{r}_1 = M_{q_i}\mathbf{q}, \quad \text{(A.148)}$$

which is satisfied if and only if the left hand side is majorised by $\mathbf{q}$. So now, the task would be to find suitable doubly stochastic matrices $(M_{r_1})_i$ such that the left hand side is majorised by $\mathbf{q}$ for each of the $\mathbf{v}_i$. Let us then start by considering $\mathbf{v}_1$, which in the probability coordinates is

$\mathbf{v}_1 = ((p_0+p_1)/2, (p_0+p_1)/2, p_2, p_3, p_4)$. In this case, the greatest component of $\mathbf{v}_i - \mathbf{p} + \mathbf{q}$ is the second, namely $\frac{p_0-p_1}{2} + p_1^2$. Unlike lower dimensions, however, in this case we have that $\frac{p_0-p_1}{2} + p_1^2 \leq p_0^2 \iff p_0 + p_1 \geq 1/2$ does not always hold, meaning that this time $(M_{r_1})_1 = \mathbb{1}$ does not suffice. Thus, in general we have to find $(M_{r_1})_1$ such that the second component of the vector $\tilde{\mathbf{r}}_1 := (\mathbb{1}+\Pi)((M_{r_1})_1 - \mathbb{1})\mathbf{r}_1$ is negative. The simplest would be if $\tilde{\mathbf{r}}_1 = (-a, a, 0, 0, 0)$ for some positive a. However, it can be readily checked that this is not possible. Nevertheless, we can look for vectors, for example, of the form $\tilde{\mathbf{r}}_1 = (a_1, a_2, -a_1 - a_2, 0, 0)$, for positive a_2 such that we can satisfy $\frac{p_0-p_1}{2} + p_1^2 - a_2 \leq p_0^2$ by choosing

$$a_2 = \max\{0, (p_0 - p_1)(\tfrac{1}{2} - p_0 - p_1)\}. \tag{A.149}$$

Afterwards, the next condition for the majorisation to hold is either

$$p_0^2 + p_1^2 - a_1 - a_2 \leq p_0^2 + p_1^2, \tag{A.150}$$

or

$$\tfrac{p_0-p_1}{2} + p_1^2 + p_2^2 + a_1 \leq p_0^2 + p_1^2. \tag{A.151}$$

While the first is trivially satisfied if a_1 and a_2 are both positive, the second needs to be checked. After this, all other conditions for the majorisation will be trivially satisfied. This can be achieved with the following matrix:

$$(M_{r_1})_1 = \begin{pmatrix} 1-m_1 & m_1 & 0 & 0 & 0 \\ 0 & 1-m_1 & m_1 & 0 & 0 \\ m_1 & 0 & 1-m_1 & 0 & 0 \\ 0 & 0 & 0 & 1-m_2 & m_2 \\ 0 & 0 & 0 & m_2 & 1-m_2 \end{pmatrix}, \tag{A.152}$$

with

$$\begin{aligned} a_2 &= m_2 p_4 (p_0 - p_3) = m_1(p_0 p_1 - p_2 p_3), \\ a_1 &= m_1 p_2 (p_1 - p_3), \end{aligned} \tag{A.153}$$

and we have to ensure the following conditions (only in the case $1/2 \geq p_0 + p_1$), coming from $m_1 \leq 1$ and $m_2 \leq 1$

$$\begin{aligned} &(p_0 - p_1)(\tfrac{1}{2} - p_0 - p_1)/(p_0 p_1 - p_2 p_3) \leq 1, \\ &(p_0 - p_1)(\tfrac{1}{2} - p_0 - p_1)/(p_4(p_0 - p_3)) \leq 1, \end{aligned} \tag{A.154}$$

plus we have to guarantee that Eq. (A.151) is satisfied, which leads to

$$(p_0-p_1)(\tfrac{1}{2}-p_0-p_1)p_2(p_1-p_3)/(p_0p_1-p_2p_3) \leq p_0^2 - p_2^2 - \tfrac{p_0-p_1}{2}, \tag{A.155}$$

since $a_1 = (p_0 - p_1)(\frac{1}{2} - p_0 - p_1)p_2(p_1 - p_3)/(p_0 p_1 - p_2 p_3)$. The above conditions are nontrivial, but still satisfied, as it is shown below in Sec. A.XIII.a.

Let us now come to $\mathbf{v}_2$. For this case we can try to re-use the result of $d = 4$, and show that the majorisation relation

$$\mathbf{v}_2 - \mathbf{p} + \mathbf{q} + (\mathbb{1} + \Pi)((M_{r_1})_2 - \mathbb{1})\mathbf{r}_1 = M_{q_2}\mathbf{q}, \tag{A.156}$$

holds for

$$(M_{r_1})_2 = \begin{pmatrix} m & 1-m & 0 & 0 & 0 \\ 1-m & m & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}, \tag{A.157}$$

with the appropriate coefficient $1 - m$, which is exactly the same as in the previous case, namely

$$1 - m = \max\{0, \frac{p_0 + p_1 - 2p_2 - 3(p_0^2 - p_2^2)}{3p_1(p_0 - p_2)}\}, \tag{A.158}$$

and again we have to ensure that

$$1 - m \leq 1 \iff p_0 + p_1 - 2p_2 - 3(p_0^2 - p_2^2) \leq 3p_1(p_0 - p_2). \tag{A.159}$$

However, while Eq. (A.159) holds alwasy true for $d = 4$, this is no longer the case for $d = 5$. Still, there is a range of parameters $\{E_i\}_{i=1}^5$ where it holds (see also Sec. A.XIII.a below).

Last, for the point $\mathbf{v}_2$ we can consider a matrix similar to the above, namely

$$(M_{r_1})_3 = \begin{pmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & m & 1-m & 0 & 0 \\ 0 & 1-m & m & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}, \tag{A.160}$$

but now the condition to be satisfied is $\frac{p_0+p_1+p_2+p_3}{4} - p_3 + p_3^2 - (1-m)p_2(p_1 - p_3) \leq p_0^2$ which leads to

$$1 - m = \max\{0, \frac{p_0 + p_1 + p_2 - 3p_3 - 4(p_0^2 - p_3^2)}{4p_2(p_1 - p_3)}\}. \tag{A.161}$$

The requirement $1 - m \leq 1$ thus translates to

$$p_0 + p_1 + p_2 - 3p_3 - 4(p_0^2 - p_3^2) \leq 4p_2(p_1 - p_3). \tag{A.162}$$

While this last relation might not hold in general, it can be satisfied in some appropriately chosen range of the parameters $\{E_i\}_{i=1}^4$. Thus, in order to fully prove the $d = 5$ case with this method, one would need to find other possible $(M_{r_1})_2$ and $(M_{r_1})_3$ for the respective complementary parameter regions. Nevertheless, this can possibly still be achieved while keeping $M_{r_2} = \mathbb{1}$.

In conclusion, we have seen a possible strategy to prove that all the vertices listed in condition (I) can be reached with the transformations preserving equal marginals. This exploits the geometry of such achievable thermal marginals and tries to avoid the difficulties encountered in the "passing on the norm approach, by considering the assumption that it is always possible to choose one of the matrices M_{r_i} as the identity. This assumption is in fact supported by the geometric proofs in dimensions $d = 3$ and $d = 4$.

A.XIII.a. *Validity of doubly stochasticity of M_{r_i} for $d = 5$*

Here we study the validity of the relations that guarantee that the matrices M_{r_i} needed for the proof of $d = 5$ are indeed doubly stochastic. We start with Eq. (A.146), which was also needed for the $d = 4$ case. To prove Eq. (A.146) we can see that

$$p_0 + p_1 - 2p_2 - 3(p_0^2 - p_2^2) \leq 3p_1(p_0 - p_2). \tag{A.163}$$

The above relation in fact holds since we can rewrite it as

$$(p_0 - p_2)(1 - 3(p_0 + p_1 + p_2)) + (p_1 - p_2) \leq 0, \tag{A.164}$$

and we can see that indeed

$$\begin{aligned}(p_0 - p_2)(1 - 3(p_0 + p_1 + p_2)) + (p_1 - p_2) \leq \\ (p_0 - p_2)(2 - 3(p_0 + p_1 + p_2)) \leq 0,\end{aligned} \tag{A.165}$$

since $p_0+p_1+p_2 \geq 2/3$ holds for $d = 4$, but not always $d = 5$. In fact, for $d = 5$ we can have points close to $p_0 = p_1 = p_2 = 1/5$.

To prove Eq. (A.154), we have to prove that, whenever $\frac{1}{2} \geq p_0 + p_1$ it holds that

$$(p_0 - p_1)(\tfrac{1}{2} - p_0 - p_1) \leq p_4(p_0 - p_3) \leq p_0p_1 - p_2p_3. \tag{A.166}$$

This is indeed true, since we can rewrite it as

$$(p_0-p_1)(\tfrac{1}{2}-p_0-p_1)-p_4(p_0-p_3) \leq (p_0-p_1)(\tfrac{1}{2}-p_0-p_1-p_4) \leq 0, \tag{A.167}$$

where the last relation holds whenever $\frac{1}{2} \geq p_0 + p_1$ because we can write

$$\tfrac{1}{2} - p_0 - p_1 - p_4 = p_2 + p_3 - \tfrac{1}{2} \leq p_0 + p_1 - \tfrac{1}{2} \leq 0. \tag{A.168}$$

Let us know consider Eq. (A.155). Since $p_2(p_1 - p_3) \leq (p_0p_1 - p_2p_3)$ we can rewrite it as

$$(p_0 - p_1)(\tfrac{1}{2} - p_0 - p_1) \leq p_0^2 - p_2^2 - \tfrac{p_0-p_1}{2}, \tag{A.169}$$

which is always satisfied since $p_0 - p_1 \leq 2(p_0^2 - p_1^2) \leq 2p_0^2 - p_2^2 - p_1^2$.

Finally, let us consider Eq. (A.162). We can rewrite the expression as

$$(p_0 - p_3)(1 - 4(p_0 + p_3)) + (p_1 - p_3)(1 - 4p_2) + p_2 - p_3 \leq 0, \tag{A.170}$$

and again, we can see that

$$\begin{aligned}(p_0 - p_3)(3 - 4(p_0 + p_2 + p_3)) \leq \\ (p_0 - p_3)(1 - 4(p_0 + p_3)) + (p_1 - p_3)(1 - 4p_2) + p_2 - p_3\end{aligned} \tag{A.171}$$

and in particular both the above expressions are surely negative whenever

$$p_0 + p_2 + p_3 \geq \tfrac{3}{4}, \tag{A.172}$$

which is not always true, but it is so in a certain range of parameters.

II.6 Ideal Projective Measurements Have Infinite Resource Costs

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Overview: Here, we developed a framework for characterizing measurements in a model where the system is unitarily coupled to an apparatus initially in a thermal state. We identify three properties of ideal measurements, which are (i) unbiased, (ii) faithful, and (iii) non-invasive, which cannot simultaneously be satisfied if the measurement apparatus has a nonzero initial temperature. We study the resulting non-ideal measurements and the relationship between the three properties, and show that ideal measurements require either infinite energy, time or control complexity. Our measurement model provides a simple tool to estimate lower bounds on the energy costs of measurements with any desired quality (how 'faithful' these measurements are), which show that the involved resource costs cannot be neglected when considering measurements as a means of supplying work to a quantum system.

Contribution: I jointly developed the ideas and concepts for this work with my colleagues, in particular, the definition of the three properties and their relationship. I helped writing the main paper, and I wrote Appendices I-VII and proved the results therein.

Ideal Projective Measurements Have Infinite Resource Costs

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We show that it is impossible to perform ideal projective measurements on quantum systems using finite resources. We identify three fundamental features of ideal projective measurements and show that when limited by finite resources only one of these features can be salvaged. Our framework is general enough to accommodate any system and measuring device (pointer) models, but for illustration we use an explicit model of an N-particle pointer. For a pointer that perfectly reproduces the statistics of the system, we provide tight analytic expressions for the energy cost of performing the measurement. This cost may be broken down into two parts. First, the cost of preparing the pointer in a suitable state, and second, the cost of a global interaction between the system and pointer in order to correlate them. Our results show that, even under the assumption that the interaction can be controlled perfectly, achieving perfect correlation is infinitely expensive. We provide protocols for achieving optimal correlation given finite resources for the most general system and pointer Hamiltonians, phrasing our results as fundamental bounds in terms of the dimensions of these systems.

1 Introduction

The foundations of any physical theory are laid by its axioms, postulates and laws. In quantum theory, the projection postulate presents one of these central pillars. It says that upon measuring a quantum system, its post-measurement state is given by an eigenstate of the measured observable and the corresponding probability for obtaining this state is given by the Born rule. In this way, an ideal projective measurement leaves the system in a pure state that is perfectly correlated with the measurement outcome.

Similarly, the key tenets of thermodynamics are formed by its three fundamental laws. Intense efforts in quantum thermodynamics [1–3] have placed these laws on rigorous mathematical footing [4–13]. Of particular interest is the third law of thermodynamics in the quantum regime, which tells us that no quantum system can be cooled to the ground state (which, in non-degenerate cases, is a *pure* state) in finite time and with finite resources [11, 12, 14–17]. This is in apparent contradiction with the projection postulate [18] — how is it that an ideal, error-free, measurement leaves the system in a state forbidden by the laws of thermodynamics?

In reality, we know that measurements in the lab are performed in finite time and with finite resources. These measurements are prone to small errors, implying that the post-measurement state of the system is never truly pure. However, with technological advances making errors ever smaller, one would assume rising thermodynamic costs as the post-measurement state of the system approaches purity.

Here, we resolve this apparent contradiction. We show that the resource cost of an ideal quantum measurement in a finite temperature environment is indeed infinite. Our operational approach is based on correlations between a system and a pointer, allowing us to make quantifiable statements about the cost. Within this framework we identify that an ideal projective measurement has three model-independent properties; it is: *unbiased*, *faithful*, and *non-invasive* — properties that cannot hold simultaneously for measurements with finite resources (energy and time). Our framework is general enough to accommodate any measurement model for which we provide quantitative results for an example case. In doing so, we refrain from making statements about what is commonly perceived as the 'measurement problem' (how or why the system is left in a particular state and what it means to obtain a 'result' [19, 20]).

Past approaches to quantifying the cost of a quantum measurement have typically assumed that projective measurements can be carried out *perfectly* and that their cost can be attributed to the work value of the measurement outcome [5, 21–24]. Others adopt the stance that Landauer's erasure bound represents the cost of resetting devices to pure states [4, 25, 26], without providing conclusive evidence that the bound is achievable. These works assume an unlimited supply of pure states, circumventing the third law of thermodynamics and resulting in finite energy costs. However, when limited to thermal environments, measurements

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produce errors, which can be mitigated by either reducing the temperature of the environment, or by using larger measuring devices. Both strategies can be quantified in terms of their thermodynamic cost, for which we provide exact analytic results. Our results demonstrate that even the simplest quantum measurements on qubits are never for free.

2 Ideal measurements

Consider an unknown quantum system ρ_S and a measuring device (*pointer*) ρ_P. To measure the system, one must couple it to the pointer and effect a joint transformation that correlates them: $\rho_S \otimes \rho_P \longrightarrow \tilde{\rho}_{SP}$. In an *ideal* measurement, the system and pointer become perfectly correlated, such that upon "observing" the pointer, one infers which pure state the system is in with probability 1. More precisely, each eigenstate $|i\rangle_S$ of the measured observable of the system is assigned a set $\{|\tilde{\psi}_n^{(i)}\rangle_P\}_n$ of orthogonal states of the pointer corresponding to a projector $\Pi_i = \sum_n |\tilde{\psi}_n^{(i)}\rangle\langle\tilde{\psi}_n^{(i)}|$. The projectors are orthogonal, forming a complete set, $\Pi_i\Pi_j = \delta_{ij}\Pi_i$ and $\sum_i \Pi_i = \mathbb{1}_P$. Upon finding the pointer in a state $|\tilde{\psi}_n^{(i)}\rangle_P$ (chosen to reflect $|i\rangle_S$), one concludes that the measurement outcome is "i", and that the system is left in the state $|i\rangle_S$. Up to arbitrary off-diagonal elements w.r.t. the basis $\{|i\rangle\otimes|\tilde{\psi}_n^{(j)}\rangle\}_{i,j,n}$, the ideal post-interaction state with perfect correlation has the form

$$\tilde{\rho}_{SP} = \sum_i \rho_{ii}\, |i\rangle\langle i| \otimes \rho^{(i)} + \text{off-diag.}\,, \tag{1}$$

where $\rho_{ii} = \langle i|\rho_S|i\rangle$ are diagonal elements w.r.t. the basis $\{|i\rangle\}_S$ and $\rho^{(i)}$ is a pointer state, associated to one and only one of the outcomes i, i.e., $\Pi_i\rho^{(j)} = \delta_{ij}\rho^{(j)}$. The form of $\tilde{\rho}_{SP}$ in Eq. (1) is the result of an *ideal measurement* and can be entangled or simply classically correlated. This ideal measurement satisfies three fundamental properties:

(i) **Unbiased**. The probability of finding the pointer in a state associated with outcome i after the interaction is the same as the probability of finding the system in the state $|i\rangle_S$ before the interaction,

$$\mathrm{tr}\big[\mathbb{I}\otimes\Pi_i\tilde{\rho}_{SP}\big] = \mathrm{tr}\big[|i\rangle\langle i|_S\,\rho_S\big] = \rho_{ii} \quad \forall i\ \forall\rho_S. \tag{2}$$

A measurement is unbiased if the pointer reproduces the measurement statistics of the system.

(ii) **Faithful**. There is a one-to-one correspondence between the pointer outcome and the post-measurement system state

$$C(\tilde{\rho}_{SP}) := \sum_i \mathrm{tr}\big[|i\rangle\langle i|\otimes\Pi_i\,\tilde{\rho}_{SP}\big] = 1 \quad \forall\rho_S, \tag{3}$$

i.e., $\tilde{\rho}_{SP}$ has perfect correlation: on observing the pointer outcome i (associated to Π_i), one concludes that the system is left in the state $|i\rangle_S$ with certainty.

(iii) **Non-invasive**. The probability of finding the system in the state $|i\rangle_S$ is the same before and after the interaction with the pointer,

$$\mathrm{tr}\big[|i\rangle\langle i|_S\,\tilde{\rho}_{SP}\big] = \mathrm{tr}\big[|i\rangle\langle i|_S\,\rho_S\big] = \rho_{ii} \quad \forall\, i\ \forall\rho_S. \tag{4}$$

This property only holds for the basis $|i\rangle_S$ and coherences appearing on the off-diagonal can, in general, be destroyed.

These three properties, stated here without particular hierarchy, capture the pairwise relation between (i) the pre-measurement system state and the measurement outcome, (3) between the measurement outcome and the post-measurement system state, and (4) between the pre- and post-measurement system states, respectively.

All quantitative statements we make about the faithfulness of a measurement [property (3)] depend on the function $C(\tilde{\rho}_{SP})$ in Eq. (3). This function's value represents the average probability of correctly inferring the post-measurement state upon observing the pointer, which is 1 for any unbiased measurement if and only if the post-interaction state is of the form of $\tilde{\rho}_{SP}$ in Eq. (1). One could choose more complicated functions or even measures of correlation. However, in our paradigm it is sufficient to be classically correlated to have perfect 'correlation' in the sense that $C = 1$. Note that quantum correlations are not strictly necessary, since $C(|0\rangle_S|0\rangle_P) = 1$. The advantage of the expression in Eq. (3) is that it quantifies the probability that the pointer indicates an outcome which is *correct* and yields the maximal value 1 if and only if the post-interaction state is of the form of Eq. (1).

Example. Consider a measurement of a qubit system using a single qubit pointer in the ground state. We model the measurement with a controlled NOT operation $U_{\text{CNOT}} = |0\rangle\langle 0|_S \otimes \mathbb{1}_P + |1\rangle\langle 1|_S \otimes X_P$, where $X = |0\rangle\langle 1| + |1\rangle\langle 0|$. The post-measurement state $\tilde{\rho}_{SP} = U_{\text{CNOT}}\ (\rho_S \otimes |0\rangle\langle 0|_P)\ U_{\text{CNOT}}^\dagger$ is of the form of Eq. (1), meaning the measurement is unbiased, faithful, and non-invasive. Indeed, whenever both system and pointer have dimension $d_S = d_P = d$, and the pointer is initially in a pure state (w.l.o.g. the ground state), we can define a unitary $U_d := |0\rangle\langle 0|_S \otimes \mathbb{1}_P + \sum_{i\neq 0}|i\rangle\langle i|_S \otimes X_P^{(i)}$, where $X_P^{(i)} = |0\rangle\langle i| + |i\rangle\langle 0| + \sum_{j\neq 0,i}|j\rangle\langle j|$, realizing an ideal measurement. A more detailed discussion of this example is presented in Appendix A.II.

3 Non-ideal measurements

We call a measurement in which any of the properties (i) – (iii) fails to hold *non-ideal*. This is due to the fact that, in general, the properties do not imply one another, i.e., satisfying a single property does not imply any of the other two, as illustrated in Fig. 1. Things become more subtle when a *pair* of properties is satisfied. In two cases, satisfying a pair of properties implies the third. As we show in detail in Appendix A.III, a measurement which is faithful and unbiased is also non-invasive, and a measurement that is faithful and non-invasive is unbiased. However, in general, a measurement being unbiased and non-invasive for a particular input state *does not* imply it is faithful, unless it is unbiased and non-invasive *for all* input states ρ_S, as illustrated in Fig. 1.

In what follows we prove that faithful measurements (perfect correlations) are possible if and only if one can prepare states with sufficiently many vanishing eigenvalues. Since, by the third law of thermodynamics, one cannot prepare states of non-full rank with finite resources, property (ii) fails to hold and therefore ideal measurements are not physically feasible. To see this, consider the most general interaction between a system and pointer — a completely positive and trace-preserving (CPTP) map, which can be understood as a unitary on the system and an extended pointer. In order for such a unitary to realize a faithful measurement according to Eq. (3), the rank of the final state $\tilde{\rho}_{SP}$ must be bounded from above by the dimension of the pointer d_P (with $d_P \geq d_S$), since $d_S \leq \sum_i \mathrm{rank}(\rho^{(i)}) \leq d_P$. When $d_S = d_P = d$, this implies that the initial rank of the pointer ρ_P must be 1, i.e., a pure state. For larger pointers, their initial state need not be pure, but it cannot have full rank — one must have $\mathrm{rank}(\rho_P) \leq d_P/d_S$. Practically, this requires pure state preparation for some non-trivial pointer subspaces. Thus, faithful and therefore ideal measurements are not possible without a supply of pure states (states at absolute zero temperature). States with non-full rank require infinite time, energy or complexity (interaction range) to be prepared and are prohibited by the third law of thermodynamics [11, 12, 14–17]. Conversely, whenever the pointer state does not start with full rank, operations such as U_d allow one to achieve perfect correlation.

Since faithful (and hence also ideal) measurements are not possible, we want to determine how closely they can be approximated. Since laboratory experiments take place at non-zero temperature, the natural state of a pointer is in thermodynamic equilibrium with its environment, i.e., the state $\tau_P(\beta)$, with inverse temperature $\beta = 1/k_\mathrm{B}T$. At any finite temperature, a thermal state has full rank, and any deviation from it requires an input of work.

While faithfulness is not necessarily the most important property, it is the one that certainly cannot be upheld in practice, whereas one of the other two properties can in principle be maintained also for practical measurements. Here, we take the point of view that the crucial property to demand of *any* measurement is to be *unbiased*. This guarantees that with sufficient repetitions, one obtains a mean value for the measured observable that accurately reflects the mean value of the underlying system ρ_S and the degree of trust in this outcome can be quantified using standard statistical methods. Conversely, if one imposed that the measurement were non-invasive, one would be able to perform repeated measurements on the system without changing the statistics of the measured observable. However, without properties (i) or (ii) it would not be possible to reliably relate the measurement data to statements about ρ_S. We therefore consider non-ideal measurements between a system ρ_S and a thermal pointer $\tau_P(\beta)$ with the property that the measurements are *unbiased*. In Appendix A.IV, we derive the general structure of maps realizing unbiased measurements in full detail.

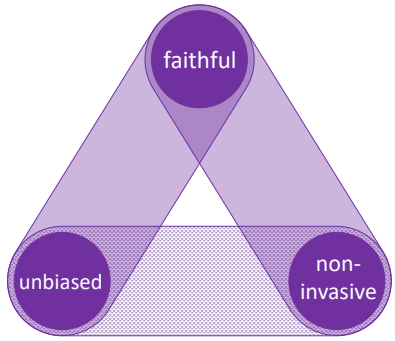

Figure 1: The properties attributed to an ideal measurement. In a non-ideal measurement these three properties do not hold simultaneously, and satisfying one of them does not imply any of the other two. When ρ_S is fixed, in two out of three cases, satisfying a pair of properties implies that the third property also holds. A measurement which is faithful and unbiased implies that it is also non-invasive, and a measurement that is faithful and non-invasive implies that it is unbiased. When ρ_S is relaxed to *all* initial system states, then satisfying any pair of properties implies the third, and one recovers the last relation, namely that a measurement which is unbiased and non-invasive (*for all* ρ_S) implies that it is also faithful.

4 Maximally faithful measurements

After imposing the measurements to be unbiased, we are then interested in the ones that produce the best correlations, in other words those which are as close to faithful as possible and thus approximate ideal measurements in a meaningful way. For such unbiased maximally correlating measurements, one may then determine and minimize the finite energy cost. To provide a self-contained description of this cost, we

consider the joint system of S and P to be closed, implying that we restrict to unitary maps. In Appendix A.V we further discuss this restriction and show that such unitarily correlating unbiased measurements are indeed possible at any temperature. Moreover, we find that unitarily correlating unbiased measurements have a maximal achievable correlation $C_{\max}$ that can only be reached if sufficient energy is supplied. To see this, let us denote the system and pointer Hamiltonians by H_S and H_P and the corresponding dimensions by d_S and d_P. For unbiased measurements, the restriction to unitary maps $\rho_S \otimes \tau_P \to \tilde{\rho}_{SP}$ implies a truncation of d_P at an integer multiple of d_S. We then order the spectrum of the pointer Hamiltonian and divide it into d_S disjoint sets $\{E_i^{(k)}\}_{i=0,\dots,d_P/d_S-1}$ for $k = 0,\dots,d_S-1$, such that $H_P = \sum_{k=0}^{d_S-1}\sum_{i=0}^{d_P/d_S-1} E_i^{(k)} \,|\, E_i^{(k)} \rangle\langle E_i^{(k)} \,|$ and $E_i^{(k)} \geq E_j^{(k')}$ whenever $k \geq k'$ or when $k = k'$ and $i \geq j$. Up to swaps between degenerate energies, the set $\{E_i^{(0)}\}_i$ contains the d_P/d_S smallest energies and consequently $\{\frac{1}{\mathcal{Z}} e^{-\beta E_i^{(0)}}\}_i$ are the largest populations. These populations are assigned to the 'correlated subspace'. For unbiased unitaries, there is an algebraic maximum to the achievable correlations between any system and thermal pointer, given by

$$C_{\max}(\beta) = \sum_{i=0}^{d_P/d_S-1} e^{-\beta E_i^{(0)}}/\mathcal{Z}\,, \tag{5}$$

which is independent of ρ_{ii} precisely due to unbiasedness. A more detailed derivation can be found in Appendix A.VII.

Note that, because we assigned the largest populations to the 'correlated subspace', $C_{\max}$ can be interpreted as the maximum probability of the post-measurement system being in the same state as the pointer. For an arbitrary unbiased measurement generally $C(\tilde{\rho}_{SP}) \leq C_{\max}(\beta)$. For an arbitrary unbiased measurement achieving $C_{\max}$ one can select a pointer basis $\{|\tilde{\psi}_i^{(k)}\rangle\}_{i,k}$ such that the resulting state can be written

$$\tilde{\rho}_{SP} = \sum_{k=0}^{d_S-1} \frac{\rho_{kk}}{\mathcal{Z}} \Big(\sum_{i=0}^{d_P/d_S-1} e^{-\beta E_i^{(0)}} \,|\, k \rangle\langle k \,| \otimes |\, \tilde{\psi}_i^{(k)} \rangle\langle \tilde{\psi}_i^{(k)} \,| + \sum_{m\neq k} \sum_{i=0}^{d_P/d_S-1} e^{-\beta \tilde{E}_{i,m}} U_{\mathrm{nc}}^{(k)} \,|\, m \rangle\langle m \,| \otimes |\, \tilde{\psi}_i^{(k)} \rangle\langle \tilde{\psi}_i^{(k)} \,|\, U_{\mathrm{nc}}^{(k)\dagger} \Big), \tag{6}$$

where the $U_{\mathrm{nc}}^{(k)}$ for $k = 0,\dots,d_S$ are unitaries on the non-correlated subspaces spanned by the vectors $|\, m \rangle\, |\tilde{\psi}_i^{(k)}\rangle$ for $i = 0,\dots,d_P/d_S-1$ and $m \neq k$, $\{\tilde{E}_{i,m\neq k}\}_{i,m} = \{E_i^{(n>0)}\}_{n,i}$. From this form, we see that perfect correlation $C = 1$ is only possible if the pointer temperature reaches absolute zero, or, more generally, if the rank of the pointer is bounded by $\mathrm{rank}(\rho_P) \leq d_P/d_S$. Note that the way the system state is altered through measurement is not completely fixed by Eq. (6). The relation between the bases $\{\tilde{\psi}_i^{(k)}\}_{i,k}$ and $\{|\, E_i^{(k)} \rangle\}_{i,k}$, as well as the choices of $U_{\mathrm{nc}}^{(k)}$ and the ordering of the energies $\tilde{E}_{i,m}$ leave room for adjusting the final energy cost.

The exact form of the unbiased and maximally (but not perfectly) correlated lowest energy states depends both on ρ_S and requires diagonalization of H_P. We now present a solution for an example case and refer to Appendix A.IX for a detailed step-by-step instruction.

5 Energy cost of measurements

We now investigate the relation between the energy cost ΔE of an unbiased measurement achieving the maximum correlation $C_{\max}$ between a system and the pointer. Here, we wish to showcase different ways of increasing $C_{\max}$, which depends on temperature and dimension. It is readily seen from Eq. (5) that $C_{\max}$ increases when the pointer size is increased at fixed temperature or when the initial temperature is lowered at fixed pointer size. While Eq. (6) provides a general form for $\tilde{\rho}_{SP}$, quantitative insight about ΔE cannot be gained without fixing the pointer Hamiltonian. An exception is when the pointer dimension is infinite. There, the third law of thermodynamics can be circumvented by using (a part of) the pointer as a 'fridge' and creating asymptotically pure subspaces (see, e.g., [27]), a scenario which we include as a limiting case in our analysis. For the general case we refer to Appendix A.IX, but here, as a concrete example (described in more detail in Appendix A.VIII), we will consider a single-qubit system and a pointer consisting of N initially non-interacting qubits with identical Hamiltonians H_P.

We have $\tilde{\rho}_{SP} = U_{\mathrm{corr}}\,(\rho_S \otimes \tau_P(\beta)^{\otimes N})\, U_{\mathrm{corr}}^\dagger$, where from now on we take $\tau_P(\beta) = 1/\mathcal{Z}_P(|\,0\rangle\langle 0\,| + e^{-\beta E_P}\,|\,1\rangle\langle 1\,|)$ with the partition function $\mathcal{Z}_P = \mathrm{tr}[e^{-\beta H_P}]$. Since we would like to increase $C_{\max}$ as much as possible and we know that $C_{\max}$ depends on the initial temperature of the pointer, we also consider cooling the pointer prior to the correlating interaction in order to get closer to a faithful result.

In principle, there exist many models of refrigeration, e.g., [16]. To achieve ground state cooling, however, some form of resource has to diverge. For instance, infinite time is required in adiabatic Landauer erasure [1], infinite energy in finite size fridges [16], or infinite complexity (or time) in fridges of infinite size [27]. In short, quantum measurements inherit the limitations imposed by the third law of thermodynamics [12, 15]. Since our example also aims to quantify the energy cost of correlations with increasing pointer size N, including fridges of arbitrary size may compromise statements about correlations costs at fixed N. We therefore consider a fridge model of the same type and size as the pointer, i.e., for each pointer qubit we add one fridge qubit, see Fig. 2. For larger refriger-

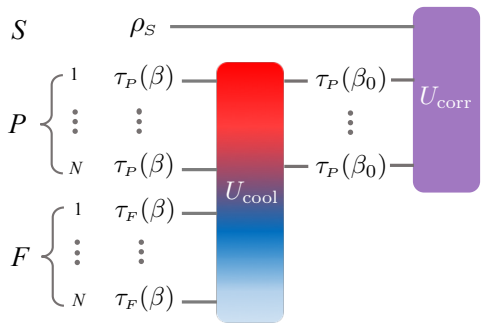

Figure 2: The measurement procedure. In step I an N-qubit pointer is coupled to an N-qubit fridge and cooled from β to β_0. In step II, a unitary correlates the pointer with the unknown qubit system.

ation systems the cost of cooling could be decreased by a constant factor (see [16]), but would still diverge as one approaches zero temperature unless the fridge size is itself infinite.

Within this framework, we describe the measurement by two consecutive unitary operations, which we call *cooling* and *correlating*. The total transformation on the system, pointer, and fridge is $U_{\text{tot}}\left(\rho_S \otimes \tau_P(\beta)^{\otimes N} \otimes \tau_F(\beta)^{\otimes N}\right) U_{\text{tot}}^\dagger$, where $\tau_F(\beta) = 1/\mathcal{Z}_F(|0\rangle\langle 0| + e^{-\beta E_F}|1\rangle\langle 1|)$ and $U_{\text{tot}} = (U_{corr} \otimes \mathbb{I}_F) \cdot (\mathbb{I}_S \otimes U_{\text{cool}})$, see Fig. 2. Both unitaries drive the respective systems out of equilibrium and come at a thermodynamic cost. Neglecting the price for perfect control over these operations, the work cost of implementing them is lower-bounded by the total energy change of the system, pointer, and fridge, $W \geq \Delta E$. The total cost in energy can be split into the sum of the two parts: cooling and correlating, which we write $\Delta E = \Delta E_{\text{I}} + \Delta E_{\text{II}}$. Our objective is to minimise ΔE when performing a non-ideal measurement for a fixed value of the correlation function $C(\tilde{\rho}_{SP}) = C_{\max} < 1$.

6 Minimal energy cost

To minimise ΔE_{I} we use Ref. [16], which details the optimal cost for the single-qubit fridge. Cooling the pointer from $T = 1/\beta$ to $E_P/(\beta E_F)$ such that $\tau_P(\beta)^{\otimes N} \mapsto \tau_P(\beta \frac{E_F}{E_P})^{\otimes N}$ requires at least

$$\Delta E_{\text{I}} = N(E_F - 1)\left(\frac{1}{e^{-\beta E_F}+1} - \frac{1}{e^{-\beta E_P}+1}\right). \quad (7)$$

To minimise ΔE_{II} we are interested in determining $\min_{U_{\text{corr}}} \Delta E_{\text{II}}$ such that $C(\tilde{\rho}_{SP}) = C_{\max}(\beta)$. For the case of a single-qubit system and N-qubit pointer (with N odd), we have

$$C_{\max}(\beta) = \frac{1}{\mathcal{Z}^N} \sum_{k=0}^{N/2} \binom{N}{k} e^{-kE_P\beta}. \quad (8)$$

For even N, the formula is slightly different with the same qualitative behaviour. As expected, in the limit of infinite pointer size ($N \to \infty$) for fixed β, or in the limit of zero temperature ($\beta \to \infty$) for any N, the correlations become perfect, $\lim_{N\to\infty} C_{\max}(\beta) = \lim_{\beta\to\infty} C_{\max}(\beta) = 1$. In Appendix A.VIII, we construct the optimal unitary U_{opt} that solves the optimisation problem for ΔE_{II} for arbitrary N and β, i.e., the unitary that achieves the algebraic maximum correlation for minimal energy cost. In particular, this construction allows us to specify an analytic expression for ΔE_{II} in terms of β, N, and ρ_S, which implies an achievable lower bound on the minimal energy cost of non-ideal measurements that approximate ideal ones as well as possible.

Note that in the limit $N \to \infty$ the energy cost of achieving $C_{\max}(\beta)$ is finite but infinite time (or full control over N-body interactions with $N \to \infty$) is required (see Appendix A.VIII). For any finite N, the only way to achieve correlations higher than $C_{\max}(\beta)$ is to cool the pointer. Thus, we consider the scenario where starting at some finite β, we cool the pointer ($\beta \to \beta_0 > \beta$) and then correlate it with the system to the algebraic maximum for the new temperature $C_{\max}(\beta_0)$. Results for exemplary temperatures are shown in Fig. 3 for $N = 6$. Within our cooling paradigm, the energy cost for reaching the ground state in finite time is infinite. Other paradigms allow cooling to the ground state using finite energy, but require infinite time [27]. Thus, without access to pure states, a measurement satisfying properties (i) – (iii) has an infinite resource cost. The cost for the maximally correlating unitary U_{corr} is always finite and given by $\Delta E_{\text{II}}^{(C=1)} = \frac{1}{2}E_P$.

7 Discussion

The projection postulate is a central concept within the foundations of quantum mechanics, asserting that ideal projective measurements leave the system in a pure state corresponding to the observed outcome. All interpretations of quantum mechanics must be compatible with this statement together with the Born rule assigning the probabilities. However, the existence of such 'true' projections is usually taken for granted. Here, we have discussed a self-contained description of measurements from a thermodynamic point of view. We have shown that, when their existence is not assumed, ideal finite-time projective measurements have an infinite cost.

We argued that a necessary condition for ideal measurements is to be faithful, i.e., have perfect correlation between the system and pointer. However, this requirement incurs infinite costs unless pure states are freely available. Nonetheless, ideal measurements can be approximated by non-ideal, unbiased ones to arbitrary precision at finite energy cost. We find that the correlation achieved by the best unbiased mea-

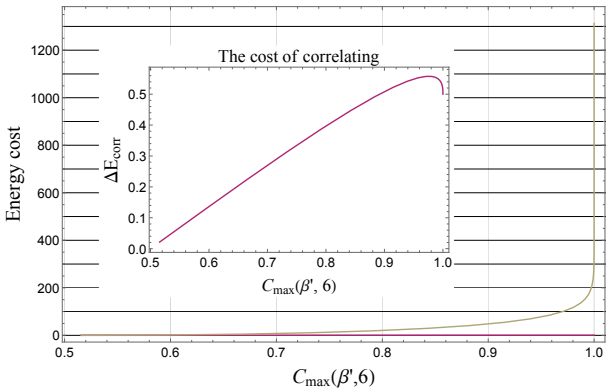

Figure 3: Cost of a non-ideal projective measurement of a qubit system using a 6-qubit pointer. Each pointer qubit is in the state $\tau_P(\beta)$. We start from room temperature (≈ 300 K) and choose an energy gap in the microwave regime such that $\beta E_P = 1/30$. Each point on the horizontal indicates the maximal algebraic correlation $C_{\max}(\beta', N = 6)$ achievable for a fixed value $\beta' = \frac{\beta E_F}{E_P}$ (or equivalently, fixed E_F), which is the result of cooling the 6-qubit pointer from β to β' using refrigeration qubits with gaps E_F. For each correlation value, the refrigeration cost $\Delta E_{\rm I}$ and the cost $\Delta E_{\rm II}$ of maximally correlating the thermal state at inverse temperature β' are shown. The inset shows the relevant energy scale for correlating the system and pointer since the cooling cost significantly dominates the correlating cost.

surement is universally bounded by the largest d_P/d_S eigenvalues of the pointer. To gain quantitative insight into this cost, we considered the measurement of a single qubit by an N-qubit pointer. We provided analytic expressions for the minimal energy cost for unitarily achieving maximal correlation for any initial temperature and any N. We find that correlations can be increased by increasing the pointer size and by cooling the pointer.

While the three mentioned properties capture the basic features of ideal measurements, they are not sufficient to characterise the 'quantum to classical' transition. Classical outcomes additionally feature 'robustness', where small perturbations of the pointer do not significantly alter the observed outcomes. This is an important consideration for broadcasting measurement outcomes. In our qubit model, this is taken into account by the size of the subspaces the system is correlated with, i.e., the number of pointer particles, N.

The insight that ideal measurements carry a diverging cost also sheds light on thermodynamics because it implies that, in practice, all measurements that can be performed are intrinsically non-ideal. Consequently, a central question is how well one can approximate ideal measurements and which consequences these approximate realizations have on tasks that arise in quantum thermodynamics. For instance, to interpret work as a random variable in the quantum regime, two projective measurements are commonly assumed to characterize work [28–30]. The impossibility of these measurements with finite work, prompts two questions (recently studied in [31]): (a) what is the impact of measurement imperfection on the observed fluctuations and (b) what is the total work cost of observing work fluctuations imperfectly. Furthermore, in quantum information-based engines [24, 32, 33], it would be highly relevant to incorporate measurement imperfection and work cost into the efficiency in addition to other constraints [34]. These insights could also be useful for quantum information processing, e.g., bounding the minimal power consumption of quantum computers employing syndrome measurements for error correction.

Acknowledgments

We are grateful to Tiago Debarba and Karen Hovhannisyan for valuable comments and insights. We acknowledge support by the EU COST Action MP1209 "Thermodynamics in the quantum regime" and from the Austrian Science Fund (FWF) through the START project Y879-N27, the project P 31339-N27, and the joint Czech-Austrian project MultiQUEST (I 3053-N27 and GF17-33780L). YG acknowledges support by the Austrian Science Fund (FWF) through the Zukunftskolleg ZK03.

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Appendix: Mathematical Model for Measurement Procedures

In this Appendix, we give a detailed description of *unbiased* measurement procedures introduced in the main text. As we have argued, ideal measurements (unbiased, faithful, and non-invasive) are not generally implementable in finite time or with finite energy. In practice, real measurements may nonetheless approximate ideal measurements by investing energy; loosely speaking, the approximation becomes better, the more energy that is invested. To make this more precise, we will explicitly determine the fundamental energy cost of projective[1] measurements.

[1]Arbitrary quantum measurements represented by positive-operator-valued measures (POVMs) can be realized by projective measurements on a Hilbert space obtained by appending an auxiliary system of, at most [35], the same dimension as the original system. We therefore concentrate on projective measurements.

A.I Framework

System. We consider a quantum system S with Hilbert space $\mathcal{H}_S$ of dimension $d_S = \dim(\mathcal{H}_S)$ initially in an arbitrary unknown quantum state represented by a density operator $\rho_S \in \mathcal{L}(\mathcal{H}_S)$, i.e., a Hermitian operator with $\mathrm{tr}(\rho_S) = 1$ in the space of linear operators $\mathcal{L}(\mathcal{H}_S)$ over the system Hilbert space $\mathcal{H}_S$. We are then interested in describing (projective) measurements of the system w.r.t. a basis $\{|i\rangle_S\}_i$ of $\mathcal{H}_S$, which we take to be the eigenbasis of the system Hamiltonian H_S, i.e., we can write $H_S = \hbar\sum_i \Omega_i |i\rangle\langle i|_S$, where $\hbar(\Omega_j - \Omega_i)$ is the energy gap between the i-th and j-th levels. For instance, an example that we will focus on later is that of the simplest quantum system — a qubit — with vanishing ground state energy and energy gap $E_S = \hbar\Omega$. That is, $\mathcal{H}_S = \mathbb{C}^2$, and the system Hamiltonian H_S has eigenstates $|0\rangle_S$ and $|1\rangle_S$ and spectral decomposition $H_S = E_S |1\rangle\langle 1|_S$.

Pointer. Similarly, we consider a pointer system P with Hilbert space $\mathcal{H}_P$ of dimension $d_P = \dim(\mathcal{H}_P)$ and Hamiltonian H_P. We then take the resource-theoretic point of view that the pointer is initially in a state that is freely available, i.e., a thermal state $\tau_P(\beta) \in \mathcal{L}(\mathcal{H}_P)$ at ambient temperature $T = 1/\beta$. We order the spectrum of the pointer Hamiltonian in terms of its excitations into d_S sectors of size d_P/d_S, i.e., $H_P = \sum_{k=0}^{d_S-1}\sum_{i=0}^{d_P/d_S-1} E_i^{(k)} |E_i^{(k)}\rangle\langle E_i^{(k)}|$ with $E_i^{(k)} \leq E_j^{(k')}$ $\forall i,j$ for $k' > k$. The thermal (Gibbs) state is given by

$$\tau_P(\beta) = \sum_{k=0}^{d_S-1}\sum_{i=0}^{d_P/d_S-1} p_i^{(k)} |E_i^{(k)}\rangle\langle E_i^{(k)}| \tag{A.1}$$

where $p_i^{(k)} = \exp\big(-\beta E_i^{(k)}\big)/\mathcal{Z}$ and $\mathcal{Z}$ is the pointer's partition function $\mathcal{Z} = \mathrm{tr}(e^{-\beta H_P}) = \sum_{i,k} e^{-\beta E_i^{(k)}}$.

Measurement procedure. We now wish to consider a measurement of the system's energy, i.e., of the observable H_S, or, in other words, a projective measurement of the system in the energy eigenbasis[2]. The corresponding *measurement procedure* may be defined via a completely positive and trace-preserving (CPTP) map $\mathcal{E} : \mathcal{L}(\mathcal{H}_{SP}) \to \mathcal{L}(\mathcal{H}_{SP})$, where $\mathcal{H}_{SP} = \mathcal{H}_S \otimes \mathcal{H}_P$, that maps $\rho_{SP} = \rho_S \otimes \tau_P$ to a post-measurement state $\tilde{\rho}_{SP} \in \mathcal{L}(\mathcal{H}_{SP})$. This may be understood as a generalized interaction between the system, the pointer, and some auxiliary system. Here, we do not wish to address the question of which measurement outcome is ultimately realized (which pure state the system is left in), or how and why

[2]Note that a projective measurement of the system in any other (orthonormal) basis can be subsumed into this discussion by including an additional unitary transformation (and its energy cost) on the initial state ρ_S to switch between the energy eigenbasis and the desired measurement basis.

this may be the case. That is, we do not attempt to make statements about what is often perceived as the "measurement problem", but rather take the point of view that system and pointer are left in a joint state in which the internal states of the system are correlated with the internal states of the pointer. Each of the latter is designated to correspond to one of the system states $|i\rangle_S$, such that, upon finding the pointer in a state chosen to reflect $|i\rangle_S$, one concludes that the measurement outcome is "i". More precisely, each eigenstate $|i\rangle_S$ of the measured observable of the system is assigned a set $\{|\tilde{\psi}_n^{(i)}\rangle_P\}_n$ of orthogonal states of the pointer corresponding to a projector $\Pi_i = \sum_n |\tilde{\psi}_n^{(i)}\rangle\langle\tilde{\psi}_n^{(i)}|$. The projectors are chosen to be orthogonal and to form a complete set, i.e., $\Pi_i\Pi_j = \delta_{ij}\Pi_i$ and $\sum_i \Pi_i = \mathbb{1}_P$. In an ideal measurement, upon obtaining the outcome "i", one may further conclude that the post-measurement *system* is left in the state $|i\rangle_S$. This is one of three features that can be identified for ideal projective measurements. As explained in the main text, ideal measurements are

unbiased:

$$\mathrm{tr}\big[\mathbb{1}_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] = \mathrm{tr}\big[|i\rangle\langle i|_S\, \rho_S\big] = \rho_{ii}\, \forall\, i, \quad \text{(A.2)}$$

faithful:

$$C(\tilde{\rho}_{SP}) := \sum_i \mathrm{tr}\big[|i\rangle\langle i|_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] = 1, \quad \text{(A.3)}$$

non-invasive:

$$\mathrm{tr}\big[|i\rangle\langle i|_S \otimes \mathbb{1}_P\, \tilde{\rho}_{SP}\big] = \mathrm{tr}\big[|i\rangle\langle i|_S\, \rho_S\big] = \rho_{ii}\, \forall\, i. \quad \text{(A.4)}$$

A.II Example: 2-Qubit Measurements

To illustrate the properties above and to understand why these conditions are not met by general non-ideal measurement procedures with finite energy input, we consider a simple example. Consider a measurement procedure using a single pointer qubit and assume that by some means it has been prepared in the ground state, i.e., $\rho_P = |0\rangle\langle 0|_P$. We model the interaction with the pointer by applying a controlled NOT operation $U_{\text{CNOT}} = |0\rangle\langle 0|_S \otimes \mathbb{1}_P + |1\rangle\langle 1|_S \otimes X_P$, with the usual Pauli operator $X = |0\rangle\langle 1| + |1\rangle\langle 0|$. Denoting the matrix elements of the initial state as $\rho_{ij} = \langle i|\rho_S|j\rangle$, we can then write the post-measurement state $\tilde{\rho}_{SP}$ as

$$\tilde{\rho}_{SP} = U_{\text{CNOT}}\, \rho_{SP} U_{\text{CNOT}}^\dagger = \sum_{i,j=0,1} \rho_{ij}\, |ii\rangle\langle jj|\,. \quad \text{(A.5)}$$

The system and pointer are now perfectly (classically) correlated in the sense that whenever the pointer is found in the state $|0\rangle_P$ ($|1\rangle_P$), the system is left in the corresponding state $|0\rangle_S$ ($|1\rangle_S$). In other words, for the choice $\Pi_i = |i\rangle\langle i|_P$, we find that the measurement is faithful,

$$C(\tilde{\rho}_{SP}) = \sum_{i=0,1} \mathrm{tr}\big[|ii\rangle\langle ii|\, \tilde{\rho}_{SP}\big] = \sum_{i=0,1} \rho_{ii} = \mathrm{tr}[\rho_S] = 1\,. \quad \text{(A.6)}$$

The post-measurement system state $\tilde{\rho}_S = \mathrm{tr}_P[\tilde{\rho}_{SP}] = \sum_i \rho_{ii}\, |i\rangle\langle i|_S$ is in general different from the initial system state since it not longer has any off-diagonal elements w.r.t. the measurement basis, but the measurement is nonetheless non-invasive since the diagonal elements match those of the initial system state ρ_S. At the same time, the chosen unitary U_{CNOT} guarantees that the probabilities for finding the pointer in the states $|0\rangle_P$ and $|1\rangle_P$, match those of the original system state, i.e., for $i = 0, 1$ we have

$$\mathrm{tr}\big(|i\rangle\langle i|_P\, \mathrm{tr}_S(\tilde{\rho}_{SP})\big) = \mathrm{tr}(|i\rangle\langle i|_S\, \rho_S) = \rho_{ii}\,. \quad \text{(A.7)}$$

Consequently, the measurement is not biased towards one of the outcomes and reproduces the statistics of the original system state, while being perfectly correlated (i.e., faithful).

However, in general strong correlation and unbiasedness of the measurement do not imply one another. For instance, one can construct an unbiased but also generally uncorrelated measurement by replacing U_{CNOT} with $U_{\text{SWAP}} = |00\rangle\langle 00| + |01\rangle\langle 10| + |10\rangle\langle 01| + |11\rangle\langle 11|$, leaving the system in the state $|0\rangle_S$ no matter which state the pointer is in. Although all available information about the pre-measurement system is thus stored in the pointer, measuring the latter reveals no (additional) information about the post-measurement system. Alternatively, consider the unitary $|1\rangle\langle 1|_S \otimes \mathbb{1}_P + |0\rangle\langle 0|_S \otimes X_P$ instead of U_{CNOT}, both of which lead to the same correlation $C(\tilde{\rho}_{SP})$, but the probabilities for observing the two outcomes are now exchanged w.r.t. to ρ_S, i.e., the pointer is found in the state $|0\rangle_P$ ($|1\rangle_P$) with probability ρ_{11} (ρ_{00}) after the interaction.

For the purpose of examining real measurements, these examples are of course pathological due to the assumption of reliably preparing the pointer in a pure state (without having to have performed a projective measurement in order to model a projective measurement or having to cool to the ground state using finite resources [12]). Let us therefore relax this assumption and assume instead that the pointer has been prepared at some finite non-vanishing temperature $T = 1/\beta$, such that $\rho_P = p\,|0\rangle\langle 0|_P + (1-p)\,|1\rangle\langle 1|_P$ for some $p = (1 + e^{-\omega\beta})^{-1} = \mathcal{Z}^{-1}$ with $0 < p < 1$. A quick calculation then reveals that the previously perfect correlations are reduced to $C(\tilde{\rho}_{SP}) = p = \mathcal{Z}^{-1} < 1$ and that the measurement procedure using U_{CNOT} is in general biased, i.e.,

$$\mathrm{tr}\big[|i\rangle\langle i|_P\, \mathrm{tr}_S[\tilde{\rho}_{SP}]\big] = \rho_{ii}(2p-1) + 1 - p\,. \quad \text{(A.8)}$$

However, while we generally have to give up the notion of a perfect projective measurement in the

sense that the outcomes are perfectly correlated with the post-measurement states (as we have shown in the main text), one may retain the unbiasedness of the measurement. That is, if we replace U_{CNOT} by $U_{\text{unb.}} = |00\rangle\langle 00| + |01\rangle\langle 11| + |11\rangle\langle 10| + |10\rangle\langle 01|$, we obtain the same imperfect correlation value $C(\tilde{\rho}_{SP}) = p = \mathcal{Z}^{-1}$, but the unbiasedness condition of Eq. (A.7) is satisfied. To reiterate, measurement procedures using finite resources (finite time, finite energy, and finite complexity, i.e., operations with finite interaction range), cannot be ideal, since finite resources are not sufficient to prepare pointers in the required pure states. Realistic measurement procedures hence are non-ideal.

A.III Non-Ideal Measurement Procedures

When any one of the three properties (A.2), (A.3) or (A.4) fails to hold, we call the corresponding measurement procedure *non-ideal*. For non-ideal measurements, the relation between the remaining properties is more complicated. In particular, none of the three properties alone implies any of the other two. For instance, consider an ideal post-interaction state

$$\tilde{\rho}_{SP} = \sum_i \rho_{ii}\, |i\rangle\langle i| \otimes \rho^{(i)} + \text{off-diag.}\,, \tag{A.9}$$

where we have not explicitly written the off-diagonal elements w.r.t. the basis $\{|i\rangle \otimes |\tilde{\psi}_n^{(j)}\rangle\}_{i,j,n}$ and $\rho^{(i)}$ is a pointer state that is associated to one and only one of the outcomes i, that is, $\Pi_i \rho^{(j)} = \delta_{ij}\rho^{(j)}$. Any measurement procedure based on a map $\mathcal{E}$ for which the values ρ_{ii} in (A.9) are replaced with arbitrary probabilities $p_i \neq \rho_{ii}$, results in a joint post-interaction state $\tilde{\rho}_{SP}$ satisfying (A.3), but not (A.2) or (A.4), resulting in a non-ideal measurement that is faithful, but neither unbiased or non-invasive. Similarly, the state $\tilde{\rho}_{SP} = \rho_S \otimes \rho_P$ obtained from a trivial interaction $\mathcal{E}[\rho_{SP}] = \rho_{SP}$ complies with (A.4), but not with (A.3) or (A.2). Finally, the map $\mathcal{E}$ realizing a complete exchange of the initial system and pointer states (assuming, for the purpose of this example that $d_S = d_P$), results in an unbiased (A.2) measurement procedure that does not satisfy either (A.3) or (A.4).

Satisfying any single one of the three properties is hence not sufficient for distinguishing ideal from non-ideal measurements. When two out of the three properties hold, things become more subtle. In two cases, a joint final state $\tilde{\rho}_{SP}$ satisfying a *pair* of properties implies the third property, and hence that the measurement is ideal for the particular given initial system state ρ_S. First, a measurement that is both faithful (A.3) *and* unbiased (A.2) implies that it is also non-invasive (A.4). To show this, we start with the property of unbiasedness and, summing the right-hand side of Eq. (A.2) over all i, we have $\sum_i \rho_{ii} = 1$. The left-hand side of Eq. (A.2) thus gives

$$\sum_i \operatorname{tr}\big[\mathbb{1}_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] = \sum_{i,j} \operatorname{tr}\big[|j\rangle\langle j|_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] = 1. \tag{A.10}$$

At the same time, property (A.3) demands that the sum in the second step of (A.10) yields 1 already just for the terms where $i = j$, implying

$$\sum_{i\neq j} \operatorname{tr}\big[|j\rangle\langle j|_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] = 0. \tag{A.11}$$

Since all diagonal matrix elements of a density operator are non-negative, this further implies $\operatorname{tr}\big[|j\rangle\langle j|_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] = 0\ \forall i \neq j$, which we can insert back into (A.2) to see that $\operatorname{tr}\big[|i\rangle\langle i|_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] = \rho_{ii}$. Inserting all of this into the left-hand side of Eq. (A.4) together with $\mathbb{1}_P = \sum_j \Pi_j$, we obtain

$$\operatorname{tr}\big[|i\rangle\langle i|_S \otimes \sum_j \Pi_j\, \tilde{\rho}_{SP}\big] = \operatorname{tr}\big[|i\rangle\langle i|_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] = \rho_{ii}, \tag{A.12}$$

which concludes the proof that unbiased and faithful measurements are also non-invasive.

Second, a measurement that is both faithful (A.3) *and* non-invasive (A.4) is also unbiased (A.2). Now starting with (A.4), we again sum the left-hand side over all i and resolve the identity $\mathbb{1}_P = \sum_j \Pi_j$, to obtain

$$\sum_{i,j} \operatorname{tr}\big[|i\rangle\langle i|_S \otimes \Pi_j\, \tilde{\rho}_{SP}\big] = \sum_i \rho_{ii} = 1. \tag{A.13}$$

This time, unbiasedness (A.2) implies

$$\sum_{i\neq j} \operatorname{tr}\big[|i\rangle\langle i|_S \otimes \Pi_j\, \tilde{\rho}_{SP}\big] = 0, \tag{A.14}$$

and in turn $\operatorname{tr}\big[|i\rangle\langle i|_S \otimes \Pi_j\, \tilde{\rho}_{SP}\big] = 0\ \forall i \neq j$, as before. Inserting this into (A.4) then implies $\operatorname{tr}\big[|i\rangle\langle i|_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] = \rho_{ii}$. Finally inserting into the left-hand side of (A.2) yields

$$\begin{aligned}\operatorname{tr}\big[\mathbb{1}_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] &= \sum_j \operatorname{tr}\big[|j\rangle\langle j|_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] \\ &= \operatorname{tr}\big[|i\rangle\langle i|_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big] = \rho_{ii},\end{aligned} \tag{A.15}$$

confirming the unbiasedness condition.

For the remaining combination this is not the case. A measurement procedure that is unbiased (A.2) and non-invasive (A.4) for a fixed system state ρ_S is not necessarily faithful (A.3). Consider, e.g., the initial single-qubit system state $\rho_S = \frac{3}{4}|0\rangle\langle 0| + \frac{1}{4}|1\rangle\langle 1|$, i.e., where $\rho_{00} = \frac{3}{4}$ and $\rho_{11} = \frac{1}{4}$, and the two-qubit final state $\tilde{\rho}_{SP} = \sum_{m,n=0,1} |m,n\rangle\langle m,n|$ with $p_{01} = p_{10} = p_{11} = \frac{1}{8}$ and $p_{00} = \frac{5}{8}$. For $\Pi_i = |i\rangle\langle i|_P$, one has the reduced states $\operatorname{tr}_S(\tilde{\rho}_{SP}) = \operatorname{tr}_P(\tilde{\rho}_{SP}) = \rho_S$, so we have unbiasedness and non-invasiveness, but $C(\tilde{\rho}_{SP}) = \frac{3}{4} < 1$.

Moreover, the measurement procedure (corresponding to the transformation above (whose details are not given in the example) may not be unbiased or non-invasive for other initial system states ρ_S. This singles out the property of faithfulness when one is interested in checking the properties of a measurement procedure for *any* given initial system state. Nonetheless, caution is advisable here. For a particular initial state ρ_S of the system, all three properties may be satisfied, yet, this may not be so for other initial states. Simply consider the example in Eq. (A.8). For an initial state with $\rho_{00} = \rho_{11} = \frac{1}{2}$, the measurement satisfies all three properties, but for $p < 1$ the measurement procedure is biased and has non-perfect correlation for almost all (other) states ρ_S.

Indeed, demanding that any of the properties hold only for particular initial system states ρ_S is somewhat contradictory to the notion of performing a measurement that reveals previously unknown information about a system. In other words, measurements should not require detailed knowledge about ρ_S to ensure that one may trust the measurement outcomes, or inferences made from them. The definitions of the properties (A.2), (A.3) and (A.4) must hence be extended to demand that measurement procedures are only called unbiased, faithful, or non-invasive, if the respective properties (A.2), (A.3) or (A.4) hold *for all initial systems states* ρ_S.

With such an extended definition, one then indeed finds that any two properties imply the third. In particular, it is then the case that measurement procedures that are unbiased *and* non-invasive, are also faithful, and thus ideal. The proof of this statement, relies on insights into the general structure of all maps representing unbiased measurement procedures, and as such appears later in Appendix A.VI. In purely qualitative terms, maps that are either unbiased or non-invasive need to transfer the diagonal elements of the system state ρ_S to particular (different) subspaces of the joint Hilbert space of system and pointer. The only way to simultaneously satisfy both the requirements for unbiasedness and non-invasiveness for arbitrary ρ_S forces all information to be concentrated in the subspaces corresponding to the images of the projectors $|i\rangle\langle i| \otimes \Pi_i$, such that the resulting state $\tilde{\rho}_{SP}$ satisfies (A.3) independently of the details of ρ_S.

Ultimately, this means that only one of the three properties can be satisfied exactly for all initial system states in any realistic measurement procedure. Given that the constraint of finite resources rules out that realistic non-ideal measurements are faithful, we have a choice between the measurement being unbiased or non-invasive. Arguably, biased measurements that are not even faithful are of little use, since the outcomes would not provide any level of certainty about either the pre- or post-measurement system state. In the following, we are therefore interested in unbiased measurement procedures for which the correlations between the system and the pointer are as large as possible. Given such non-ideal measurement, we then wish to minimize the associated energy costs.

A.IV General Unbiased Measurements

Here, we will identify the basic structure and important properties of a general model of non-ideal measurement procedures. To do so, we separate what we believe to accurately model such a measurement procedure into two steps. These are:

I **Preparation**: Some energy is invested to prepare the pointer system in a suitable quantum state.

II **Correlating**: The pointer interacts with the system to be measured, creating correlations between them.

A.IV.1 Step I: Preparation

Before interacting with the system, the pointer can be prepared in a suitable quantum state ρ_P at the expense of some initial energy investment ΔE_{I} accounting for the (CPTP) transformation $\mathcal{E}_{\mathrm{I}} : \mathcal{L}(\mathcal{H}_P) \to \mathcal{L}(\mathcal{H}_P)$ mapping $\tau(\beta)$ to $\rho_P = \mathcal{E}_{\mathrm{I}}[\tau(\beta)]$. In particular it may be desirable to lower the entropy of the initial pointer state. In principle, one may use any given amount of energy to prepare an arbitrary pointer state that is compatible with the specified energy and whose entropy is lower than that of $\tau(\beta)$. The energy cost for reaching a particular state ρ_P is bounded from below by the free energy difference, i.e.,

$$\Delta E_{\mathrm{I}} \geq \Delta F\big(\tau(\beta) \to \rho_P\big) = \Delta E_P - T\Delta S_P\,, \quad \text{(A.16)}$$

with $\Delta E_P = \mathrm{tr}\big[H_P(\rho_P - \tau(\beta))\big]$ and $\Delta S_P = S\big(\rho_P\big) - S\big(\tau(\beta)\big)$, and where $S(\rho) = -\mathrm{tr}\big[\rho\log(\rho)\big]$ is the von Neumann entropy. However, the exact work cost of the preparation depends on the control over the system and the available auxiliary degrees of freedom, and may exceed this bound. In particular, the free energy difference to the ground state is finite although this state cannot be reached with a finite work investment in finite time [12]. The precise resource requirements in terms of energy, control, and time for preparing arbitrary quantum states are hence difficult to capture[3], whereas the refrigeration of quantum systems is a well-understood task, whose energy cost has been quantified for various levels of control one assumes about the quantum systems involved in the cooling procedure [16].

[3]Certainly, any such preparation can be described by a CPTP map, which in turn can be seen as a unitary acting on $\rho_P \otimes |\Phi\rangle\langle\Phi|$ in a Hilbert space enlarged by an auxiliary system with Hilbert space $\mathcal{H}_A \ni |\Phi\rangle$. However, this brings one back to the question of quantifying the cost for preparing the pure state $|\Phi\rangle$ of the auxiliary system.

It is therefore practically useful (and reasonable) to assume that the preparation only involves refrigeration. That is, we assume in the following that the temperature of the pointer is lowered from T to $T_0 \leq T$, reaching a thermal state $\tau(\beta_0)$ with $\beta_0 = 1/T_0$. On the one hand, step I thus becomes less general than it could potentially be since one does not explore the entire Hilbert space $\mathcal{H}_P$. On the other hand, the thermal state can be considered to be energetically optimal, since it minimizes the energy at fixed entropy. Moreover, at fixed energy the thermal state also maximizes the entropy and hence minimizes the free energy, which in turn bounds the work cost from below.

A.IV.2 Step II: Correlating

During the second step of the measurement procedure, the system interacts with the pointer in such a way that correlations between the two are established via a CPTP map $\mathcal{E}_{\text{II}} : \mathcal{L}(\mathcal{H}_{SP}) \to \mathcal{L}(\mathcal{H}_{SP})$ that maps $\rho_{SP} = \rho_S \otimes \rho_P$ to $\tilde{\rho}_{SP} = \mathcal{E}_{\text{II}}[\rho_{SP}]$. A particularly important special case is the case of unitary correlating maps U, i.e., such that $\tilde{\rho}_{SP} = U\rho_{SP}U^\dagger$, representing measurement procedures where the joint system of S and P can be considered to be closed for the purpose of the correlating step. Then, the energy cost for the second step can be calculated via

$$\Delta E_{\text{II}} = \operatorname{tr}\big[(H_S + H_P)(\tilde{\rho}_{SP} - \rho_{SP})\big]. \tag{A.17}$$

In any case the generated correlations can in principle be (but need not be) genuine quantum correlations. For (non-ideal) projective measurements as defined here, it nonetheless suffices that classical correlations are established with respect to the measurement basis (here the eigenbasis of H_S) and a chosen basis of the pointer system. More specifically, we assign a set of orthogonal projectors

$$\Pi_i := \sum_n |\tilde{\psi}_n^{(i)}\rangle\langle\tilde{\psi}_n^{(i)}|, \tag{A.18}$$

with $\Pi_i\Pi_j = \delta_{ij}\Pi_i$ (in particular, $\langle\tilde{\psi}_m^{(i)}|\tilde{\psi}_n^{(j)}\rangle = \delta_{ij}\delta_{mn}$) and $\sum_i \Pi_i = \mathbb{1}_P$. The orthogonality and completeness of the projectors ensure that every pointer state is associated with a state of the measured system, i.e., every outcome provides a definitive measurement result "i". We further amend the correlations function defined in Eq. (A.6) for a single-qubit pointer to reflect the use of the more general projectors, i.e., we redefine the quantifier $C(\tilde{\rho}_{SP})$ as

$$C(\tilde{\rho}_{SP}) := \sum_i \operatorname{tr}\big[|i\rangle\langle i|_S \otimes \Pi_i\, \tilde{\rho}_{SP}\big]. \tag{A.19}$$

A.IV.3 Unbiased measurements

We are now in a position to give a formal definition of what we consider as an abstract measurement procedure.

Definition 1: Measurement procedure

A measurement procedure $\mathcal{M}(\beta)$ that realizes a (non-ideal) projective measurement at ambient temperature $T = 1/\beta$ of an (unknown) quantum state $\rho_S \in \mathcal{L}(\mathcal{H}_S)$ w.r.t. to an orthonormal basis $\{|n\rangle\}_n$ of $\mathcal{H}_S$ is given by the tuple $(\mathcal{H}_P, H_P, \Pi, \mathcal{E})$, consisting of a pointer Hilbert space $\mathcal{H}_P$, a pointer Hamiltonian $H_P \in \mathcal{L}(\mathcal{H}_P)$, a complete set $\Pi = \{\Pi_i\}_i$ of orthogonal projectors on $\mathcal{H}_P$, and a CPTP map $\mathcal{E} : \mathcal{L}(\mathcal{H}_{SP}) \to \mathcal{L}(\mathcal{H}_{SP})$ with $\mathcal{H}_{SP} = \mathcal{H}_S \otimes \mathcal{H}_P$, together with the induced CPTP map $\mathcal{E}_{\mathcal{M}} : \mathcal{L}(\mathcal{H}_S) \to \mathcal{L}(\mathcal{H}_{SP})$ given by

$$\mathcal{E}_{\mathcal{M}} : \ \rho_S \mapsto \tilde{\rho}_{SP} = \mathcal{E}[\rho_S \otimes \tau(\beta)]. \tag{A.20}$$

Note that any definition of a thermal state $\tau(\beta)$ implies that the state has full rank. This definition includes, in particular, the case that we consider here, where the map $\mathcal{E} = \mathcal{E}_{\text{II}} \circ (\mathbb{1}_S \otimes \mathcal{E}_{\text{I}})$ is split into two separate steps. As we have already motivated in our earlier example, we are interested in considering measurement procedures that represent the measured quantum state without bias. While perfect correlations cannot be guaranteed in this way, one may however ask that averages of the measured quantity match for the pointer and the system. Moreover, it is desirable that this is so independently of the specific initial states of the system and the pointer. All of this is captured by the following definition.

Definition 2: Unbiased measurement

A measurement procedure $\mathcal{M}(\beta)$ is called *unbiased*, iff $\operatorname{tr}\big[\Pi_i \operatorname{tr}_S(\tilde{\rho}_{SP})\big] = \operatorname{tr}[|i\rangle\langle i|_S\, \rho_S]\ \forall i$ and $\forall \rho_S$.

Since we wish to restrict our further considerations to unbiased measurements, it will be useful to know more about the structure of these measurement procedures, in particular, about the involved CPTP map $\mathcal{E}$ and projectors Π_i, given that one has selected a suitable pointer system with Hilbert space $\mathcal{H}_P$ and Hamiltonian H_P. To this end, note that our previous example using U_{CNOT} was unbiased only for pointers that can be prepared in the ground state (or any pure state for that matter). This can only be the case if the initial temperature vanishes or if infinite resources are available in step I, whereas we are interested in describing more realistic conditions. To capture this, we formalise the following:

Definition 3: Finite-resource measurement

A measurement procedure $\mathcal{M}(\beta)$ at a nonzero ambient temperature $T = 1/\beta$ uses *finite resources* if the map $\mathcal{E}_{\mathcal{M}}$ is rank non-increasing.

On the other hand, measurement procedures which *reduce* the rank use either infinite energy E, take infinite time t (an infinite sequence of finite interaction range operations) or are infinitely complex (infinite interaction range operations) [12, 14–17, 27]. Now, in order to take a first step towards unraveling the structure of unbiased measurements we formulate the following lemma.

Lemma 1

All unbiased finite-resource measurement procedures $\mathcal{M}(\beta)$ with (thermal, full-rank) pointer system with Hilbert space $\mathcal{H}_P$, Hamiltonian H_P, and orthogonal projectors Π_i can be realized by CPTP maps $\mathcal{E}$ of the form $\mathcal{E} = \mathcal{E}_{\text{II}} \circ (\mathbb{1}_S \otimes \mathcal{E}_{\text{I}})$, where $\mathcal{E}_{\text{I}}$ is a CPTP map from $\mathcal{L}(\mathcal{H}_P)$ to itself (achievable in finite time t and satisfying $\Delta E_{\text{I}} < \infty$), and the CPTP map $\mathcal{E}_{\text{II}}$ from $\mathcal{L}(\mathcal{H}_{SP})$ to itself has Kraus operators $K_l = \sum_i K_l^{(i)}$ with

$$K_l^{(i)} = \sum_{j=0}^{d_S-1} \sum_{n=0}^{d_P-1} \sum_{m=0}^{d_i-1} k_{jmn}^{(i,l)} \, |\, j \,\rangle\!\langle\, i \,|_S \otimes |\, \tilde{\psi}_m^{(i)} \,\rangle\!\langle\, \psi_n \,|_P \,, \tag{A.21}$$

with $d_S = \dim(\mathcal{H}_S)$, $d_P = \dim(\mathcal{H}_P) \geq d_S$, and coefficients $k_{jmn}^{(i,l)}$ such that

$$\sum_l (K_l^{(i)})^\dagger K_l^{(i)} = |\, i \,\rangle\!\langle\, i \,|_S \otimes \mathbb{1}_P. \tag{A.22}$$

Proof of Lemma 1. Before we get into the technical details of the proof, let us phrase the Lemma 1 more informally. It states that the map $\mathcal{E}$ consists of an arbitrary (finite energy, $\Delta E_{\text{I}} < \infty$, finite time $t < \infty$) preparation of the pointer ($\mathcal{E}_{\text{I}}$), followed by a map $\mathcal{E}_{\text{II}}$ that maps the subspaces $|\, i \,\rangle_S$ to those corresponding to Π_i, respectively. Moreover, note that unbiasedness of course implies that the pointer system must be large enough ($d_P \geq d_S$) to accommodate all the possible measurement outcomes. Let us then prove the lemma. As mentioned before, the CPTP map $\mathcal{E}$ may be separated into a map $\mathcal{E}_{\text{I}}$ acting nontrivially only on the pointer Hilbert space, and a CPTP map $\mathcal{E}_{\text{II}}$ acting on the resulting state $\rho_{SP} = \rho_S \otimes \rho_P$, which we can write with respect to the basis $|\, i \,\rangle_S$ as

$$\rho_{SP} = \begin{array}{c} \left(\begin{array}{c|c|c|c} \rho_{00}\rho_P & \cdot & \cdot & \cdots \\ \hline \cdot & \rho_{11}\rho_P & \cdot & \cdots \\ \hline \cdot & \cdot & \rho_{22}\rho_P & \cdots \\ \hline \vdots & \vdots & \vdots & \ddots \end{array}\right) \\ \begin{array}{cccc} |\,0\,\rangle_S & |\,1\,\rangle_S & |\,2\,\rangle_S & \cdots \end{array} \end{array}, \tag{A.23}$$

Without loss of generality, we can then write the final state $\tilde{\rho}_{SP} = \mathcal{E}_{\text{II}}[\rho_{SP}]$ with respect to the product basis $\{|\, i \,\rangle_S \otimes |\, \tilde{\psi}_m^{(j)} \,\rangle_P\}_{i,j,m}$ in the form

$$\begin{array}{c} \begin{array}{ccccccccccc} \Pi_0 & \Pi_1 & \Pi_2 & & \Pi_0 & \Pi_1 & \Pi_2 & & \Pi_0 & \Pi_1 & \Pi_2 \end{array} \\ \left(\begin{array}{ccc|c|ccc|c|ccc|c} A_{00} & \cdots & \cdot & \cdot & \cdot & \cdots & \cdot & \cdot & \cdot & \cdots & \cdot & \cdot \\ \vdots & A_{01} & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ \cdot & \cdots & A_{02} & \cdot & \cdot & \cdots & \cdot & \cdot & \cdot & \cdots & \cdot & \cdot \\ \cdot & \cdots & \cdot & \ddots & \cdot & \cdots & \cdot & \cdot & \cdot & \cdots & \cdot & \cdot \\ \cdot & \cdots & \cdot & \cdot & A_{10} & \cdots & \cdot & \cdot & \cdot & \cdots & \cdot & \cdot \\ \vdots & \ddots & \vdots & \vdots & \vdots & A_{11} & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ \cdot & \cdots & \cdot & \cdot & \cdot & \cdots & A_{12} & \cdot & \cdot & \cdots & \cdot & \cdot \\ \cdot & \cdots & \cdot & \cdot & \cdot & \cdots & \cdot & \ddots & \cdot & \cdots & \cdot & \cdot \\ \cdot & \cdots & \cdot & \cdot & \cdot & \cdots & \cdot & \cdot & A_{20} & \cdots & \cdot & \cdot \\ \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & A_{21} & \vdots & \vdots \\ \cdot & \cdots & \cdot & \cdot & \cdot & \cdots & \cdot & \cdot & \cdot & \cdots & A_{22} & \cdot \\ \cdot & \cdots & \cdot & \cdot & \cdot & \cdots & \cdot & \cdot & \cdot & \cdots & \cdot & \ddots \end{array}\right) \\ \begin{array}{ccc} |\,0\,\rangle_S & |\,1\,\rangle_S & |\,2\,\rangle_S \cdots \end{array} \end{array} \tag{A.24}$$

where we have indicated the columns corresponding to the subspaces of fixed vectors $|\, i \,\rangle_S$ and projectors Π_i for $i = 1, 2, 3$, and dots indicate matrix elements that may be nonzero but are not explicitly shown. In particular, the latter can include subspaces for $i > 3$, and the case for $d_S \leq 3$ can be obtained by truncating the shown matrix by removing the corresponding rows and columns. The colored sub-blocks A_{0i}, A_{1i}, A_{2i} and so forth are $d_i \times d_i$ block matrices in terms of which the unbiasedness condition of Def. 2 can be written as

$$\sum_{j=0}^{d_S-1} \text{tr}[A_{ji}] = \rho_{ii} \quad \forall i. \tag{A.25}$$

Crucially, the unbiasedness condition in Eq. (A.25) is to hold for all possible system states ρ_S, and hence for all possible values of ρ_{ii}. This, in turn, implies that all sub-blocks with second subscript i must be proportional to ρ_{ii}. That is $A_{ji} = \rho_{ii}\tilde{A}_{ji} \quad \forall j$ with $\sum_{j=0}^{d_S-1} \text{tr}[\tilde{A}_{ji}] = 1 \ \forall i$. Since the unbiasedness condition is not sensitive to terms appearing in the off-diagonal blocks, a convenient representation of the relevant terms of the post-interaction state $\tilde{\rho}_{SP}$ under the map $\mathcal{E}_{\text{II}}$ is :

$$\Gamma_{\mathcal{E}_{\text{II}}} = \left[\begin{array}{c|c|c|c} \rho_{00}\tilde{A}_{00} & \rho_{11}\tilde{A}_{01} & \rho_{22}\tilde{A}_{02} & \cdots \\ \hline \rho_{00}\tilde{A}_{10} & \rho_{11}\tilde{A}_{11} & \rho_{22}\tilde{A}_{12} & \cdots \\ \hline \rho_{00}\tilde{A}_{20} & \rho_{11}\tilde{A}_{21} & \rho_{22}\tilde{A}_{22} & \cdots \\ \hline \vdots & \vdots & \vdots & \ddots \end{array}\right], \tag{A.26}$$

which we call the *correlation matrix*. Here it can immediately be seen that the unbiasedness condition, which implies $\sum_{j=0}^{d_S-1} \text{tr}[\tilde{A}_{ji}] = 1$ says that the sum of the trace of the blocks in column i of $\Gamma_{\mathcal{E}}$ must be equal to ρ_{ii} for unbiasedness to hold[4].

[4] Note that this representation is not square, since in principle the dimension of the matrix-valued entries of each column are different. In the case that the map $\mathcal{E}$ representing the measurement is unitary, the representation becomes square and the dimension of all blocks across all columns is equal.

From the initial state (A.23) and the final state (A.24) it becomes apparent that unbiasedness can be guaranteed for maps that connect the subspaces corresponding to $|i\rangle_S$ only with those corresponding to Π_i. More precisely, each of these connecting maps can be viewed as an arbitrary CPTP map $\mathcal{E}_{\Pi}^{(i)}$ from the d_P-dimensional space spanned by the vectors in the set $\{|i\rangle_S \otimes |\psi_n\rangle_P\}_{n=0,\dots,d_P-1}$, where $\{|\psi_n\rangle_P\}_n$ is an arbitrary basis of $\mathcal{H}_P$, to the $(d_S \times d_i)$-dimensional space spanned by the vectors in the set $\{|j\rangle_S \otimes |\tilde{\psi}_m^{(i)}\rangle_P\}_{\substack{j=0,\dots,d_S-1\\ m=0,\dots,d_i-1}}$. The Kraus operators for these CPTP maps are precisely the $\{K_l^{(i)}\}_l$ of Eq. (A.21) and in matrix notation we may denote these maps as

$$\rho_{ii}\rho_P \overset{\mathcal{E}_{\Pi}^{(i)}}{\longmapsto} \left(\begin{array}{c|c|c|c} A_{0i} & \cdot & \cdot & \cdots \\ \hline \cdot & A_{1i} & \cdot & \cdots \\ \hline \cdot & \cdot & A_{2i} & \cdots \\ \hline \vdots & \vdots & \vdots & \ddots \end{array}\right) = \rho_{ii} \left(\begin{array}{c|c|c|c} \tilde{A}_{0i} & \cdot & \cdot & \cdots \\ \hline \cdot & \tilde{A}_{1i} & \cdot & \cdots \\ \hline \cdot & \cdot & \tilde{A}_{2i} & \cdots \\ \hline \vdots & \vdots & \vdots & \ddots \end{array}\right). \tag{A.27}$$

Since the domains as well as the images for different i lie in orthogonal subspaces of $\mathcal{H}_{SP}$, the maps $\mathcal{E}_{\Pi}^{(i)}$ can be combined to the map $\mathcal{E}_{\Pi}$ with Kraus operators[5] $\{K_l = \sum_i K_l^{(i)}\}_l$. Once can check that the unbiasedness condition is satisfied for these Kraus operators by a simple calculation, which we will not repeat here. If the initial ambient temperature is nonzero and the measurement procedure uses finite resources (time and energy), the pointer state ρ_P has full rank and unbiasedness can only be achieved via maps of the form mentioned, as claimed in Lemma 1, which concludes the proof. □

Inspecting again the example from Appendix A.II, one notes that the controlled NOT operation U_{CNOT} is not of the form required for a finite-resource unbiased measurement, as expected. However, when the pointer can be prepared in a pure state (w.l.o.g. the ground state $|0\rangle_P$) one observes that the measurement procedure using U_{CNOT} in the correlating step becomes unbiased because some of the sub-blocks A_{ji} are only trivially proportional to ρ_{ii}. In particular, $A_{01} = \tilde{A}_{01} = A_{10} = \tilde{A}_{10} = 0$.

Having understood the general structure of all unbiased measurements, we now want to turn to some specific instances of such measurement procedures.

A.V Extremal Measurements

With the help of Lemma 1 we can now describe the set of *all* unbiased measurements for a given quantum system ρ_S and pointer. The measurement within this set may further be categorized according to their specific properties, in particular, their energy cost, the amount of correlations created between the system and the pointer (how faithful the measurement is), and the level of control required for their implementation (e.g., what type of auxiliary systems are available and which operations can be performed on the system, pointer, and auxiliaries). Given an (unknown) quantum system S it would ideally be desirable to answer the question: What is the maximal correlation achievable between the system and *any suitable* pointer given a fixed work input ΔE? A more restricted version of this question is: What is the maximal correlation achievable between the system and *a particular* pointer given a fixed work input ΔE?

Since we assume that the system state ρ_S is unknown before the measurement, the correlation measure $\bar{C}$ that we are interested in optimizing is obtained from averaging $C(\tilde{\rho}_{SP})$ from Eq. (A.3) over all system states. We observe that for any particular systems state ρ_S, the correlation measure $C(\tilde{\rho}_{SP})$ does not depend on any of the matrix elements of ρ_S except for those on the diagonal, i.e.,

$$C(\tilde{\rho}_{SP}) = \rho_{00}\text{tr}[\tilde{A}_{00}] + \rho_{11}\text{tr}[\tilde{A}_{11}] + \rho_{22}\text{tr}[\tilde{A}_{22}] + \dots . \tag{A.28}$$

Averaging over all states ρ_S is hence equivalent to an average over all probability distributions corresponding to the diagonal of ρ_S. Moreover, for each of these values ρ_{ii} $(i = 0, \dots, d_S - 1)$, the average over all probability distributions results in the value $1/d_S$, such that the average of $C(\tilde{\rho}_{SP})$ is given by

$$\bar{C} = \tfrac{1}{d_S}\text{tr}\big[\tilde{A}_{00} + \tilde{A}_{11} + \tilde{A}_{22} + \dots\big], \tag{A.29}$$

which, in terms of the representation presented in Eq. (A.26), corresponds to taking the trace of the blocks appearing along the diagonal.

Despite this simple form of $\bar{C}$, the optimization over all pointer systems and operations thereon is a daunting task. Indeed, even for a fixed pointer at initial temperature $T = 1/\beta$, identifying the optimal measurement procedure in terms of the best ratio of (average) correlation increase per unit energy cost (averaged over the input system states) is highly nontrivial. To illustrate the difficulty, first note that an (attainable) bound exists for correlating (quantified by the mutual information) two arbitrary systems that are initially thermal at the same temperature at optimal energy expenditure [36, 37]. While the known protocol for attaining this bound is in general not unitary (it involves lowering the temperature), in some cases the bound is tight already when one correlates the systems unitarily. However, it was recently shown [38, 39] that the optimal (in the sense of the mentioned bound being tight) trade-off between correlations and energy cost cannot always be achieved unitarily.

Of course, in our case, the initial state of the system is not known, and cannot be expected to be thermal in general. Moreover, the mutual information is

[5] Note that the number of nonzero Kraus operators may be different for each i, but one may always add trivial (vanishing) Kraus operators to each set $\{K_l^{(i)}\}_l$ with fixed i.

not a suitable figure of merit for quantifying the desired correlations between system and pointer because the latter don't distinguish between classical and genuinely quantum correlations. For instance, for a single-qubit system and pointer, the states $|\Phi^+\rangle_{SP} = (|00\rangle + |11\rangle)/\sqrt{2}$ and $\rho_{SP} = \frac{1}{2}(|00\rangle\langle 00| + |11\rangle\langle 11|)$ have different values of mutual information but are equally well (i.e., perfectly) correlated w.r.t. to the desired measurement basis. The above arguments on optimally correlating protocols hence do not apply directly, but with the added complication of the unknown system state and the unbiasedness condition, we cannot rule out the possibility that the optimal unbiased measurement procedures are not realized by a unitary correlating step.

Nonetheless, it can be argued that any nonunitary realization of the second part $\mathcal{E}_{\text{II}}$ of the CPTP map $\mathcal{E}$ must require higher levels of control than a corresponding unitary realization due to the requirement of realizing nonunitary maps $\mathcal{E}_{\text{II}}$ as unitaries on a larger Hilbert space. Specifically, any CPTP map $\mathcal{E}_{\text{II}}$ can be thought of as a unitary on a larger Hilbert space $\mathcal{H}_{SP} \otimes \mathcal{H}_E$ (with a factoring initial condition) [40], that is, one may write any $\mathcal{E}_{\text{II}}$ as

$$\mathcal{E}_{\text{II}}[\rho_{SP}] = \operatorname{tr}_E\left[U_{SPE}\left[\rho_{SP} \otimes |\chi\rangle\langle\chi|\right]U_{SPE}^\dagger\right] \tag{A.30}$$

for some unitary U_{SPE} on $\mathcal{H}_{SP} \otimes \mathcal{H}_E$ and for some pure state $|\chi\rangle \in \mathcal{H}_E$. At the same time, employing a unitary to correlate pointer and system enables us to unambiguously quantify the work cost of the correlating step without assumptions about the Hamiltonian of potential auxiliary systems.

We are therefore particularly interested in describing all unbiased measurement procedures, where $\mathcal{E}_{\text{II}}$ is realized unitarily, such that

$$\tilde{\rho}_{SP} = \mathcal{E}_{\text{II}}[\rho_{SP}] = U\rho_{SP}U^\dagger \tag{A.31}$$

with $UU^\dagger = U^\dagger U = \mathbb{1}_{SP}$. In this sense, our focus lies on unbiased measurement procedures where all control that one may have over external systems (beyond S and P) is used in the initial step represented by $\mathcal{E}_{\text{I}}$ to prepare the pointer in a suitable state, e.g., by lowering its temperature. Here we make use of the fact that the work cost of refrigeration with various levels of control has been extensively studied [16]. This leaves us with the task of analyzing the structure of the representations U of the unitary maps $\mathcal{E}_{\text{II}}$.

A first step towards the completion of this task is to note that the unbiasedness condition for measurement procedures with a *unitary* correlating step $\mathcal{E}_{\text{II}}$ means that it is inefficient (in terms of energy cost) to use a pointer Hilbert space $\mathcal{H}_P$ whose dimension is not an integer multiple of the system dimension. This can be explained in the following way. By inspection of the maps in Eq. (A.27), one notes that $\mathcal{E}_{\text{II}}^{(i)}$ maps the $d_P \times d_P$ density matrix ρ_P to a $d_S \operatorname{rank}(\Pi_i) \times d_S \operatorname{rank}(\Pi_i)$ density matrix. That is, the size of each of the d_S blocks $\tilde{A}_{ji}$ $\forall j$ is determined by the rank of Π_i. If the map $\mathcal{E}_{\text{II}}$ is unitary, this implies that all $\mathcal{E}_{\text{II}}^{(i)}$ are unitary, and hence

$$d_P = d_S \operatorname{rank}(\Pi_i) \quad \forall i. \tag{A.32}$$

Conversely, this implies that all projectors Π_i have the same rank d_P/d_S, which must be an integer larger or equal to 1, $d_P = \lambda d_S$ for $\lambda \in \mathbb{N}$. The implication of this for the correlation matrix in (A.26) is that it is now square

$$\Gamma_{U_{\text{unb}}} = \left[\begin{array}{c|c|c|c} \rho_{00}\tilde{A}_{00} & \rho_{11}\tilde{A}_{01} & \rho_{22}\tilde{A}_{02} & \cdots \\ \hline \rho_{00}\tilde{A}_{10} & \rho_{11}\tilde{A}_{11} & \rho_{22}\tilde{A}_{12} & \cdots \\ \hline \rho_{00}\tilde{A}_{20} & \rho_{11}\tilde{A}_{21} & \rho_{22}\tilde{A}_{22} & \cdots \\ \hline \vdots & \vdots & \vdots & \ddots \end{array}\right]. \tag{A.33}$$

In principle, one could initially consider a pointer with a Hilbert space dimension larger than required for the desired λ. However, the energy levels exceeding λd_S would have to be truncated before the preparation step to avoid unnecessary additional energy costs. The general form of the unitaries realising such unbiased measurement procedures is summarised below.

Lemma 2

Let $\mathcal{M}_U(\beta)$ be an unbiased finite-resource ($\Delta E < \infty, T = 1/\beta > 0$) measurement procedure with *unitary* correlating step $\mathcal{E}_{\text{II}}$ using a pointer system Hilbert space $\mathcal{H}_P$ and Hamiltonian H_P with $d_P = \lambda d_S$ for $\lambda \in \mathbb{N}$. The unitary map U realizing the correlating step, i.e.,

$$\mathcal{E}_{\text{II}}[\rho_S \otimes \rho_P] = U\rho_S \otimes \rho_P U^\dagger, \tag{A.34}$$

can be split into two consecutive unitary operations, $U = V\tilde{U}$, where $\tilde{U}$ and V are of the form

$$\tilde{U} = \sum_{i=0}^{d_S-1} |i\rangle\langle i|_S \otimes \tilde{U}^{(i)}, \tag{A.35}$$

$$V = \sum_{i,j=0}^{d_S-1} \sum_{m=1}^{\lambda} |j\rangle\langle i|_S \otimes |\tilde{\psi}_m^{(i)}\rangle\langle\tilde{\psi}_m^{(j)}|_P, \tag{A.36}$$

and $\tilde{U}^{(i)}$ are arbitrary unitaries on $\mathcal{H}_P$.

Proof of Lemma 2. The structure of the unitaries in the correlating step can be understood by noting that unitaries have only a single non-trivial Kraus operator. The operators $\tilde{U}^{(i)}$ in the first unitary $\tilde{U}$ then simply correspond to the single Kraus operators of the maps $\mathcal{E}_{\text{II}}^{(i)}$ from Eq. (A.27), rearranging the joint density matrix only in the subspaces of fixed $|i\rangle_S$, creating the distinction between the sub-blocks $\tilde{A}_{ji}$ for different j. The second part, realized by the unitary V then just swaps these sub-blocks, such that all $\tilde{A}_{0i}$ are left in the subspace corresponding to $|0\rangle_S$ and Π_i, all $\tilde{A}_{1i}$ are left in the subspace corresponding to $|1\rangle_S$

and Π_i, and so forth. The only freedom in choosing unitary correlation steps for unbiased measurements hence lies in the choice of the $\tilde{U}^{(i)}$. □

A.VI Unbiased and Non-invasive Measurements are Faithful

With the compact result of Lemmas 1 and 2 at hand, let us now briefly return to the relationship between the properties unbiasedness and non-invasiveness. As we have now seen, measurement procedures that are unbiased for all initial system states ρ_S are required to map the subspace of the joint system-pointer Hilbert space corresponding to the image of the operator $|i\rangle\langle i|_S \otimes \mathbb{1}_P$ to the subspace corresponding to the image of $\mathbb{1}_S \otimes \Pi_i$ for all i. At this point it becomes clear that, formulating the analogous statements to Lemmas 1 and 2 for measurement procedures that are non-invasive instead of unbiased for all ρ_S, results in maps from the subspace corresponding to the image of the operator $|i\rangle\langle i|_S \otimes \mathbb{1}_P$ to itself. Since the image of $|i\rangle\langle i|_S \otimes \mathbb{1}_P$ is spanned by the set of non-trivial joint eigenvectors of the set of projectors $\{|i\rangle\langle i|_S \otimes \Pi_j\}_j$, and the image of $\mathbb{1}_S \otimes \Pi_i$ is spanned by the set of non-trivial joint eigenvectors of the set of projectors $\{|j\rangle\langle j|_S \otimes \Pi_i\}_j$, a map that is supposed to satisfy both unbiasedness and non-invasiveness for all ρ_S must be a map from the image of $|i\rangle\langle i|_S \otimes \mathbb{1}_P$ to the span of the set of non-trivial eigenvectors of $|i\rangle\langle i|_S \otimes \Pi_i$, such that $\mathrm{tr}(|i\rangle\langle i|_S \otimes \Pi_i \tilde{\rho}_{SP}) = \rho_{ii}\ \forall i$. By construction, one thus has $\sum_i \mathrm{tr}(|i\rangle\langle i|_S \otimes \Pi_i \tilde{\rho}_{SP}) = \sum_i \rho_{ii} = 1$, and the measurement is faithful.

A.VII Maximally Correlating Unbiased Measurements

To gain further insight into the fundamental limitations of non-ideal measurements, we now wish to focus on a special case where Lemma 2 applies, that is, an unbiased measurement procedure with unitary correlating step, such that — at least for the purpose of controlling their interaction — the joint system of pointer and measured system can be considered closed. That is, procedures where $\mathcal{E}_{\mathrm{II}}[\rho_S \otimes \rho_P] = U \rho_S \otimes \rho_P U^\dagger$. Apart from this restriction, we will only consider preparation steps that modify the temperature of the initial pointer system, i.e. $\mathcal{E}_{\mathrm{I}}$ is a refrigeration step. In such a scenario, $T = 1/\beta$ might be the initial temperature of the pointer, or, e.g., one below the ambient temperature, reached by investing energy for cooling the pointer. For such measurement procedures, we now wish to find the maximum attainable correlation between the system and pointer. As we show, there is an algebraic maximum $C_{\max}$ for the correlations that can be unitarily created between the system and the thermal pointer, regardless of the initial system state ρ_S. Recall the definition of the correlation function in Eq. (A.19), which we rewrite as

$$C(\tilde{\rho}_{SP}) := \sum_i \mathrm{tr}\left[\tilde{\Pi}_{ii} \tilde{\rho}_{SP}\right], \tag{A.37}$$

by making the association $\tilde{\Pi}_{ij} = |i\rangle\langle i| \otimes \Pi_j\ \forall i, j$. By definition, the correlation function is only sensitive to terms appearing along the diagonal w.r.t. any chosen common eigenbasis (with nontrivial eigenvalues) of the set of operators $\tilde{\Pi}_{ii}$. We refer to the subspace of $\mathcal{H}_S \otimes \mathcal{H}_P$ spanned by these eigenvectors as $\mathcal{H}_{\mathrm{corr}}$, and to its complement as H_{nc}, such that $\mathcal{H}_S \otimes \mathcal{H}_P = \mathcal{H}_{\mathrm{corr}} \oplus \mathcal{H}_{\mathrm{nc}}$. In particular, this implies that the unitary transformation achieving the algebraic maximum (over all unitaries U_{unb} realizing unbiased measurement procedures in the sense of Lemma 2)

$$\max_{U_{\mathrm{unb}}} C(\tilde{\rho}_{SP}) = C_{\max} \tag{A.38}$$

is not unique since $C(\tilde{\rho}_{SP})$ is invariant under operations of the form $U_{\mathrm{corr}} \oplus U_{\mathrm{nc}}$, where U_{corr} and U_{nc} act nontrivially only on the subspaces $\mathcal{H}_{\mathrm{corr}}$ and $\mathcal{H}_{\mathrm{nc}}$, respectively.

Within the orbit of all unitaries that one may perform (including those corresponding to biased measurements) on $\rho_S \otimes \tau_P$, the global maximum value of the function $C(\tilde{\rho}_{SP})$ is achieved when the state $\tilde{\rho}_{SP}$ is block-diagonal w.r.t. to the subspace partition into $\mathcal{H}_{\mathrm{corr}} \oplus \mathcal{H}_{\mathrm{nc}}$, and the eigenvalues of the joint final state restricted to the d_P-dimensional correlated subspace $\mathcal{H}_{\mathrm{corr}}$, given by $\tilde{\rho}_{\mathrm{corr}} = \Pi_{\mathrm{corr}} \tilde{\rho}_{SP} \Pi_{\mathrm{corr}}$ with $\Pi_{\mathrm{corr}} = \sum_i |i\rangle\langle i| \otimes \Pi_i$, are the d_P largest eigenvalues of $\tilde{\rho}_{SP}$. These eigenvalues depend on ρ_S. However, we have to take into account unbiasedness and the fact that we are looking for a unitary. In particular, from Eq. (A.32) we know that the Π_i must all have the same rank, namely $\mathrm{rank}(\Pi_i) = d_P/d_S = 2^{N-1}\ \forall i$. It then becomes apparent that one is restricted to selecting the d_P/d_S largest eigenvalues of $\rho_P = \tau_P(\beta)$ for each of the d_S subspaces corresponding to the image of a projector $|i\rangle\langle i| \otimes \Pi_i$. Since the assignment of eigenvalues to each subspace labelled by i is the same, and the corresponding matrix elements of the initial system state sum to 1, the maximal achievable correlation is independent of ρ_S. In the notation of Eq. (A.1), this corresponds to all probability populations that belong to the sector where $(k = 0)$, i.e.,

$$C_{\max}(\beta) = \sum_{i=0}^{d_S-1} \sum_{j=0}^{d_P/d_S-1} \rho_{ii}\, p_j^{(0)} = \tfrac{1}{\mathcal{Z}} \sum_{i=0}^{d_P/d_S-1} e^{-\beta E_i^{(0)}}. \tag{A.39}$$

As mentioned in Eq. (A.29), this function is independent of the system. The remaining probability weights (i.e., the $p_j^{(k)}$ for $k \neq 0$ and $j = 0, \ldots, d_P/d_S - 1$) are distributed in the non-correlated subspace.

The corresponding general form of the joint final state $\tilde{\rho}_{SP}$ of any unitarily maximally correlating unbiased measurement procedure (starting from an initially thermal pointer state) can then be compactly

specified in terms of its correlation matrix $\Gamma_{U_{C_{\max}}}$ as defined in Eq. (A.26). To write $\Gamma_{U_{C_{\max}}}$ in a simple form, let $a_i^{(0)}$ for $i = 0, 1, \ldots, d_S$ be $d_P/d_S \times d_P/d_S$ Hermitian matrices whose eigenvalues are the d_P/d_S largest eigenvalues of the initial pointer state $\rho_P = \tau_P(\beta)$, i.e.,

$$a_i^{(0)} = M_i^{(0)} \big(\mathrm{diag}(p_0^{(0)}, p_1^{(0)}, \ldots, p_{d_P/d_S-1}^{(0)}\big) M_i^{(0)\dagger}, \tag{A.40}$$

where the $M_i^{(0)}$ are $d_P/d_S \times d_P/d_S$ unitary matrices. The correlation matrix $\Gamma_{U_{C_{\max}}}$ is then of the form

$$\Gamma_{U_{C_{\max}}} = \left[\begin{array}{c|c|c|c} \rho_{00} a_0^{(0)} & \rho_{11}\tilde{A}_{01} & \rho_{22}\tilde{A}_{02} & \cdots \\ \hline \rho_{00}\tilde{A}_{10} & \rho_{11} a_1^{(0)} & \rho_{22}\tilde{A}_{12} & \cdots \\ \hline \rho_{00}\tilde{A}_{20} & \rho_{11}\tilde{A}_{21} & \rho_{22} a_2^{(0)} & \cdots \\ \hline \vdots & \vdots & \vdots & \ddots \end{array}\right], \tag{A.41}$$

where the block matrices on the diagonal (blue) are the corresponding entries of $\tilde{\rho}_{SP}$ restricted to the correlated subspace $\mathcal{H}_{\mathrm{corr}}$, whereas the block matrices on the off-diagonal of $\Gamma_{U_{C_{\max}}}$ (here shown in green) correspond to the diagonal blocks of $\tilde{\rho}_{SP}$ restricted to the non-correlated subspace $\mathcal{H}_{\mathrm{nc}}$. Additional off-diagonal entries may appear in the projection of $\tilde{\rho}_{SP}$ onto $\mathcal{H}_{\mathrm{nc}}$ between blocks $\tilde{A}_{ij}$ and $\tilde{A}_{i'j}$ with the same column index j but different row indices $i \neq i'$ with $i, i' \neq j$, while maintaining an unbiased measurement with maximal correlation. The additional constraint of Eq. (A.25) ensuring an unbiased measurement procedure can here be written as $\mathrm{tr}[a_j^{(0)}] + \sum_{i, i\neq j} \mathrm{tr}[\tilde{A}_{ij}] = 1, \forall j$. The remaining freedom of applying unitaries that leave the subspaces $\mathcal{H}_{\mathrm{corr}}$ and $\mathcal{H}_{\mathrm{nc}}$ invariant and are compatible with unbiasedness can be used for minimization of the corresponding energy cost. Before we discuss this procedure for arbitrary system and pointer dimensions, it will be instructive to consider the special case where the system is a single qubit and the pointer consists of N identical two-level systems.

A.VIII Optimally Correlating Unitary for a Single-Qubit System and N-Qubit Pointer

In the previous appendix, we have identified the structure of all unitarily correlating unbiased measurements that create maximal correlations $C_{\max}$ (a subclass of the maps $\mathcal{E}_{\mathrm{II}}$). We are now interested in further restricting this set of measurements to identify those unitaries that achieve $C_{\max}$ for the least energy. That is, we wish to determine the optimal U_{opt} which solves the optimisation problem

$$\min_{U_{\mathrm{corr}}} \Delta E_{\mathrm{II}} \ \text{ s.t. } \ C(\tilde{\rho}_{SP}) = C_{\max}\,. \tag{A.42}$$

For arbitrary system dimensions and Hamiltonians, the explicit form of the solutions U_{opt} is rather involved and the proofs of optimality become very technical in nature. Before we move on to such general cases in Appendix A.IX, let us therefore here illustrate the general method by focusing on an example of interest.

Here, we consider a two-dimensional system, i.e., a qubit with Hilbert space $\mathcal{H}_S = \mathbb{C}^2$, dimensions $d_S = 2$, and a Hamiltonian H_S with eigenstates $|0\rangle_S$ and $|1\rangle_S$ and spectral decomposition $H_S = E_S\,|1\rangle\langle 1|_S$. In addition, we assume that the system state is initially unknown such that the corresponding density operator is maximally mixed, $\rho_S = \frac{1}{2}\mathbb{1}_2$. Meanwhile, we consider a measurement apparatus modelled as an N-qubit pointer, $d_P = 2^N$, where each qubit has the same local Hamiltonian with vanishing ground state energy and energy gap matching the system energy gap, $E_P = E_S$. The total pointer Hamiltonian is thus $H_P = \sum_{k=0}^{1}\sum_{i=0}^{2^{N-1}-1} E_i^{(k)}\,|E_i^{(k)}\rangle\langle E_i^{(k)}|$, where we have adopted the sector notation introduced in Eq. (A.1). Note that the pointer spectrum is highly degenerate since there are 2^N eigenvalues but only $N+1$ different energy levels, $E_i^{(k)}/E_S \in \{0, 1, \ldots, N\}$. With respect to the energy eigenbasis the initial pointer state before the correlating step is

$$\tau_P(\beta)^{\otimes N} = \sum_{k=0}^{1}\sum_{i=0}^{2^{N-1}-1} p_i^{(k)}\,|E_i^{(k)}\rangle\langle E_i^{(k)}|\,, \tag{A.43}$$

with $p_i^{(k)} = e^{-\beta E_i^{(k)}}/\mathcal{Z}$ and $\mathcal{Z} = \mathrm{tr}(e^{-\beta H_P}) = \sum_{i,k} e^{-\beta E_i^{(k)}}$. For this setting, we will now solve the optimization problem of Eq. (A.42) for $\tilde{\rho}_{SP} = U_{\mathrm{corr}}\,(\rho_S \otimes \tau_P(\beta)^{\otimes N})U_{\mathrm{corr}}^\dagger$.

From (A.39), the maximum algebraic correlation achievable between an $N-$qubit pointer and a qubit system is

$$C_{\max}(\beta) = \tfrac{1}{\mathcal{Z}} \sum_{i=0}^{2^{N-1}-1} e^{-\beta E_i^{(0)}}\,, \tag{A.44}$$

and the post-interaction correlation matrix associated with this scenario is given by

$$\Gamma_{U_{C_{\max}}} = \left[\begin{array}{c|c} \rho_{00} a_0^{(0)} & \rho_{11}\tilde{A}_{01} \\ \rho_{00}\tilde{A}_{10} & \rho_{11} a_1^{(0)} \end{array}\right]. \tag{A.45}$$

Here, $a_0^{(0)}$ and $a_1^{(0)}$ are $2^{N-1} \times 2^{N-1}$ dimensional Hermitian matrices whose eigenvalues are the 2^{N-1} largest eigenvalues of $\tau_P^{\otimes N}$, that is, there are a unitary matrices $M_0^{(0)}$ and $M_1^{(0)}$ such that

$$a_i^{(0)} = M_i^{(0)} \big(\mathrm{diag}(p_0^{(0)}, \cdots, p_{2^{N-1}-1}^{(0)})\big) M_i^{(0)\dagger} \quad \text{for } i = 0, 1. \tag{A.46}$$

In our example, we further have $\rho_{00} = \rho_{11} = \frac{1}{2}$ but we leave the symbols ρ_{00} and ρ_{11} for clarity where necessary. For the interaction to be unbiased according to Eq. (A.41), there is now no choice but to set $\tilde{A}_{01} = a_1^{(1)}$ and $\tilde{A}_{10} = a_0^{(1)}$, where

$$a_i^{(1)} = M_i^{(1)} \big(\mathrm{diag}(p_0^{(1)}, p_1^{(1)}, \ldots, p_{2^{N-1}-1}^{(1)}\big) M_i^{(1)\dagger}, \tag{A.47}$$

and $M_i^{(1)}$ for $i = 0, 1$ are unitaries not yet fixed by the requirements of unbiasedness or maximal correlation. The eigenvalues $\{p_0^{(1)}, p_1^{(1)}, \ldots, p_{2^{N-1}-1}^{(1)}\}$ of $a_0^{(1)}$ and $a_1^{(1)}$ correspond to the second (smaller) half of the eigenvalues of $\tau_P^{\otimes N}$, and hence we have $\mathrm{tr}(a_i^{(0)}) + \mathrm{tr}(a_i^{(1)}) = 1\ \forall i$. The correlation matrix becomes

$$\Gamma_{U_{C_{\max}}} = \left[\begin{array}{c|c} \rho_{00}a_0^{(0)} & \rho_{11}a_1^{(1)} \\ \rho_{00}a_0^{(1)} & \rho_{11}a_1^{(0)} \end{array}\right]. \tag{A.48}$$

In the present case where $d_S = 2$ and a maximally correlated, unbiased measurement, the correlation matrix $\Gamma_{U_{C_{\max}}}$ indeed catches all nonzero elements of $\tilde{\rho}_{SP}$, which is block diagonal,

$$\tilde{\rho}_{SP} = \mathrm{diag}(\rho_{00}a_0^{(0)}, \rho_{11}a_1^{(1)}, \rho_{00}a_0^{(1)}, \rho_{11}a_1^{(0)}). \tag{A.49}$$

In general, for $d_S > 2$, additional nonzero off-diagonal elements may appear in $\tilde{\rho}_{SP}$ that are not explicitly captured by $\Gamma_{U_{C_{\max}}}$.

The cost of correlating, and the function we wish to minimise, is given by the energy difference of the initial and final states,

$$\Delta E_{\Pi} = E(\tilde{\rho}_{SP}) - E(\rho_{SP}). \tag{A.50}$$

Since the initial energy is fixed by the initial temperature (and any preparation one wishes to include), we focus on minimising $E(\tilde{\rho}_{SP})$. In order to facilitate the computation, it will be useful to decompose $E(\tilde{\rho}_{SP})$ in terms of the correlated and non-correlated subspaces

$$\begin{aligned} E(\tilde{\rho}_{SP}) &= E_{\mathrm{corr}}(\tilde{\rho}_{SP}) + E_{\mathrm{nc}}(\tilde{\rho}_{SP}) \\ &= \mathrm{tr}[\sum_i \tilde{\Pi}_{ii} H_{SP} \tilde{\rho}_{SP}] + \mathrm{tr}[\sum_{\substack{i,j \\ i\neq j}} \tilde{\Pi}_{ij} H_{SP}\ \tilde{\rho}_{SP}], \end{aligned} \tag{A.51}$$

where, again we have $\tilde{\Pi}_{ij} = |i\rangle\langle i| \otimes \Pi_j$ and the combined system-pointer Hamiltonian is $H_{SP} = H_S \otimes \mathbb{1}_P + \mathbb{1}_S \otimes H_P$. For our qubit example, measured by an N-qubit pointer, this becomes

$$\begin{aligned} E(\tilde{\rho}_{SP}) = \mathrm{tr}[(\tilde{\Pi}_{00} + \tilde{\Pi}_{11}) H_{SP} \tilde{\rho}_{SP}] \\ + \mathrm{tr}[(\tilde{\Pi}_{01} + \tilde{\Pi}_{10}) H_{SP}\ \tilde{\rho}_{SP}]. \end{aligned} \tag{A.52}$$

The class of unitaries that achieve $C_{\max}$ rearranges the elements of ρ_{SP} to place the 'heaviest' populations of probabilities (eigenvalues of τ_P) into the correlated subspaces of $\tilde{\rho}_{SP}$. From this constraint we already know which elements (eigenvalues) of the post-interaction state $\tilde{\rho}_{SP}$ are assigned to which subspaces. In order to minimise the energy, one must therefore find the optimal assignment of the energy eigenbasis to these subspaces, which amounts to determining the Π_i.

We now proceed as follows: First, we will minimise the energy in the correlated subspaces, after which we will minimise the energy in the non-correlated subspaces, a strategy that presents a global energy minimum for the entire state.

Noting that $\tilde{\Pi}_{ij} \perp \tilde{\Pi}_{i'j'}$ whenever $i \neq i'$ or $j \neq j'$ we observe that also $(\tilde{\Pi}_{00} + \tilde{\Pi}_{11})$ is a projector. We can hence rewrite the first term on right-hand side of Eq. (A.52) as

$$\begin{aligned} \mathrm{tr}\big[\sum_{i=0,1} \tilde{\Pi}_{ii}\, H_{SP}\, \tilde{\rho}_{SP}\big] &= \mathrm{tr}\big[\big(\sum_{i=0,1} \tilde{\Pi}_{ii}\big)^2 H_{SP}\, \tilde{\rho}_{SP}\big] \\ &= \mathrm{tr}\big[\big(\sum_{i=0,1} \tilde{\Pi}_{ii}\big) H_{SP}\, \tilde{\rho}_{SP} \big(\sum_{j=0,1} \tilde{\Pi}_{jj}\big)\big] \\ &= \sum_{i=0,1} \mathrm{tr}\big[\tilde{\Pi}_{ii}\, H_{SP}\, \tilde{\rho}_{SP}\, \tilde{\Pi}_{ii}\big], \end{aligned} \tag{A.53}$$

where we have used the orthogonality of the projectors again in the last step. Once the trace has been restricted to the subspace corresponding to the space spanned by the nontrivial eigenvectors of $\tilde{\Pi}_{ii}$, we can further rewrite Eq. (A.53) as

$$\begin{aligned} &\sum_{i=0,1} \mathrm{tr}\big[\tilde{\Pi}_{ii}\, H_{SP}\, \tilde{\rho}_{SP}\, \tilde{\Pi}_{ii}\big] \\ &= \sum_{i=0,1} \mathrm{tr}\big[\tilde{\Pi}_{ii}\, (H_S + H_P)\, \tilde{\rho}_{SP}\, \tilde{\Pi}_{ii}\big] \\ &= \rho_{11}\, E_S\, \mathrm{tr}\big[a_1^{(0)}\big] + \sum_{i=0,1} \rho_{ii}\, \mathrm{tr}\big[\Pi_i H_P \Pi_i\, a_i^{(0)}\big]. \end{aligned} \tag{A.54}$$

Here, one should note that, strictly speaking, $\Pi_i H_P \Pi_i$ are $d_P \times d_P$ matrices, while $a_i^{(0)}$ are $d_P/d_S \times d_P/d_S$ matrices. However, the image of $\Pi_i H_P \Pi_i$ is also of dimension d_P/d_S, and one may hence think of $\Pi_i H_P \Pi_i$ as nonzero $d_P/d_S \times d_P/d_S$ matrices padded by rows and columns of zeros. In a slight abuse of notation we use the same symbol for the entire operator and its nontrivial block, since it is clear from the context, which object is referred to. In particular, $(\Pi_i H_P \Pi_i)\, a_i^{(0)}$ refers to the product of two $d_P/d_S \times d_P/d_S$ matrices. To simplify the last line of Eq. (A.54), let us first write $a^{(0)} := \mathrm{diag}(p_0^{(0)}, \cdots, p_{2^{N-1}-1}^{(0)})$, where we may assume w.l.o.g. that $a^{(0)}$ is diagonal w.r.t. the same basis as $\Pi_i H_P \Pi_i$ and the populations are ordered in non-increasing order. Any mismatch can be absorbed into the choice of the $M_i^{(0)}$. Further note for the first term that $\mathrm{tr}\big[a_1^{(0)}\big] = \mathrm{tr}\big[a^{(0)}\big]$, whereas the second term can be expressed as

$$\begin{aligned} \mathrm{tr}\big[\Pi_i H_P \Pi_i\, a_i^{(0)}\big] &= \mathrm{tr}\big[\Pi_i H_P \Pi_i\, M_i^{(0)} a^{(0)} M_i^{(0)\dagger}\big] \\ &= \mathrm{tr}\big[M_i^{(0)\dagger} \Pi_i H_P \Pi_i\, M_i^{(0)} a^{(0)}\big]. \end{aligned} \tag{A.55}$$

The quantity that we wish to minimize in this first step is thus of the form

$$\begin{aligned} E_{\mathrm{corr}}(\tilde{\rho}_{SP}) = \mathrm{tr}\big[\big(\rho_{00} M_0^{(0)\dagger} \Pi_0 H_P \Pi_0 M_0^{(0)} \\ + \rho_{11} M_1^{(0)\dagger} \Pi_1 (H_P + E_S) \Pi_1 M_1^{(0)}\big) a^{(0)}\big]. \end{aligned} \tag{A.56}$$

The minimization is to be carried out over all choices of projectors Π_i, i.e., choosing the basis $\{|\tilde{\psi}_n^{(i)}\rangle\}_{i,n}$ in

relation to the eigenbasis of H_{SP}, as well as over all choices of the $M_i^{(0)}$, or, in general the $M_i^{(j)}$. While the minimisation over the Π_i requires some more in-depth analysis (that we perform below), a simple observation can made right away. For a given initial state, the state in the unitary orbit of the initial state with minimal energy w.r.t. to a given Hamiltonian must be diagonal in the eigenbasis of this Hamiltonian, i.e., the corresponding passive state. Meanwhile, the conditions of unbiasedness and maximal algebraic correlations impose a certain block structure once a basis has been fixed and allow for but do not require off-diagonal elements. Therefore, it is clear that the unbiased, minimal energy solution with maximal algebraic correlations must be diagonal w.r.t. H_{SP}, restricting the unitaries connecting the bases $\{|\tilde{\psi}_n^{(i)}\rangle\}_{i,n}$ with the eigenbasis of H_{SP} as well as the unitaries $M_i^{(j)}$ to be *permutation* matrices. Moreover, suppose that for the fixed choice of $M_i^{(0)} = \mathbb{1}\ \forall i$ one has found an optimal choice of Π_i. Then any nontrivial modification of any of the $M_i^{(0)}$ can only increase the energy in the respective subspace. Without loss of generality we therefore set $M_i^{(0)} = \mathbb{1}\ \forall i$.

With this, we can now rewrite the energy in the correlated subspace as

$$E_{\text{corr}}(\tilde{\rho}_{SP}) = \text{tr}\big[\,\big(\rho_{00}\Pi_0 H_P \Pi_0 + \rho_{11}\Pi_1(H_P + E_S)\Pi_1\big)a^{(0)}\big] = \mathbf{x}\cdot\mathbf{a}^0, \tag{A.57}$$

where $\mathbf{a}^0$ is the vector of diagonal entries of the matrix $a^{(0)}$ and $\mathbf{x} \in \mathbb{R}^{2^{N-1}}$ is the vector of diagonal entries of the matrix $\big(\rho_{00}\Pi_0 H_P \Pi_0 + \rho_{11}\Pi_1(H_P + E_S)\Pi_1\big)$. Every component x_i of $\mathbf{x}$ is seen to be sum of two energies from the Π_0 and Π_1 subspaces of H_P, respectively modulated by the respective system populations ρ_{ii}.

We now switch to a slightly less cumbersome notation for the pointer. Let the set $\mathcal{S}_N$ with elements s_i for $i \in \{0, \cdots, d_P - 1\}$ be the set of energies (in units of E_S) in the energy spectrum of the pointer, ordered in non-decreasing order, such that $H_P = \sum_i s_i\,|s_i\rangle\langle s_i|$ and $s_i \leq s_j\ \forall i < j$. For example, a 3-qubit pointer with gap $E_P = E_S$ and vanishing ground state would be associated with the set $\mathcal{S}_3 = \{0, 1, 1, 1, 2, 2, 2, 3\}$. The elements of the vector $\mathbf{x}$ can now be written as

$$x_i = \rho_{00}s_j + \rho_{11}(s_l + E_S) \quad j \neq l, \quad 0 \leq j, l \leq (2^N - 1), \quad s_{j,l} \in \mathcal{S}_N\ , \tag{A.58}$$

such that the indices j, l are used only once. Thus, the x_i are composed by selecting pairs of elements, without replacement, from the set $\mathcal{S}_N$. There are several statements we can make immediately about the set $\mathcal{S}_N$. First, it has 2^N elements which are distributed binomially such that the energy kE_P appears $\binom{N}{k}$ times. Second, in the case that we are probing an unknown state, $\rho_S = \frac{1}{2}\mathbb{1}_2$, the sum of the elements of $\mathbf{x}$ is constant. Namely

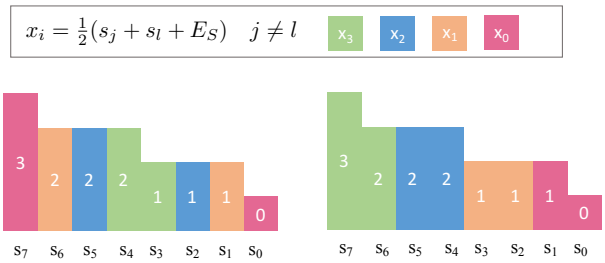

Figure A.1: The energies of a 3−qubit pointer $\mathcal{S}_3 = \{s_0, \cdots, s_7\}$ with gap $E_P = 1$ are arranged in non-increasing order. The schematic show two ways of choosing x_i from the set $\mathcal{S}_3$. The right hand side selects nearest neighbour pairs and thus represents the optimal pairing that minimises $E_{\text{corr}}(\tilde{\rho}_{SP})$ in (A.60).

$$\sum_{i=0}^{2^{N-1}-1} x_i = \sum_{i=0}^{2^N-1} (s_i + \tfrac{1}{4}E_S) = c\,. \tag{A.59}$$

Since $x_i \geq 0\ \forall i$, this means we can treat the set $\{x_i/c\}_i$ as a normalised probability distribution. Let X denote the set of all possible vectors $\mathbf{x}$, then, minimising the energy in the correlated subspace amounts to

$$\min E_{\text{corr}}(\tilde{\rho}_{SP}) = \min_{\mathbf{x}\in X}(\mathbf{x}\cdot\mathbf{a}^0) = \mathbf{x}^*\cdot\mathbf{a}^0. \tag{A.60}$$

The set X can be understood as the set of all possible ways of choosing pairs from $\mathcal{S}_N$ without replacement. The size (i.e., the cardinality) of X, denoted $|X|$, grows factorially with N, so searching by brute-force for the optimal vector is not feasible. The solution $\mathbf{x}^*$ for the minimization problem in Eq. (A.60) is given by the vector that pairs the smallest weights $p_i^{(0)}$ with the largest values x_i. Specifically, let $\mathbf{v}, \mathbf{w} \in \mathbb{R}^{2^{N-1}}$ be two normalised vectors with their components ordered in non-increasing order such that $v_0 \geq v_1 \cdots$ and $w_0 \geq w_1 \cdots$. We say that $\mathbf{v}$ majorises $\mathbf{w}$, written $\mathbf{v} \succ \mathbf{w}$, when $\sum_i^k v_i \geq \sum_i^k w_i\ \forall\, k$. In other words the cumulative sum of the components of the vector $\mathbf{v}$ grows faster than for $\mathbf{w}$. The vector $\mathbf{x}^*$ that presents the solution to the optimization problem is hence the vector that majorises all other vectors $\mathbf{x} \in X$, i.e.,

$$\mathbf{x}^* \succ \mathbf{x} \qquad \forall\, \mathbf{x} \in X. \tag{A.61}$$

This vector is constructed by maximising each x_i^* term by term from the bottom up, populating the components of $\mathbf{x}^*$ such that $x_{2^{N-1}-1} \geq \cdots \geq x_0$. This construction amounts to picking nearest neighbour pairs from the set $\mathcal{S}_N$, starting with the largest pair, as illustrated in Fig. A.1. Thus, the components of the optimal solution take the form

$$x_i^* = \rho_{00}s_{2i} + \rho_{11}(s_{2i+1} + E_S) \qquad s_i \in \mathcal{S}_N\,. \tag{A.62}$$

By construction, the majorisation of Eq. (A.61) is satisfied, and we have found the minimum energy solution in the correlated subspace.

By constructing $\mathbf{x}^*$ from nearest neighbour pairs in $\mathcal{S}_N$, we have fixed the energy eigenbasis in the pointer Hilbert space. The projectors on the pointer are then

$$\Pi_0 = \sum_{i=0}^{2^{N-1}-1} |\, s_{2i} \rangle\!\langle s_{2i} \,|\,, \quad \Pi_1 = \sum_{i=0}^{2^{N-1}-1} |\, s_{2i+1} \rangle\!\langle s_{2i+1} \,|\,. \tag{A.63}$$

We now proceed to minimise the energy in the non-correlated subspaces. Following a similar calculation and series of arguments as leading to Eq. (A.57), we write the energy as

$$\begin{aligned} E_{\mathrm{nc}}(\tilde{\rho}_{SP}) &= \mathrm{tr}\big[\, \rho_{11}\Pi_1 H_P \Pi_1\, M_1^{(1)} a^{(1)} M_1^{(1)\dagger} \\ &\quad + \rho_{00}\Pi_0(H_P + E_S)\Pi_0 M_0^{(1)} a^{(1)} M_0^{(1)\dagger}\big] \\ &= \mathbf{y}_1 \cdot \mathbf{a}_1^1 + \mathbf{y}_0 \cdot \mathbf{a}_0^1 \\ &= \mathbf{y}_1 \cdot M_1^{(1)}\mathbf{a}^1 + \mathbf{y}_0 \cdot M_0^{(1)}\mathbf{a}^1, \end{aligned} \tag{A.64}$$

where $\mathbf{a}_1^1$ and $\mathbf{a}_0^1$ are vectors whose components are the eigenvalues of the matrix $a^{(1)}$ in, as of yet, undetermined permutations (fixed by $M_1^{(1)}$ and $M_0^{(1)}$). The vector $\mathbf{a}^1$, in turn, collects exactly these eigenvalues in non-increasing order. However, since the energy basis for the pointer has been fixed by Eq. (A.63), the vectors $\mathbf{y}_1$ and $\mathbf{y}_0$ are completely determined. Their components are given by

$$(y_1)_i = \rho_{11} s_{2i+1}, \quad (y_0)_i = \rho_{00}(s_{2i} + E_S)\,. \tag{A.65}$$

To minimise the energy in the non-correlated subspace, we are thus looking for the solution to the optimisation problem

$$\min E_{\mathrm{nc}}(\tilde{\rho}_{SP}) = \min_{M_0^{(1)}, M_1^{(1)}} (\mathbf{y}_0 \cdot M_0^{(1)}\mathbf{a}^1 + \mathbf{y}_1 \cdot M_1^{(1)}\mathbf{a}^1)\,, \tag{A.66}$$

i.e., to find the optimal permutation matrices $M_0^{(1)}$ and $M_1^{(1)}$. Because of the freedom to choose these two permutations independently, the optimizations in the two subspaces decouple and it can be easily seen that the optimal solution for both is to pair up the smallest energies with the largest weights. In other words, to select $M_0^{(1)} = M_1^{(1)} = \mathbb{1}$. For $\rho_{00} = \rho_{11}$, this in turn implies

$$\min E_{\mathrm{nc}}(\tilde{\rho}_{SP}) = (\mathbf{y}_0 + \mathbf{y}_1) \cdot \mathbf{a}^1 = \mathbf{x}^* \mathbf{a}^1\,. \tag{A.67}$$

where we have noted that for the special case[6] of $d_S = 2$ one may collect $\mathbf{y}_0$ and $\mathbf{y}_1$ into $\mathbf{y}_0 + \mathbf{y}_1 = \mathbf{x}^*$.

$M^*\mathbf{a}^1$ must be ordered in non-increasing order to achieve the global minimum, which in turn implies that $M_* = \mathbb{1}$.

Collecting the results for the correlated and non-correlated subspaces and substituting for the forms of $\mathbf{a}^0$ and $\mathbf{a}^1$, the total energy after the interaction is

$$\begin{aligned} \min E(\tilde{\rho}_{SP}) &= \min(E_{\mathrm{corr}}(\tilde{\rho}_{SP}) + E_{\mathrm{nc}}(\tilde{\rho}_{SP})) \\ &= \mathbf{x}^* \cdot (\mathbf{a}^{\mathbf{0}} + \mathbf{a}^{\mathbf{1}}) \\ &= \tfrac{1}{2} \sum_{i=0}^{2^{N-1}-1} \big(s_{2i} + s_{2i+1} + E_S\big)(p_i^{(0)} + p_i^{(1)}). \end{aligned} \tag{A.68}$$

Since the initial state is diagonal w.r.t. the energy eigenbasis, the initial energy can also be easily computed to be

$$\begin{aligned} E(\rho_{SP}) = \tfrac{1}{2} \sum_{i=0}^{2^{N-1}-1} &\big[(2s_i + E_S)p_i^{(0)} \\ &+ (2s_{(2^{N-1}+i)} + E_S)p_i^{(1)}\big]. \end{aligned} \tag{A.69}$$

Thus from the above and Eq. (A.69) we have

$$\begin{aligned} \Delta E_{\Pi} &= E(\tilde{\rho}_{SP}) - E(\rho_{SP}) \\ &= \tfrac{1}{2} \sum_{i=0}^{2^{N-1}-1} (s_{2i} + s_{2i+1} - 2s_i)p_i^{(0)} \\ &\quad + (s_{2i} + s_{2i+1} - 2s_{(2^{N-1}+i)})p_i^{(1)}, \end{aligned} \tag{A.70}$$

where we note that, E_S (the gap of the system) no longer plays any role. Finally, observe that the cost of correlating is always finite. If one substitutes for the $p_i^{(j)}$ from Eq. (A.43) and takes the limit in which the pointer is in a pure state, i.e., $\beta \to \infty$, then the maximal correlation indeed is perfect correlation, $C = 1$, and the corresponding cost of correlating is given by

$$\lim_{\beta\to\infty} \Delta E_{\Pi} = \Delta E_{\Pi}^{(C=1)} = \tfrac{1}{2} E_P\,. \tag{A.71}$$

Notably, this expression is independent of N and hence true also when $N = 1$. Therefore, regardless of how many qubits the pointer consists of, if these qubits are initially in the ground state, the cost of correlating the system and pointer is precisely the cost of exciting only a single qubit (modulated by $\rho_{00} = \rho_{11} = \frac{1}{2}$).

These results can equivalently be expressed in terms of the sector notation introduced in Appendix A.I. In this case, the projectors in Eq. (A.63) become

$$\Pi_0 = \sum_{k=0}^{1} \sum_{i=0}^{2^{N-2}-1} |\, E_{2i}^{(k)} \rangle\!\langle E_{2i}^{(k)} \,|\,, \tag{A.72a}$$

$$\Pi_1 = \sum_{k=0}^{1} \sum_{i=0}^{2^{N-2}-1} |\, E_{2i+1}^{(k)} \rangle\!\langle E_{2i+1}^{(k)} \,|\,. \tag{A.72b}$$

Similarly, the energy after optimally correlating in Eq. (A.68) is

$$\begin{aligned} E(\tilde{\rho}_{SP}) = \tfrac{1}{2} \sum_{j=0}^{1} \sum_{i=0}^{2^{N-2}-1} &\big(E_{2i}^{(j)} + E_{2i+1}^{(j)} + E_S\big) \\ &\times (p_{i+j2^{N-2}}^{(0)} + p_{i+j2^{N-2}}^{(1)}), \end{aligned} \tag{A.73}$$

[6] This equality holds in the special case that $d_S = 2$, for the more general case see Appendix A.IX.

and the cost of correlating is

$$\Delta E_{\text{II}} = \tfrac{1}{2}\sum_{j=0}^{1}\sum_{i=0}^{2^{N-2}-1}\big[\big(E_{2i}^{(j)}+E_{2i+1}^{(j)}-2E_{i+j2^{N-2}}^{(0)}\big)p_{i+j2^{N-2}}^{(0)} + \big(E_{2i}^{(j)}+E_{2i+1}^{(j)}-2E_{i+j2^{N-2}}^{(1)}\big)p_{i+j2^{N-2}}^{(1)}\big]. \tag{A.74}$$

A.IX Construction of the optimal unitary for arbitrary systems

In the previous appendix we provided the construction for correlating a qubit system with an $N-$qubit pointer to $C_{\max}$. We also proved that this construction was an energy minimum. This construction generalises to any unknown quantum systems $\rho_S = \frac{1}{d_S}\mathbb{1}_{d_S}$ and thermal pointers $\tau_P(\beta)$, with arbitrary Hamiltonians H_S and H_P. Below we provide the recipe for constructing such a unitary.

Consider the thermal pointer $\tau_P(\beta)$ and order the spectrum of the pointer Hamiltonian in terms of its excitations into d_S sectors of size d_P/d_S, i.e., $H_P = \sum_{k=0}^{d_S-1}\sum_{i=0}^{d_P/d_S-1} E_i^{(k)}\,|E_i^{(k)}\rangle\langle E_i^{(k)}|$ with $E_i^{(k)} \leq E_j^{(k')}\ \forall i,j$ for $k' > k$. Diagonalise the pointer and the system in their ordered energy eigenbases,

$$\begin{aligned}\tau_P(\beta) &= \sum_{k=0}^{d_S-1}\sum_{i=0}^{d_P/d_S-1} p_i^{(k)}\,|E_i^{(k)}\rangle\langle E_i^{(k)}|\,,\\ \rho_S &= \sum_i \rho_{ii}\,|i\rangle\langle i|\,,\end{aligned} \tag{A.75}$$

where $p_i^{(k)} = 1/\mathcal{Z}e^{-\beta E_i^{(k)}}$.

Assign the largest d_P/d_S eigenvalues of the pointer state $\tau_P(\beta)$ (captured in the matrix $a^{(0)}$) to the correlated subspace. The form of the correlation matrix is given in Eq. (A.41). To minimise the energy contribution from the correlated subspace, given by $E_{\text{corr}}(\tilde{\rho}_{SP}) = \text{tr}[\sum_i \tilde{\Pi}_{ii}H_{SP}\tilde{\rho}_{SP}]$, choose the pointer Hilbert space projectors to be

$$\Pi_i = \sum_{k=0}^{d_S-1}\sum_{i=0}^{(d_P/d_S^2)-1} |E_{d_S\cdot j+i}^{(k)}\rangle\langle E_{d_S\cdot j+i}^{(k)}| \qquad \forall\, j\in\{0,\cdots,d_S-1\}\,. \tag{A.76}$$

This choice fixes the basis vectors for the pointer and thus it remains to distribute the remaining probability weights (the remaining eigenvalues of τ_P) in the non-correlated subspace. This is achieved by pairing the largest weights with the smallest energies. The remaining weights are

$$a^{(i)} = \big(\text{diag}(p_0^{(0)},\cdots,p_{2^{N-1}-1}^{(0)})\big) \quad i\in\{1,\cdots,d_S-1\}, \tag{A.77}$$

and the resulting correlation matrix, arising from the optimal unitary U_{opt} has the form

$$\Gamma_{U_{\text{opt}}} = \left[\begin{array}{c|c|c|c} \rho_{00}a^{(0)} & \rho_{11}a^{(1)} & \cdots & \rho_{d_S-1\,d_S-1}a^{(1)} \\ \hline \rho_{00}a^{(1)} & \rho_{11}a^{(0)} & \cdots & \rho_{d_S-1\,d_S-1}a^{(2)} \\ \hline \vdots & \vdots & \ddots & \vdots \\ \hline \rho_{00}a^{(d_S-1)} & \rho_{11}a^{(d_S-1)} & \cdots & \rho_{d_S-1\,d_S-1}a^{(0)} \end{array}\right] \tag{A.78}$$

In turn, this fixes the the matrices $\tilde{A}_{ij}$ in Eq. (A.41) to be

$$\tilde{A}_{ij} = a^{(\pi[i,j])} \tag{A.79}$$

where $\pi[m,k]$ denotes the $m,k-$th element of the $d_S\times d_S$ matrix composed of permutations of the entries of the set $\{0,1,\ldots,d_S-1\}$ under the constraint that the diagonal entries $\pi[m,m] = 0\ ,\forall m$. For the optimal energy solution, this permutation matrix takes the form

$$\pi = \begin{pmatrix} 0 & 1 & 1 & 1 & \ldots \\ 1 & 0 & 2 & 2 & \ldots \\ 2 & 2 & 0 & 3 & \ldots \\ 3 & 3 & 3 & 0 & \ldots \\ \vdots & \vdots & \vdots & \vdots & \ddots \end{pmatrix}, \tag{A.80}$$

which encodes how the correlations in the matrices $a^{(i)}$ are paired with the non-correlated subspaces. The final state admits a simplified form, namely

$$\tilde{\rho}_{SP} = \sum_{k=0}^{d_S-1}\frac{\rho_{kk}}{\mathcal{Z}}\Big(\sum_{i=0}^{d_P/d_S-1} e^{-\beta E_i^{(0)}}\,|k\rangle\langle k|\otimes|E_k^{(i)}\rangle\langle E_k^{(i)}| + \sum_{m\neq k}^{d_S-1}\sum_{i=0}^{d_P/d_S-1} e^{-\beta E_i^{(\pi[m,\,k])}}\,|m\rangle\langle m|\otimes|E_k^{(i)}\rangle\langle E_k^{(i)}|\Big). \tag{A.81}$$

It can be seen that this state is not unique due to the inherent description in terms of energy. Thus the final state $\tilde{\rho}_{SP}$ depends on one's choice of how to represent the basis and excitations and in general is degenerate. We leave it as an open investigation as to whether, within this class there is a preferred state with special and interesting properties.

II.7 Work Estimation and Work Fluctuations in the Presence of Non-Ideal Measurements

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Overview: In this publication, we investigated the influence of non-ideal measurements on the estimation of work in non-equilibrium processes in the context of the two-point measurement (TPM) scheme. As we show, when estimates of the work distribution are obtained at finite work cost, the resulting estimates are subject to imperfections, and more accurate estimates incur higher work costs. We further show that Jarzynski's equality can nevertheless be maintained exactly at the expense of a correction that depends only on the system's energy scale, but the more general fluctuation relation due to Crooks no longer holds when the cost of the work estimation procedure is finite.

Contribution: I helped to develop the ideas for this project, discussed them with my coauthors and supervised the project. I wrote the main text of the manuscript.

Work estimation and work fluctuations in the presence of non-ideal measurements

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From the perspective of quantum thermodynamics, realisable measurements cost work and result in measurement devices that are not perfectly correlated with the measured systems. We investigate the consequences for the estimation of work in non-equilibrium processes and for the fundamental structure of the work fluctuations when one assumes that the measurements are non-ideal. We show that obtaining work estimates and their statistical moments at finite work cost implies an imperfection of the estimates themselves: more accurate estimates incur higher costs. Our results provide a qualitative relation between the cost of obtaining information about work and the trustworthiness of this information. Moreover, we show that Jarzynski's equality can be maintained exactly at the expense of a correction that depends only on the system's energy scale, while the more general fluctuation relation due to Crooks no longer holds when the cost of the work estimation procedure is finite. We show that precise links between dissipation and irreversibility can be extended to the non-ideal situation.

I. INTRODUCTION

Energy is a resource and, as with any resource, it is of interest to understand how much of it is spent or can be obtained during a given process, or simply, how much of it is stored, for instance, in a battery. A quite different, but familiar, resource that one handles on a daily basis is money. Money does not usually come for free: it is exchanged for goods and services, and as a consequence it is in one's interest to know how much things cost and how much money is at hand, e.g., stored in a wallet or bank account. But while checking the exact amount of money (or lack thereof) in one's wallet is free, it is not unusual to expect that banks charge certain fees for storing and transferring money. Unfortunately, when it comes to energy, Nature is similarly unforthcoming. Fees apply to the storage and transfer of energy and an energy cost is incurred for obtaining estimates of the work transferred within any thermodynamic process, or stored in a quantum system. In this work, we show that obtaining these estimates with a finite amount of work implies an imperfection in the estimates themselves: estimates which are more accurate incur higher costs.

From a thermodynamic point of view, acknowledging the energetic cost of measurements is crucial, e.g., for a complete understanding of Maxwell's demon or Szilard's engine [1, 2]. The work-cost of those measurements that are ideal and projective has been investigated by means of the work-value of measurement outcomes [3–5] or via Landauer's erasure bound for resetting the memory that stores these outcomes [6–10]. However, a common observation among Refs. [11–13] is that the benefits derived from using measurements as sources of free energy are either matched or surpassed by the corresponding costs.

The crux of our argument is that energy delivered by measurements is not free of charge and must be supplied to realise the measurement in the first place. As we show, this statement is bolstered by the first, second and, in particular, third law of thermodynamics. It was recently shown in [14] that ideal projective measurements require one to prepare the measurement apparatus in a pure initial state. The third law stipulates that such zero-entropy states can only be prepared asymptotically using infinite time, infinite energy, or operations of infinite complexity (see e.g., [15]). Consequently, ideal measurements do not exist, in a strict sense, since they always incur diverging costs. This implies that any realistic measurement using finite resources is non-ideal. It is precisely these considerations that become conceptually important when the purpose of the measurement is to assess the energy consumption itself.

Significant focus in quantum statistical mechanics has been dedicated to the quantification of work and its fluctuations in thermodynamic processes [16–19]. Studies have also looked at the two-point measurement (TPM) scheme (one of the most prominent approaches for estimating work in an out-of-equilibrium process) [20] in the context of Jarzynski's and Crooks' fluctuation relations [21]. In this work we revisit these concepts and investigate the consequences for these quantities when one does *not* assume ideal measurements. We explicitly show how the average work of the ideal TPM is modified and discuss the operational meaning of the corresponding estimates. We show that while Jarzynski's equality can be maintained exactly at the expense of a correction that only depends on the system's Hamiltonian, the more general relation due to Crooks (as well as related results linking irreversibility and dissipation [22, 23]) no longer

hold in the presence of non-ideal measurements. Our results provide a qualitative connection between the cost of obtaining information about work and the trustworthiness of this information.

The paper is structured as follows. In Sec. II we set the stage for our investigation: First, we review the usual TPM scheme in Sec. II.1 before discussing the key properties of ideal and non-ideal measurements following Ref. [14] in Secs. II.2 and II.3, respectively. In Sec. III we discuss a modified TPM scheme based on non-ideal measurements and the resulting work estimates, before investigating the implications for fluctuation relations of Jarzynski and Crooks in Sec. IV. Finally, we discuss our findings in Sec. V.

II. FRAMEWORK

II.1. Two-point measurement scheme

To formulate our ideas we adopt a commonplace view in quantum thermodynamics, namely, that work is a central resource that is required to move systems away from freely available thermal equilibrium states [24] — an approach that has staged a diverse range of investigations within the broader field [25–27]. In this paradigm, previous research has investigated the work-cost (or gain) of quantum processes [28–32], refrigeration [33, 34], or for establishing correlations [35–38].

In this so called 'resource-theoretic' approach, consider a quantum system with Hamiltonian $H^{(0)} = \sum_i E_i^{(0)} |E_i^{(0)}\rangle\langle E_i^{(0)}|$ initially at thermal equilibrium with its environment at temperature T, described by a Gibbs state $\tau^{(0)} = \exp(-\beta H^{(0)})/\mathcal{Z}^{(0)}$ with partition function $\mathcal{Z}^{(0)} = \mathrm{Tr}\big(\exp(-\beta H^{(0)})\big)$ and $\beta = (k_{\mathrm{B}}T)^{-1}$. Suppose the system is driven out of equilibrium by a process Λ

$$(\tau^{(0)}, H^{(0)}) \xrightarrow{\Lambda} (\rho^{(\mathrm{f})}, H^{(\mathrm{f})}), \tag{1}$$

resulting in a final Hamiltonian $H^{(\mathrm{f})} = \sum_i E_i^{(\mathrm{f})} |E_i^{(\mathrm{f})}\rangle\langle E_i^{(\mathrm{f})}|$ and a final state $\rho^{(\mathrm{f})} = U_\Lambda \tau^{(0)} U_\Lambda^\dagger$, where U_Λ is a unitary determined by Λ. The work that is performed on or extracted from the system during such a process can be estimated via the two-point measurement (TPM) scheme [20, 21] consisting of two *ideal projective measurements* with respect to the eigenbases of $H^{(0)}$ and $H^{(\mathrm{f})}$ before and after the protocol Λ is implemented, respectively. After obtaining the outcomes labelled by "n" and "m" in these measurements one concludes that the system is left in the states $|E_n^{(0)}\rangle$ and $|E_m^{(\mathrm{f})}\rangle$, respectively. To any transition between these pure states one may associate a probability $p_{n\to m} = |\langle E_m^{(\mathrm{f})}|U_\Lambda|E_n^{(0)}\rangle|^2$ together with a work value $W_{n\to m} = E_m^{(\mathrm{f})} - E_n^{(0)}$, while the probability for obtaining the first outcome is $p_n^{(0)} = \exp(-\beta E_n^{(0)})/\mathcal{Z}^{(0)}$. The average work performed during the protocol is thus

$$\langle W\rangle_\Lambda = \sum_{m,n} p_n^{(0)} p_{n\to m} \big(E_m^{(\mathrm{f})} - E_n^{(0)}\big), \tag{2}$$

which equals the change in average energy during the protocol Λ, i.e.,

$$\langle W\rangle_\Lambda = \mathrm{Tr}(H^{(\mathrm{f})}\rho^{(\mathrm{f})}) - \mathrm{Tr}(H^{(0)}\tau^{(0)}) =: \Delta E_\Lambda. \tag{3}$$

Estimates of $\langle W\rangle_\Lambda$ could thus be obtained from performing ideal measurements and collecting the corresponding outcome statistics. In the following, we will show how the quantity $\langle W\rangle_\Lambda$ in Eq. (2) and its estimate are modified when replacing the two ideal measurements in the TPM by more general non-ideal measurements (see Fig. 1).

II.2. Ideal measurements

The notion of perfect projective measurements that leave the system in pure states with certainty is of course idealized. To understand this idealization and its consequences, we review the framework for non-ideal measurements in Ref. [14]: Measurements are performed by coupling the measured system to a suitably prepared measurement apparatus (the "pointer") via an energy investment. Assuming that the initial system and pointer states are ρ_S and ρ_P, respectively, the measurement can be described by a physical process that correlates the system and pointer, resulting in a joint post-measurement state $\tilde{\rho}_{SP}$. For each of the states $|n\rangle_S$ (with $n = 0, 1, \ldots, d_S - 1$) in the measurement basis $\{|n\rangle_S\}_n$, one assigns a corresponding outcome subspace of the pointer Hilbert space via a projector Π_n, such that $\sum_n \Pi_n = \mathbb{1}_P$ and $\Pi_m \Pi_n = \delta_{mn}\Pi_n$. We then define an ideal measurement to have the following three properties:

(i) **Unbiased**: The (post-interaction) pointer reproduces the measurement statistics of the (pre-interaction) system exactly, i.e.,

$$\mathrm{Tr}\big(\mathbb{1}_S \otimes \Pi_n \tilde{\rho}_{SP}\big) = \mathrm{Tr}\big(|n\rangle\langle n|_S\, \rho_S\big) \;\; \forall\, n \;\; \forall\, \rho_S\,. \tag{4}$$

(ii) **Faithful**: The post-interaction pointer and the post-interaction system are perfectly correlated w.r.t. the measurement basis (projectors), that is,

$$C(\tilde{\rho}_{SP}) := \sum_n \mathrm{Tr}\big(|n\rangle\langle n|_S \otimes \Pi_n\, \tilde{\rho}_{SP}\big) = 1\,. \tag{5}$$

In other words, given a measurement outcome n, the probability that the system is left in the state $|n\rangle_S$ is 1.

(iii) **Non-invasive**: The diagonal entries (w.r.t. the measurement basis) of the pre-measurement system state and the unconditional post-measurement system state are the same, i.e.,

$$\mathrm{Tr}\big(|n\rangle\langle n|_S\, \rho_S\big) = \mathrm{Tr}\big(|n\rangle\langle n|_S\, \tilde{\rho}_S\big) \quad \forall\, n\; \forall\, \rho_S\,, \tag{6}$$

where $\tilde{\rho}_S := \mathrm{Tr}_P(\tilde{\rho}_{SP})$.

A measurement that *does not* satisfy all three properties is called *non-ideal*. In particular, it was shown in Ref. [14] that ideal projective measurements are not exactly realizable in practise because they require the preparation of initially pure pointer states (at least in some nontrivial subspace) to satisfy condition (ii). However, the third law of thermodynamics prevents one from reaching the ground-state of any system with finite resources [15, 33, 34, 39–42]. Since any other (pure) state necessarily has higher energy than the ground state, the third law thus excludes ideal measurements.

II.3. Non-ideal measurements

In order to understand non-ideal measurements better, we consider how the laws of thermodynamics place constraints on the ability to perform measurements in quantum mechanics. To begin, we note that the first law establishes a lower bound on the work-cost of measurement when one explicitly considers the system and measurement apparatus in the physical description (note that work-costs in terms of lower bounds are sometimes assumed, even if the costs are not always explicitly considered [43–45]). The second law implies that any reduction of the system's entropy (e.g., by a projective measurement that leaves the system in a pure state) must be compensated by an entropy increase of at least the same magnitude in the environment or the measurement apparatus [11]. Finally, the third law provides the most severe constraint on the cost of measurement. From the above [14] an ideal measurement can only be implemented using a pure state measurement apparatus. By the third law, it is impossible to create pure states using finite resources (e.g., with finite time, energy or complexity), which implies that any physical measurement using finite resources is non-ideal. Nevertheless, it was shown that non-ideal measurements employing finite resources can approximate ideal projective measurements arbitrarily well.

To understand the sense in which a non-ideal measurement can be considered 'close' to ideal, we recall properties (i)-(iii). All three properties are independent – for a given measurement any one of them can be satisfied, while the other two are not [14, Appendix A.3]. At the same time, satisfying any two properties (for all ρ_S) also implies the third. Since the third law of thermodynamics prevents measurements from being exactly faithful (i.e., satisfying (ii) exactly), this means that any non-ideal measurement can only be unbiased or non-invasive, but not both.

Consider a non-ideal measurement that is non-invasive. From the above we know it can be neither faithful nor unbiased. Then neither the individual measurement outcomes nor the statistics generated from many measurements allow reliable inferences about either the post- or pre-measurement system state, respectively. Such a measurement does not seem to reveal any information about the measured system. Instead we consider non-ideal measurements that are *unbiased*, and take this to be the relevant requirement to speak meaningfully about a measurement. For unbiased measurements one may then attempt to maximise the correlation between pointer outcomes and post-measurement system states to approach an ideal measurement.

To formalise this, we consider an arbitrary system state ρ_S in finite dimension d_S, measured in the energy eigenbasis $\{|E_i\rangle_S\}_{i=0}^{d_S-1}$. The system interacts with the measurement apparatus, represented as a finite-size pointer with Hamiltonian $H_P = \sum_i E_i^{(P)} |E_i^{(P)}\rangle\langle E_i^{(P)}|$ and dimension d_P. In order to account transparently for all resources from a thermodynamic point of view, we assume the pointer is initially in a thermal[1] state $\tau_P = \exp(-\beta_P H_P)/\mathcal{Z}_P$. The interaction can be modelled by a suitable unitary[2] evolution U_{meas} leading to

$$\tilde{\rho}_{SP} := U_{\text{meas}}(\rho_S \otimes \tau_P) U_{\text{meas}}^\dagger. \tag{7}$$

The interaction between system and pointer is unitary but does not generally preserve energy, requiring an investment of energy

$$\Delta E_{\text{meas}} = \text{Tr}\big[(H_S + H_P)(\tilde{\rho}_{SP} - \rho_S \otimes \tau_P)\big] \tag{8}$$

in the form of work. For details see Appendix A.1. Following Ref. [14, Lemma 2], one may then construct U_{meas} to realise an unbiased measurement with finite energy cost, as we illustrate for a 3-dimensional system in Appendix A.1.a. The probability p_n to obtain the measurement outcome n in such a measurement is given by $p_n = \text{Tr}\big(|n\rangle\langle n|\rho_S\big)$. To ensure this is the case for all ρ_S, the unitary U_{meas} must result in a final state $\tilde{\rho}_{SP}$ which satisfies the equivalence relation

$$\mathbb{1}_S \otimes \Pi_n \tilde{\rho}_{SP} \mathbb{1}_S \otimes \Pi_n \mathrel{\hat{=}} p_n \tilde{U}^{(n)} \tau_P \tilde{U}^{(n)\dagger}, \tag{9}$$

i.e., $\mathbb{1}_S \otimes \Pi_n \tilde{\rho}_{SP} \mathbb{1}_S \otimes \Pi_n$ are $d_P \times d_P$ matrices with the same spectra as $p_n \tau_P$, i.e., they are equivalent up to applications of arbitrary unitaries $\tilde{U}^{(n)}$ on the pointer Hilbert space, as we discuss in detail in Appendix A.1.a. This implies that ρ_n, the conditional post-measurement system state for outcome n, is independent of the initial system state ρ_S. Note that this is also the case for ideal measurements (which are unbiased by definition), where

[1] If the system is assumed to be initially thermal as well, such as in the TPM scheme, there need not be a relation between the inverse temperatures of the pointer and the system in principle.

[2] In principle, one may effectively model such processes by completely positive and trace preserving (CPTP) maps, see [14, Appendix A.4], but for an exact account of the invested work it is necessary to specify a corresponding dilation to a unitary acting on a larger space of the pointer and its environment.

the pure state after the measurement only depends on the measurement outcome, but not on the initial system state. In this regard, the difference with unbiased non-ideal measurements is that the conditional state for the latter are not pure. The most general form of any post-measurement mixed state conditioned on the outcome n is

$$\rho_n = \sum_{l,l'} q_{ll'|n} \, |E_l\rangle\langle E_{l'}| \,. \tag{10}$$

Each state is normalised $\sum_l q_{ll|n} = 1, \ \forall\, n$ and in general the coefficients $q_{ll'|n}$ are independent of ρ_S. In the rest of the paper we formulate our results as conditions and constraints on $q_{ll'|n}$.

One should also bear in mind that for non-ideal measurements the pointer outcomes and post-measurement system states are not perfectly correlated. This can be quantified by the correlation function in Eq. (5),

$$C(\tilde{\rho}_{SP}) = \sum_{i=0}^{d_S-1} \mathrm{Tr}\big(|E_i\rangle\langle E_i|_S \otimes \Pi_i \, \tilde{\rho}_{SP}\big), \tag{11}$$

where Π_i are the orthogonal pointer projectors associated to different outcomes. The value of $C(\tilde{\rho}_{SP})$ represents the average probability of correctly inferring the post-measurement state upon observing the pointer. For ideal measurements $C(\tilde{\rho}_{SP}) = 1$. However, for non-ideal measurements there is an algebraic maximum, the *maximal correlation* $C_{\max} < 1$, which can be unitarily achieved. $C_{\max}$ is given by the sum of the largest $\frac{d_P}{d_S}$ eigenvalues of τ_P, see [14, Appendix A.7]. We call unbiased measurements that achieve $C(\tilde{\rho}_{SP}) = C_{\max}$ *unbiased maximally correlating* (UMC) measurements, discussed in detail in Appendix A.1.b. In the same Appendix we also show that UMC measurements lead to the following constraints on the coefficients of the post-measurement state: $q_{nn|n} = C_{\max}, \forall\, n$ and $q_{nl|n} = q_{ln|n} = 0 \ , \forall l \neq n$. As a consequence, the trace distance between the conditional post-measurement system state ρ_n and the pure state $|E_n\rangle$ evaluates to $D(\rho_n, |E_n\rangle\langle E_n|) = 1 - C_{\max}$.

By further restricting to UMC measurements of *minimal energy* (Appendix A.1.c) we obtain $q_{ll'|n} = 0 \ \forall E_l \neq E_{l'}$, because any off-diagonal elements with respect to energy eigenstates with different energies would imply extractable work, see, e.g., [46]. Minimal energy UMC measurements thus imply a back-action on the measured system. Up to off-diagonal elements in degenerate subspaces, the unconditional post-measurement state for minimal energy UMC measurements is given by $\tilde{\rho}_S = \sum_n p_n \rho_n = \sum_{n,l} q_{ll|n} p_n \, |E_l\rangle\langle E_l|$. By (6) this would be non-invasive when $\rho_S = \tilde{\rho}_S$ implying $q_{ll|n} = \delta_{ln} \ \forall n$.

At the same time, $C_{\max}$ can be understood as an indicator of the resource cost of a measurement: Qualitatively, increasing $C_{\max}$ requires more work, control over more complex pointers, more time to carry out operations, or combinations thereof [14]. In particular, for fixed pointer systems, increasing $C_{\max}$ can only be achieved by investing work in the preparation of the initial state of the pointer.

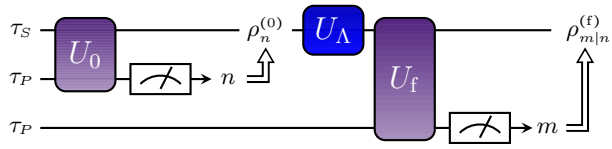

Figure 1. Work estimation using two non-ideal measurements. To estimate the work done on or extracted from a system during a process Λ, two measurements are carried out before and after the process occurs. The respective outcomes labelled "n" and "m" allow concluding that the system is left in states $\rho_n^{(0)}$ and $\rho_{m|n}^{(\mathrm{f})}$. In the TPM scheme [20], these states are (pure) eigenstates of the system Hamiltonian. For non-ideal measurements modelled by unitaries U_0 and U_f coupling the system state to pointers originally in thermal states τ_S, $\rho_n^{(0)}$ and $\rho_{m|n}^{(\mathrm{f})}$ are mixed states.

III. ESTIMATING WORK WITH NON-IDEAL MEASUREMENTS

It is hardly surprising that the procedure of checking how much work is spent or extracted during a protocol itself costs work. In other words, obtaining a work estimate is accompanied by an additional, non-negative work-cost. In Eq. (8) we saw that the specific (average) work ΔE_{meas} of UMC measurements depends on the details of the Hamiltonians, the initial state ρ_S, the temperature, and the association of the states $|E_i\rangle_S$ with the projectors Π_i. It has a finite positive minimum value $\Delta E_{\mathrm{meas}}^{\min} > 0$ and as pointed out in Ref. [14] should not be taken for granted, as it may significantly outweigh the ideal expectation $\langle W\rangle_\Lambda$ in Eq. (2). Particular attention should be paid when it comes to machines using measurements as a means of injecting free energy into the system [43–45], as such costs need to be included in an evaluation of the machine's efficiency.

In this work, we do not wish to focus on the specific cost of the measurement, but rather on the consequences for the work estimate itself. The imperfection of measurements is unavoidable and has immediate and interesting consequences for work estimation. To investigate, we modify the ordinary TPM scheme and replace the ideal measurements by non-ideal (minimal energy UMC) measurements[3] (Fig. 1). This assumption can be interpreted as the desire to restrict to measurements that are as close as possible to ideal ones while choosing the energetically cheapest way of doing so. A guiding intuition

[3] In principle these two measurements can be different but we assume that they are both minimal energy UMC.

for the following analysis is that work estimation requires two measurements. If the first is non-ideal but unbiased, it necessarily disturbs the system. In particular, the invasiveness of the first measurement changes the statistics of the second, leading to deviations in the work estimation from the ideal case.

Because the measurements are assumed to be unbiased, the probability $p_n^{(0)}$ for obtaining the outcome n in the first measurement is unchanged w.r.t. to the ideal scenario. However, since the system is disturbed by the measurement-induced back action, the conditional post-measurement state $\rho_n^{(0)}$ is no longer an eigenstate $|E_n^{(0)}\rangle$ of $H^{(0)}$. In particular, the conditional probability that the system is left in the eigenstate $|E_l^{(0)}\rangle$ after observing the pointer outcome n in the first measurement is $q_{ll|n}^{(0)} \neq \delta_{ln}$. Given the outcome n, the process Λ, thus acts on the state $\rho_n^{(0)} = \sum_l q_{ll|n}^{(0)} |E_l^{(0)}\rangle\langle E_l^{(0)}|$ via a unitary U_Λ. Unbiasedness then implies that the conditional probability to obtain outcome m in the second measurement given outcome n in the first measurement is

$$\begin{aligned} p(m|n) &= \langle E_m^{(f)} | U_\Lambda \rho_n^{(0)} U_\Lambda^\dagger | E_m^{(f)} \rangle \\ &= \sum_l q_{ll|n}^{(0)} p_{l\to m} \neq p_{n\to m}, \end{aligned} \tag{12}$$

where $p_{n\to m} = |\langle E_m^{(f)} | U_\Lambda | E_n^{(0)} \rangle|^2$ as before (for details of this scheme see Appendix A.2). For the work estimate $\langle W\rangle_{\text{non-id}}$ obtained within the non-ideal TPM scheme by (erroneously) associating outcomes n and m with energies $E_n^{(0)}$ and $E_m^{(f)}$ one obtains

$$\langle W\rangle_{\text{non-id}} = \sum_{m,n} p(m|n)\, p_n^{(0)} \left(E_m^{(f)} - E_n^{(0)}\right) \neq \langle W\rangle_\Lambda, \tag{13}$$

which generally does not match the ideal value in Eq. (2). In Appendix A.3.a we explicitly derive the corrected expression to be

$$\langle W\rangle_{\text{non-id}} = C_{\max}\langle W\rangle_\Lambda + \sum_{\substack{m,n \\ l\neq n}} p_{l\to m}\, q_{ll|n}^{(0)}\, p_n^{(0)} \left(E_m^{(f)} - E_n^{(0)}\right). \tag{14}$$

Thus, we find that between the modified and ordinary TPM schemes, the deviation of the estimated work $\langle W\rangle_{\text{non-id}}$ from the ideal value $\langle W\rangle_\Lambda$[4] is characterised by two values: a modifying prefactor $C_{\max} < 1$, along with an additional term which may be either positive or negative. Despite the generally complicated dependence on the details of the initial state, the measurement, energy spectrum and on the process Λ, we show, in Appendix A.3.b, that this deviation can be bounded,

$$|\langle W\rangle_{\text{non-id}} - \langle W\rangle_\Lambda| \leq (1 - C_{\max})\, \|H^{(f)}\|_\infty\,, \tag{15}$$

[4] obtained in the scenario with minimal energy UMC measurements using finite resources.

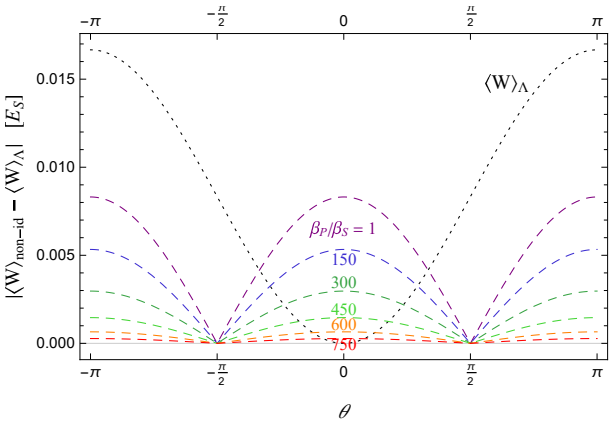

Figure 2. Deviation from the ideal work estimate for a qubit system and a 3-qubit pointer. For an atomic two-level system driven out of equilibrium by a classical electromagnetic field, the Rabi oscillations can be represented by the unitary $U_\Lambda(\theta) \equiv \exp\left(-i\frac{\theta}{2}\sigma_y\right)$ (for a derivation see Appendix A.7). The qubit is initially thermal, $\rho_S^{(0)} = \tau_S^{(0)}$, at room temperature $(k_B\beta_S)^{-1} = 300$K, and has a Hamiltonian $H_S^{(0)} = H_S^{(f)} = -E_S\sigma_z/2$ with $\sigma_z = |0\rangle\langle 0| - |1\rangle\langle 1|$ and an energy gap in the microwave regime such that $\beta_S E_S \approx 1/30$. The 3-qubit pointer is initially in the thermal state $\tau(\beta_P)^{\otimes 3}$ where each of the three qubits has the same Hamiltonian $H_P = -E_P\sigma_z/2$ and we assume $E_P = E_S/10$. The dashed lines show the deviation $|\langle W\rangle_{\text{non-id}} - \langle W\rangle_\Lambda|$ in units of E_S as a function of θ for different ratios of the system and pointer temperature, i.e., $\beta_P/\beta_S = 1$, and from $\beta_P/\beta_S = 150$ to 750 in steps of 150. The dotted black line shows the ideal work $\langle W\rangle_\Lambda$ (i.e., a pure state pointer). At $\theta = 0$, the deviation is maxmial while the ideal work estimate vanishes, whereas for $\theta = \pi$, the ratio $|\langle W\rangle_{\text{non-id}} - \langle W\rangle_\Lambda|/\langle W\rangle_\Lambda$ approaches 1/2 (the precise value is 0.49875).

where $\|A\|_\infty := \max_i \|\sum_j a_{ij}^*\|_1$ and $A = (a_{ij})$. In this sense, $C_{\max}$ can be thought of as representing the *trustworthiness* of the work estimate, in addition to its resource cost. For a fixed process Λ, the closer $C_{\max}$ is to 1, the smaller the potential distance of the work estimate from its ideal value, but also the higher the involved costs.

A principal purpose of this paper is to highlight that the difference $\langle W\rangle_{\text{non-id}} - \langle W\rangle_\Lambda$ is non-negligible and in some cases, rather significant. Indeed, in Fig. 2 we plot this difference for a standard system-pointer Hamiltonian with realistic parameters and find that the difference between the estimated ideal and non-ideal work is at times more than twice as large.

One should be careful not to confuse $\langle W\rangle_{\text{non-id}}$ with the total work performed on the system or with the system's change in average energy $\Delta E_{\text{non-id}}$. The latter can

be expressed as

$$\begin{aligned}\Delta E_{\text{non-id}} &= \sum_{m,n,k} q^{(\text{f})}_{kk|m}\, p(m|n)\, p^{(0)}_n \left(E^{(\text{f})}_k - E^{(0)}_n\right) \qquad (16)\\ &= C^2_{\max}\langle W\rangle_\Lambda + \sum_{\substack{m,n\\ l\neq n, k\neq m}} q^{(\text{f})}_{kk|m} p_{l\to m} q^{(0)}_{ll|n} p^{(0)}_m \left(E^{(\text{f})}_k - E^{(0)}_n\right)\\ &\quad + C_{\max}\sum_{m,n} p^{(0)}_n \Big[\sum_{k\neq m} q^{(\text{f})}_{kk|m}\, p_{n\to m}\left(E^{(\text{f})}_k - E^{(0)}_n\right)\\ &\qquad + \sum_{l\neq n} q^{(0)}_{ll|n}\, p_{l\to m}\left(E^{(\text{f})}_m - E^{(0)}_n\right)\Big],\end{aligned}$$

and generally contains contributions associated with both the work done on the system and heat transferred from the pointer to the system. Here, we make two observations. First, the work estimate $\langle W\rangle_{\text{non-id}}$ generally does not match the ideal work estimate $\langle W\rangle_\Lambda = \Delta E_\Lambda$, the actual work done on the system, or the change in average energy $\Delta E_{\text{non-id}}$. Second, while $\Delta E_{\text{non-id}}$ captures the average energy change of the system, the average energy change of the pointer is not yet included and has to be considered separately [14]. This contribution depends on the specific dimension of the pointer and the structure of its Hamiltonian, which in turn determine $C_{\max}$. Ideal measurements can be approached by increasing the pointer dimension (e.g., measuring a system with an N-qubit pointer and increasing N), or by cooling the pointer to a smaller but non-vanishing temperature (using a desired refrigeration paradigm [33, 34, 47–49]); these measures all increase the work cost of the measurement [14, A.7]. Achieving the limit $C_{\max}\to 1$, requires infinite time, infinite energy, or infinite control (e.g., $N\to\infty$) and in this limit one also recovers $\langle W\rangle_{\text{non-id}} = \langle W\rangle_\Lambda = \Delta E_\Lambda = \Delta E_{\text{non-id}}$. When limited by finite resources, $\langle W\rangle_{\text{non-id}} = \langle W\rangle_\Lambda$ can only be achieved for specific processes Λ, as we discuss in Appendix A.3.b.

IV. FLUCTUATION RELATIONS

Besides work estimates, higher statistical moments of work are relevant in many contexts, from the study of quenched quantum many-body systems [50, 51] to the performance of quantum thermal machines [52], or the development of effective charging protocols [53]. For discussions about the quantum-classical correspondence of work distributions see Refs. [54–58].

The quantification of work fluctuations [16–19] in the context of Jarzynski's and Crooks' fluctuation relations using the ideal TPM scheme has been studied in Ref. [21]. Here we are interested in exploring whether these universal relations are recovered within the non-ideal TPM scheme. In particular we explore the statistical properties of the work estimate $\langle W\rangle_{\text{non-id}}$. We first focus on the Jarzynski equality [59] for which the quantity of interest is the work functional $\langle e^{-\beta W}\rangle$. Using the properties of the non-ideal TPM scheme we obtain

$$\langle e^{-\beta W}\rangle_{\text{non-id}} = \chi\, e^{-\beta\Delta F}, \qquad (17)$$

where we have introduced the correction term

$$\chi := \frac{1}{\mathcal{Z}^{(\text{f})}}\sum_{n,m,l} e^{-\beta E^{(\text{f})}_m} q^{(0)}_{ll|n} |\langle E^{(\text{f})}_m|U_\Lambda|E^{(0)}_l\rangle|^2. \qquad (18)$$

Here, the partition function is $\mathcal{Z}^{(\text{f})} := \text{Tr}[e^{-\beta H^{(\text{f})}}]$, and $\Delta F := k_{\text{B}}T\log(\mathcal{Z}^{(0)}/\mathcal{Z}^{(\text{f})})$ is the difference in Helmholtz free energies between the initial state and the thermal state $\tau^{(\text{f})}$ w.r.t. $H^{(\text{f})}$. By construction $0\le\chi\le d_S$. When $\chi = 1$ one has the original Jarzynski relation, $\langle e^{-\beta W}\rangle = e^{-\beta\Delta F}$, which is satisfied by all ideal measurements. We notice that the breakdown of the original Jarzynski equality is expected here, since the invasiveness of the first non-ideal measurement spoils the requirement that the system starts the process Λ in thermal equilibrium.

Nonetheless, we find that there exists a class of non-ideal measurements for which $\chi = 1$ as well. This is the case when $\sum_n q^{(0)}_{ll|n} = 1\ \forall l$. This implies that in order to recover Jarzynski's relation, the matrix in Eq. (18) needs to be doubly stochastic. We call these measurements *minimally invasive UMC measurements*, and they correspond to unital maps, whose average effect is to preserve the identity operator on the system (for details see Appendix A.1.d). Using Eq. (17) and Jensen's inequality, $\exp\langle x\rangle \le \langle\exp x\rangle$, we can write the following second-law-like inequality

$$\langle W\rangle_{\text{non-id}} \ge \Delta F - k_{\text{B}}T\log\chi, \qquad (19)$$

which provides a lower bound for the non-ideal TPM work estimate $\langle W\rangle_{\text{non-id}}$. Since $0\le\chi\le d_S$, the extra term $-k_{\text{B}}T\log\chi$ can be either positive or negative. For non-ideal measurements that are unital ($\chi = 1$) this term vanishes and the equation above reduces to the usual second-law inequality $\langle W\rangle_{\text{non-id}} \ge \Delta F$.

Interestingly, we observe that minimal energy UMC measurements do not, in general, correspond to minimally invasive measurements (except for the special case of a single-qubit system, see Appendix A.1.d). This can be understood in the following way: For minimal energy UMC measurements, all elements of the post-interaction state $\tilde\rho_{SP}$ that depend on the outcome probabilities $\rho_{ii} = \langle i|\rho_S|i\rangle$ can be collected in a correlation matrix of dimension $d_P/d_S\times d_P/d_S$ with blocks $\Gamma_{ij} = |i\rangle\langle i|\otimes\Pi_j\tilde\rho_{SP}|i\rangle\langle i|\otimes\Pi_j$. Property (i) (unbiasedness) fixes $\text{Tr}(\Gamma_{ij}) = \rho_{jj}q_{ii|j}$ and maximal correlations are achieved when $q_{ii|i} = C_{\max}$ is satisfied. Minimal energy means that the remaining $q_{ii|j}$ for $i\neq j$ within each column are ordered such that $q_{ii|j}\ge q_{kk|j}\,\forall k\ge i$ with $k\neq j$. This is generally not compatible with row sums $\sum_j q_{ii|j} = 1$ required for $q_{ii|j}$ to be doubly stochastic and thus correspond to a minimally invasive measurement.

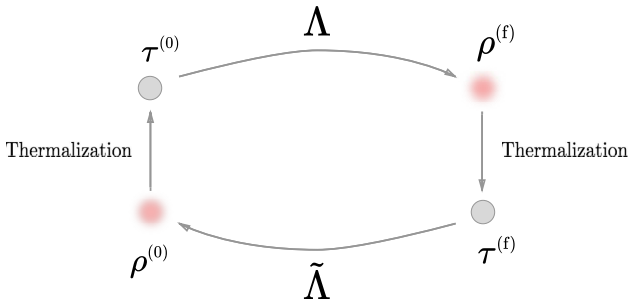

Figure 3. Schematic representing the non-reversibility of the process Λ. The system is initially at a thermal state $\tau^{(0)} = \exp(-\beta H^{(0)})/\mathcal{Z}^{(0)}$ is driven out of equilibrium by the unitary transformation U_Λ to the state $\rho^{(f)} = U_\Lambda \tau^{(0)} U_\Lambda^\dagger$. At the end of the process, the system is left to thermalise to the equilibrium state $\tau^{(f)} = \exp(-\beta H^{(f)})/\mathcal{Z}^{(f)}$, described by the Hamiltonian $H^{(f)}$. In the time reversed protocol, the thermal state $\tau^{(f)}$ is driven out of equilibrium to state $\rho^{(0)} = U_{\tilde{\Lambda}} \tau^{(f)} U_{\tilde{\Lambda}}^\dagger$. The process is reversible if $\rho^{(f)} = \tau^{(f)}$ and $\rho^{(0)} = \tau^{(0)}$.

One can, however, ensure that the measurement is minimally invasive *and* unbiased at the expense of moving away from the energy minimum. One can show that this additional cost only depends on the system Hamiltonian (and not the pointer Hamiltonian). Thus, by accepting this cost, non-ideal measurements can satisfy Jarzynski's relation.

To investigate the effect of non-ideal measurements on the irreversibilty of a process, we turn to Crooks' theorem [60]. This relates the probabilities $P_{\mathrm{F}}(W)$ of performing some work during a realisation of the TPM scheme and $P_{\mathrm{B}}(-W)$ for extracting the same amount of work when the time-reversed protocol $\tilde{\Lambda}$ is implemented:

$$P_{\mathrm{B}}(-W) = e^{-\beta(W-\Delta F)} P_{\mathrm{F}}(W). \tag{20}$$

The quantity $W - \Delta F$ is usually referred to as the *dissipated work*, the work which is lost when the final state of the TPM after the protocol, $\rho^{(f)}$ in Eq. (1), relaxes back to equilibrium at temperature T. As we discuss in Appendix A.6, Crooks' relation (20) is not recovered in the non-ideal TPM scheme, not even for minimally invasive UMC measurements (unlike the Jarzynski equality). This is in contrast to the ideal TPM scheme, where Crooks' theorem can be recovered for all unitary and unital maps [61–64]. The reason is that, here, both non-ideal measurements act as (independent) noise sources, disturbing the initial states of the forward and backward TPM processes, respectively. Reestablishing Crooks' fluctuation theorem may eventually require considering the work performed in the measurement processes and, therefore, taking into account the energy changes in the pointers.

Eq. (20) expresses the fact that finite-time processes that drive systems out of equilibrium are irreversible and thus the consumed work is unlikely to be recovered when reversing the protocol (See Fig. 3). This irreversibility can be captured [22, 23] by the average of the dissipated work appearing in Eq. (20), and is related to the entropy production during a hypothetical relaxation of $\rho^{(f)}$ to the thermal equilibrium state $\tau^{(f)}$ [64, 65]. When the measurements for determining this dissipated work are non-ideal, additional entropy is produced, resulting in greater energy dissipation in the final relaxation. In Appendix A.6.b, we calculate the average estimated work performed in addition to the free energy change to be

$$\langle W \rangle_{\text{non-id}} - \Delta F = k_{\mathrm{B}} T [\Delta S^{(0)} + D(\tilde{\rho}^{(f)} \| \tau^{(f)})], \tag{21}$$

where $\tilde{\rho}^{(f)}$ is the unconditional final state after the process Λ, $D(\rho^{(f)} \| \tau^{(f)}) \geq 0$ is the relative entropy quantifying the irreversibility in the ideal process [66, 67] and $\Delta S^{(0)} = S(\tilde{\rho}^{(0)}) - S(\tau^{(0)})$ is the corresponding change in von Neumann entropy of the system due to the first non-ideal measurement. For non-ideal, minimally invasive measurements $(\langle W \rangle_{\text{non-id}} - \Delta F) \geq 0$ in which case it may be interpreted as the entropy production. For these measurements, the von Neumann entropy of the system cannot decrease [67] and the entropy of the pointer during the measurement does not change (see, e.g., [64]). Thus, the total entropy produced in the measurement coincides with the entropy change in the system, $\Delta S^{(0)} \geq 0$. When ideal projective measurements are considered, $\Delta S^{(0)} = 0$, and the usual expression for the entropy production is recovered. In this ideal case, Kawai, Parrondo and Van den Broeck [22, 23] derived an important result in non-equilibrium thermodynamics, which is closely related to Crook's fluctuation theorem

$$\langle W \rangle_{\Lambda} - \Delta F = k_{\mathrm{B}} T D(\rho_{\mathrm{F}}(t) \| \Theta^\dagger \rho_{\mathrm{B}}(t_{\mathrm{f}} - t) \Theta). \tag{22}$$

Here, $\rho_{\mathrm{F}}(t)$ and $\rho_{\mathrm{B}}(t_{\mathrm{f}} - t)$ are the density operators in the forward and backward processes taken at the same instance of time t and Θ is the time-reversal operator in quantum mechanics. Θ is anti-unitary, satisfies $\Theta i \mathbb{1} = -i \mathbb{1} \Theta$ and $\Theta \Theta^\dagger = \Theta^\dagger \Theta = \mathbb{1}$, and is responsible for changing the sign of odd variables under time reversal (momentum, magnetic field, etc.) [68]. Equation (22) establishes a deep relationship between the physical and information-theoretical notion of irreversibility. Namely, it connects the dissipated work (left-hand side), which is a physical measure of irreversibility, with the relative entropy at any snapshot of time (right-hand side), which is an information-theoretical measure. We are able to extend this result to the non-ideal TPM scheme by using the generalised dissipation relation in Eq. (21). In the modified scheme we get that

$$\langle W \rangle_{\text{non-id}} - \Delta F = k_{\mathrm{B}} T [\Delta S_0 + \Delta D_{\mathrm{f}} + D(\rho_{\mathrm{F}}(t) \| \Theta^\dagger \rho_{\mathrm{B}}(t_{\mathrm{f}} - t) \Theta)], \tag{23}$$

where $\rho_{\mathrm{F}}(t) = U_\Lambda(t,0) \tilde{\rho}^{(f)} U_\Lambda^\dagger(t,0)$ is the system state at intermediate time $0 \leq t \leq t_{\mathrm{f}}$ in the forward process and

$\rho_{\rm B}(t_{\rm f}-t) = U_{\tilde{\Lambda}}(t_{\rm f}-t,0)\rho_{\rm B}^{(\rm f)}U_{\tilde{\Lambda}}^{\dagger}(t_{\rm f}-t,0)$ is the state of the system at the same instance of time in the backward process[5] (see Appendix A.6). We have introduced the correction term

$$\Delta D_{\rm f} = {\rm Tr}[\tilde{\rho}^{(\rm f)}\left(\log\tau^{(\rm f)} - \log\rho_{\rm B}^{(\rm f)}\right)], \tag{24}$$

whereby starting in $\tau^{(\rm f)}$, the average system state $\rho_{\rm B}^{(\rm f)}$ is obtained after the first non-ideal measurement in the backward process. The ideal case in Eq. (22) is recovered when $\rho_{\rm B}^{(\rm f)} = \tau^{(\rm f)}$ and $\tilde{\rho}^{(0)} = \tau^{(0)}$, making $\Delta D_{\rm f} = 0$ and $\Delta S^{(0)} = 0$. Finally, we stress that, analogously to what happens with the result in Eq. (22) for the ideal TPM scheme, Eqs. (21) and (23) can be turned into inequalities for open system dynamics (see Appendix A.6.b).

V. DISCUSSION

We have studied the consequences of fundamentally unavoidable measurement imperfections on the estimation of work and its fluctuations in out-of-equilibrium processes. Non-ideal measurements lead to a mismatch between the obtained estimate $\langle W\rangle_{\rm non-id}$, the desired ideal estimate $\langle W\rangle_{\Lambda}$, and the actual work performed on the system during the non-ideal TPM. In addition, an energy cost is incurred for operating the measurement apparatus. This leads to the conclusion that the process of estimating work itself has a work cost, which increases with increasing precision of the estimate. Moreover, we find that the statistical properties of the non-ideal estimate $\langle W\rangle_{\rm non-id}$ are modified. While the celebrated Jarzynski relation may be recovered exactly by imposing specific conditions on the measurement scheme, the more general Crooks theorem no longer holds. In this context, we discussed the connection between the non-ideal work estimate and the entropy production in the TPM scheme, and extended previous results for the relation between dissipation and irreversibility.

These results are of particular relevance for work extraction: When the costs for estimating the extracted work are of the order of the extracted work itself the usefulness of the procedure is dramatically limited. Conceptually, our results can be seen as a constructive resolution of the perceived shortcomings of the TPM discussed in [65]. It might also be interesting to consider work estimates as well as Jarzynski's equality and Crooks' theorem in more general contexts, such as including feedback control strategies [69, 70].

Our results about the validity of the Jarzynski equality and Crooks' fluctuation theorem for the non-ideal TPM scheme are in agreement with very recent results reported in Ref. [71], which appeared during the final stages of preparing this manuscript.

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APPENDICES

In these appendices, we provide more details on the mathematical model for non-ideal measurements from Ref. [14] and its application for work estimation described in this paper. In Appendix A.1, we describe the properties of the important class of *unbiased maximally correlated* (UMC) measurements, as well as of minimal energy UMC measurements and illustrate them for the case of 3-dim quantum system. In Appendix A.2, we give a detailed derivation of the joint probability for the measurement outcomes in the TPM scheme [20, 21] using non-ideal measurements. We then derive the corresponding work estimate in Appendix A.3 and the change in average energy throughout the estimation process in Appendix A.4. Finally, we discuss the consequences for fluctuation theorems: In Appendix A.5, we discuss the modification of Jarzynski's relation, while Appendix A.6 explains why Crooks' theorem generally no longer holds in the presence of non-ideal measurements. In Appendix A.7 we present a physical model as an example of the methods discussed in this work.

Appendix A.1: Ideal and Non-Ideal Measurements

We consider a measurement of a system described by an unknown quantum state $\rho_S \in \mathcal{D}(\mathcal{H}_S)$, where $\mathcal{D}(\mathcal{H}_S)$ represents the set of density-matrices over the Hilbert space $\mathcal{H}_S$. We model the measurement as an interaction between the system and a measurement apparatus (pointer) described by a quantum system with Hilbert space $\mathcal{H}_P$. We consider the pointer to be initially described by the thermal state $\tau_P(\beta_P) = \exp(-\beta_P H_P)/\mathcal{Z}_P$ at ambient temperature $T_P = (k_B\beta_P)^{-1}$ and with Hamiltonian $H_P = \sum_i E_i^{(P)}|E_i^{(P)}\rangle\langle E_i^{(P)}|$. This ensures that the initial state of the pointer does not contain any extractable work with respect to an environment at temperature T_P. Alternatively, one can consider the temperature T_P to be lower than the environment temperature, assuming that work has been invested to prepare the pointer by cooling it down to T_P. System and pointer are correlated by a unitary U_{meas} on the joint space $\mathcal{H}_S \otimes \mathcal{H}_P$ such that all work supplied to the joint system can be identified with the overall change in average energy due to the unitary transformation, resulting in a post-measurement state

$$\tilde{\rho}_{SP} := U_{\text{meas}}(\rho_S \otimes \tau_P)U_{\text{meas}}^\dagger. \tag{A.1.1}$$

Within the pointer Hilbert space, different subspaces are assigned to represent the different measurement outcomes. More specifically, if the system of dimension d_S is to be measured in the basis $\{|n\rangle\}_{n=0,\dots,d_S-1}$, then a set of orthogonal projectors $\{\Pi_n\}_{n=0,\dots,d_S-1}$ on the pointer Hilbert space is chosen to represent these outcomes, such that $\sum_n \Pi_n = \mathbb{1}_P$ and $\Pi_m\Pi_n = \delta_{mn}\Pi_n$ and $\mathrm{Tr}(\Pi_n)_P/d_S$ for all n. The general setup is illustrated in Fig. A.1.

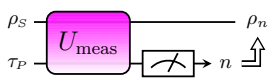

Figure A.1. Circuit representing non-ideal measurements. The system in an initial state ρ_S interacts with the pointer originally in a thermal state τ_P by means of unitary U_{meas}. The probability for obtaining a measurement outcome n is p_n, and the post-measurement system state conditioned on having obtained outcome n is ρ_n.

A.1.a. Unbiased Measurements

As explained in Sec. II (following Ref. [14]), ideal measurements have three characteristic properties, they are (ii) *faithful* (perfect correlation between the pointer and system), (i) *unbiased* (pointer exactly reproduces the system statistics in the measured basis), and (iii) *non-invasive* (system diagonal is undisturbed by the interaction). However, satisfying property (ii) requires the preparation of pure pointer states, which is not possible with finite resources according to the third law of thermodynamics. At the same time, this implies that realistic non-ideal measurements cannot satisfy both properties (i) and (iii) simultaneously, see Ref. [14, Appendix A.3]. We hence focus on non-ideal measurement procedures that are *unbiased*, i.e., which satisfy

$$\mathrm{Tr}\big(\mathbb{1}_S \otimes \Pi_n\tilde{\rho}_{SP}\big) = \mathrm{Tr}\big(|n\rangle\langle n|_S\, \rho_S\big) \;\forall\, n \quad \forall\, \rho_S\,. \tag{A.1.2}$$

Provided that the pointer cannot be prepared in a pure state (i.e., that the measurement uses finite resources according to the third law of thermodynamics), the corresponding unitary U_{meas} can be separated into two consecutive unitaries

$\tilde{U}$ and V, such that $U_{\text{meas}} = V\tilde{U}$, where

$$\tilde{U} = \sum_{n=0}^{d_S-1} |n\rangle\langle n|_S \otimes \tilde{U}^{(n)}, \qquad \text{and} \qquad V = \sum_{m,n=0}^{d_S-1} \sum_{i=1}^{d_P/d_S} |m\rangle\langle n|_S \otimes |\tilde{\psi}_i^{(n)}\rangle\langle \tilde{\psi}_i^{(m)}|_P \,. \tag{A.1.3}$$

Here, $\{|\tilde{\psi}_i^{(n)}\rangle\}_{i,n}$ is an orthonormal basis of $\mathcal{H}_P$ such that $\Pi_n = \sum_i |\tilde{\psi}_i^{(n)}\rangle\langle \tilde{\psi}_i^{(n)}|$ and the $\tilde{U}^{(n)}$ are arbitrary unitaries on $\mathcal{H}_P$, and we have assumed that the dimension d_P of the pointer is an integer multiple of the system dimension d_S (or is truncated to such a dimension). The intuition behind this decomposition is as follows. The operation V ensures unbiasedness by mapping the subspace corresponding to the system state $|n\rangle_S$ to the subspace corresponding to the pointer outcome n (i.e., Π_n), that is, V can be understood as a type of swap between these subspaces. The operation $\tilde{U}$, meanwhile, adds additional freedom by allowing unitary transformations within each of the subspaces for the states $|n\rangle_S$. To illustrate this transformation, let us consider an example of a 3-dimensional system with non-degenerate Hamiltonian, and a suitable pointer of dimension $d_P = 3\lambda$ for $\lambda \in \mathbb{N}$. The initial state $\rho_S \otimes \tau_P$ of system and pointer can be expressed in matrix form as

$$\rho_{SP} = \begin{array}{c} \left(\begin{array}{c|c|c} p_0\,\tau_P & \cdot & \cdot \\ \hline \cdot & p_1\,\tau_P & \cdot \\ \hline \cdot & \cdot & p_2\,\tau_P \end{array}\right) \\ \underbrace{}_{|0\rangle_S}\underbrace{}_{|1\rangle_S}\underbrace{}_{|2\rangle_S} \end{array}, \tag{A.1.4}$$

where $p_i = \langle i|\rho_S|i\rangle$ and the dots indicate potentially nonzero off-diagonal elements of ρ_S that are not shown here to keep the example simple, but could be included explicitly if desired. An unbiased measurement procedure realized by a unitary interaction with the pointer then leads to a final joint state of the form

$$\tilde{\rho}_{SP} = \begin{array}{c} \overbrace{\phantom{\Gamma_{00}}}^{\Pi_0}\overbrace{\phantom{\Gamma_{01}}}^{\Pi_1}\overbrace{\phantom{\Gamma_{02}}}^{\Pi_2}\overbrace{\phantom{\Gamma_{10}}}^{\Pi_0}\overbrace{\phantom{\Gamma_{11}}}^{\Pi_1}\overbrace{\phantom{\Gamma_{12}}}^{\Pi_2}\overbrace{\phantom{\Gamma_{20}}}^{\Pi_0}\overbrace{\phantom{\Gamma_{21}}}^{\Pi_1}\overbrace{\phantom{\Gamma_{22}}}^{\Pi_2} \\ \left(\begin{array}{c|c|c|c|c|c|c|c|c} \Gamma_{00} & \cdot & \cdot & & \cdot & \cdot & & \cdot & \cdot \\ \hline \cdot & \Gamma_{01} & \cdot & \cdot & & \cdot & \cdot & & \cdot \\ \hline \cdot & \cdot & \Gamma_{02} & \cdot & \cdot & & \cdot & \cdot & \\ \hline & \cdot & \cdot & \Gamma_{10} & \cdot & \cdot & & \cdot & \cdot \\ \hline \cdot & & \cdot & \cdot & \Gamma_{11} & \cdot & \cdot & & \cdot \\ \hline \cdot & \cdot & & \cdot & \cdot & \Gamma_{12} & \cdot & \cdot & \\ \hline & \cdot & \cdot & & \cdot & \cdot & \Gamma_{20} & \cdot & \cdot \\ \hline \cdot & & \cdot & \cdot & & \cdot & \cdot & \Gamma_{21} & \cdot \\ \hline \cdot & \cdot & & \cdot & \cdot & & \cdot & \cdot & \Gamma_{22} \end{array}\right) \\ \underbrace{\phantom{\Gamma_{00}\;\Gamma_{01}\;\Gamma_{02}}}_{|0\rangle_S}\underbrace{\phantom{\Gamma_{10}\;\Gamma_{11}\;\Gamma_{12}}}_{|1\rangle_S}\underbrace{\phantom{\Gamma_{20}\;\Gamma_{21}\;\Gamma_{22}}}_{|2\rangle_S} \end{array}, \tag{A.1.5}$$

where the Γ_{ij} are the $(d_P/d_S) \times (d_P/d_S)$ matrices corresponding to $|i\rangle\langle i|_S \otimes \Pi_j\, \tilde{\rho}_{SP}\, |i\rangle\langle i|_S \otimes \Pi_j$. The colour coding corresponds to the projections on to the subspaces for fixed outcomes. That is, the operators $\mathbb{1}_S \otimes \Pi_n\, \tilde{\rho}_{SP} \mathbb{1}_S \otimes \Pi_n$, can be written as

$$\begin{aligned} \mathbb{1}_S \otimes \Pi_n\, \tilde{\rho}_{SP} \mathbb{1}_S \otimes \Pi_n &= \sum_{m,m'=0}^{d_S-1} \sum_{i,j=1}^{d_P/d_S} \langle n|\rho_S|n\rangle\, \langle \tilde{\psi}_i^{(m)}|\tilde{U}^{(n)}\tau_P\tilde{U}^{(n)\dagger}|\tilde{\psi}_j^{(m')}\rangle\; |m\rangle\langle m'|_S \otimes |\tilde{\psi}_i^{(n)}\rangle\langle \tilde{\psi}_j^{(n)}|_P \\ &\hat{=}\; p_n\, \tilde{U}^{(n)}\tau_P\tilde{U}^{(n)\dagger}. \end{aligned} \tag{A.1.6}$$

In other words, the $\mathbb{1}_S \otimes \Pi_n\, \tilde{\rho}_{SP} \mathbb{1}_S \otimes \Pi_n$ for $n \in \{0, 1, \ldots, d_S - 1\}$ have rank d_P and can be understood as $d_P \times d_P$ matrices with the same spectra as $p_n \tau_P$. They are in this sense unitarily equivalent [via application of the unitaries $\tilde{U}^{(n)}$ from Eq. (A.1.3)] to $p_n \tau_P$, such that the trace of these operators gives p_n, as required by unbiasedness in Eq. (A.1.2). In terms of the indicated diagonal blocks of each subspace for fixed Π_n (fixed colour), we have $\sum_{i=0}^{2} \mathrm{Tr}(\Gamma_{in}) = p_n \;\forall\, n$. More specifically, since the measurement has to be unbiased independently of the initial system state ρ_S, one has to have

$$\mathrm{Tr}_P\big(\mathbb{1}_S \otimes \Pi_n\, \tilde{\rho}_{SP} \mathbb{1}_S \otimes \Pi_n\big) = p_n\, \rho_n, \tag{A.1.7}$$

where $\rho_n \in \mathcal{D}(\mathcal{H}_S)$ is the post-measurement system state conditioned on observing the pointer outcome n. The conditional states ρ_n correspond to coarse-grainings of the operators $\tilde{U}^{(n)}\tau_P\tilde{U}^{(n)\dagger}$ in the sense of the equivalence "$\hat{=}$" of Eq. (A.1.6). The states ρ_n are hence independent of ρ_S. We write them as $\rho_n = \sum_{l,l'} q_{ll'|n} \,|\, l \,\rangle\!\langle\, l' \,|$ with $\sum_l q_{ll|n} = 1 \;\forall\, n$ and

$$q_{ll'|n} = \sum_{i=1}^{d_P/d_S} \langle\, \tilde{\psi}_i^{(l)} \,|\, \tilde{U}^{(n)}\tau_P\tilde{U}^{(n)\dagger} \,|\, \tilde{\psi}_i^{(l')} \,\rangle\,. \tag{A.1.8}$$

A.1.b. Unbiased Maximally Correlated Measurements

Within the set of unbiased measurement procedures, one may then wish to select those measurements which maximise the correlation measure $C(\tilde{\rho}_{SP})$ that is used in property (ii) to define faithful measurements (for which $C = 1$). For unbiased measurements that are realised unitarily from an initially thermal pointer state, the maximal value $C = C_{\max}$ for unbiased measurements is obtained when the d_P/d_S largest eigenvalues of τ_P appear as the eigenvalues of the matrices Γ_{nn} for each n. This requires the off-diagonal elements in $\tilde{\rho}_{SP}$ connecting the correlated subspaces defined by the projectors $|\, n \,\rangle\!\langle\, n \,| \otimes \Pi_n$ with the uncorrelated subspaces defined by the projectors $|\, m \,\rangle\!\langle\, m \,| \otimes \Pi_n$ for $m \neq n$ to vanish, resulting in a joint state of the form

$$\tilde{\rho}_{SP} = \begin{array}{c} \begin{array}{ccccccccc} \Pi_0 & \Pi_1 & \Pi_2 & \Pi_0 & \Pi_1 & \Pi_2 & \Pi_0 & \Pi_1 & \Pi_2 \end{array} \\ \left(\begin{array}{ccc|ccc|ccc} \Gamma_{00} & \cdot & \cdot & 0 & \cdot & \cdot & 0 & \cdot & \cdot \\ \cdot & \Gamma_{01} & \cdot & \cdot & 0 & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \Gamma_{02} & \cdot & \cdot & \cdot & \cdot & \cdot & 0 \\ \hline 0 & \cdot & \cdot & \Gamma_{10} & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & 0 & \cdot & \cdot & \Gamma_{11} & \cdot & \cdot & 0 & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \Gamma_{12} & \cdot & \cdot & 0 \\ \hline 0 & \cdot & \cdot & \cdot & \cdot & \cdot & \Gamma_{20} & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & 0 & \cdot & \cdot & \Gamma_{21} & \cdot \\ \cdot & \cdot & 0 & \cdot & \cdot & 0 & \cdot & \cdot & \Gamma_{22} \end{array}\right) \\ \underbrace{\qquad\qquad}_{|\,0\,\rangle_S} \underbrace{\qquad\qquad}_{|\,1\,\rangle_S} \underbrace{\qquad\qquad}_{|\,2\,\rangle_S} \end{array}, \tag{A.1.9}$$

where the remaining blocks in the correlated subspace now satisfy $\mathrm{Tr}(\Gamma_{nn}) = p_n C_{\max}$, see Ref. [14, Eq. (5)]. From Eq. (A.1.7) we can then conclude that the coefficients of the conditional states for unbiased maximally correlated measurements (UMC) satisfy

$$q_{nn|n} = C_{\max} = \sum_{i=0}^{d_P/d_S-1} \frac{\exp\left(-\beta_P E_i^{(P)}\right)}{\mathcal{Z}_P} \qquad \forall\; n, \tag{A.1.10}$$

where we have assumed that the eigenvalues of the pointer Hamiltonian are ordered non-decreasingly, i.e., $E_i^{(P)} \geq E_j^{(P)}$ for $i \geq j$, while some of the off-diagonals of ρ_n are more restricted, $q_{nl|n} = q_{ln|n} = 0 \forall l \neq n$. In particular, this last condition implies that the conditional post-measurement state of the system for UMC measurements takes the block-diagonal form

$$\rho_n = C_{\max} \,|\, n \,\rangle\!\langle\, n \,| + (1 - C_{\max})\, \rho_n^{\text{error}}, \tag{A.1.11}$$

where ρ_n^{error} is density operator on the Hilbert space spanned by the vectors $\{|\, m \,\rangle_S\}$ for $m \in \{0, 1, \ldots, d_S - 1\}/n$ such that $\langle\, n \,|\, \rho_n^{\text{error}} \,|\, l \,\rangle = \langle\, n \,|\, \rho_l^{\text{error}} \,|\, n \,\rangle = 0 \,\forall\, l$.

The maximal correlation value $C_{\max}$ can further be used to quantify the distance between the conditional post-measurement state ρ_n of UMC measurements and the pure state $|\, n \,\rangle$. Using the trace distance $D(X, Y) = \frac{1}{2} \|X - Y\|_1 \equiv$

$\frac{1}{2}\mathrm{Tr}\sqrt{(X-Y)(X-Y)^\dagger}$ between two operators X and Y, and the block-diagonal structure of ρ_n we can calculate

$$\begin{aligned} D(\rho_n, |n\rangle\langle n|) &= \frac{1}{2}\mathrm{Tr}\sqrt{\left[\rho_n - |n\rangle\langle n|\right]^2} = \frac{1}{2}\mathrm{Tr}\sqrt{\left[(C_{\max}-1)\,|n\rangle\langle n| + (1-C_{\max})\rho_n^{\mathrm{error}}\right]^2} \\ &= \frac{1}{2}\mathrm{Tr}\sqrt{(C_{\max}-1)^2\,|n\rangle\langle n| + (1-C_{\max})^2(\rho_n^{\mathrm{error}})^2} = \frac{1}{2}\left[(1-C_{\max})\mathrm{Tr}(|n\rangle\langle n|) + (1-C_{\max})\mathrm{Tr}(\rho_n^{\mathrm{error}})\right] \\ &= 1 - C_{\max}. \end{aligned} \tag{A.1.12}$$

A.1.c. Minimal Energy UMC Measurements

One can then further restrict the set of considered unitaries U_{meas} by demanding minimal energy consumption. That is, that the measurement implemented by U_{meas} achieves the algebraic maximum of correlations *and* spends the least amount of energy as compared with all other unitary operations achieving $C_{\max}$, i.e.,

$$\Delta E_{\mathrm{meas}} = \min_{U_{\mathrm{meas}}} \mathrm{Tr}\big[(H_S + H_P)(\tilde{\rho}_{SP} - \rho_S \otimes \tau_P)\big] \quad \text{s.t.} \quad C(\tilde{\rho}_{SP}) = C_{\max}. \tag{A.1.13}$$

A requirement in order to spend the minimum amount of energy in the measurement process is not to waste energy on creating coherences [72]. Conversely, any coherence with respect to energy eigenstate with different energies would imply that there exists a unitary transformation that shifts probability from higher energies to lower energies. In other words, such states would not be passive [46]. For the sake of illustrating the effect on the form of the final state for our example, let us assume that both the system and pointer Hamiltonians are non-degenerate. Then, the final state for a minimal energy UMC measurement must be of the form

$$\tilde{\rho}_{SP} = \begin{array}{c} \begin{array}{ccccccccc} \Pi_0 & \Pi_1 & \Pi_2 & \Pi_0 & \Pi_1 & \Pi_2 & \Pi_0 & \Pi_1 & \Pi_2 \end{array} \\ \begin{pmatrix} \Gamma_{00} & \cdot & \cdot & 0 & \cdot & \cdot & 0 & \cdot & \cdot \\ \cdot & \Gamma_{01} & \cdot & \cdot & 0 & \cdot & \cdot & 0 & \cdot \\ \cdot & \cdot & \Gamma_{02} & \cdot & \cdot & 0 & \cdot & \cdot & 0 \\ 0 & \cdot & \cdot & \Gamma_{10} & \cdot & \cdot & 0 & \cdot & \cdot \\ \cdot & 0 & \cdot & \cdot & \Gamma_{11} & \cdot & \cdot & 0 & \cdot \\ \cdot & \cdot & 0 & \cdot & \cdot & \Gamma_{12} & \cdot & \cdot & 0 \\ 0 & \cdot & \cdot & 0 & \cdot & \cdot & \Gamma_{20} & \cdot & \cdot \\ \cdot & 0 & \cdot & \cdot & 0 & \cdot & \cdot & \Gamma_{21} & \cdot \\ \cdot & \cdot & 0 & \cdot & \cdot & 0 & \cdot & \cdot & \Gamma_{22} \end{pmatrix} \\ \underbrace{\qquad\qquad}_{|0\rangle_S} \underbrace{\qquad\qquad}_{|1\rangle_S} \underbrace{\qquad\qquad}_{|2\rangle_S} \end{array}, \tag{A.1.14}$$

where all remaining diagonal blocks Γ_{mn} must now be diagonal matrices themselves, and $\mathrm{Tr}(\Gamma_{nn}) = p_n C_{\max}\ \forall\, n$ as before. In addition, the energy minimisation imposes constraints on (I) the assignment of the remaining eigenvalues of τ_P to the matrices Γ_{mn} for $m \neq n$ in the uncorrelated subspaces, and (II) on the ordering of the eigenvalues within each specific block. What is crucial here is the observation that (I) corresponds to arranging the eigenvalues of τ_P in descending order and splitting the resulting list into three (in general d_S) sets $a^{(0)}$, $a^{(1)}$, and $a^{(2)}$ that we can interpret as diagonal (d_P/d_S) matrices with diagonal elements

$$a_k^{(n)} = \tau_{n\frac{d_P}{d_S}+k} \tag{A.1.15}$$

for $k = 1, \ldots, d_P/d_S$ and $n = 0, \ldots, d_S - 1$. Minimising the energy while maintaining a UMC measurement then means that the set $a^{(0)}$ of largest entries is assigned to the correlated subspaces, $\Gamma_{nn} = p_n\, a^{(0)}\ \forall\, n$, and the other $a^{(i)}$ with $i > 0$ are assigned to the blocks Γ_{mn} with $m \neq n$ such that the lower energies are combined with the larger weights (higher values of i).

At this point, it becomes useful to switch to a reduced description of the final state that captures only the diagonal blocks Γ_{ij}. We therefore define a quantity we refer to as the correlation matrix $\Gamma = (\Gamma_{ij})$ in this context, where, as

before, the Γ_{ij} are the $(d_P/d_S)\times(d_P/d_S)$ matrices corresponding to $|i\rangle\langle i|_S \otimes \Pi_j\, \tilde{\rho}_{SP}\, |i\rangle\langle i|_S \otimes \Pi_j$. With this notation, the correlation matrix of the final state for a minimal energy UMC measurement takes the form

$$\Gamma = \left[\begin{array}{c|c|c} \Gamma_{00} & \Gamma_{01} & \Gamma_{02} \\ \hline \Gamma_{10} & \Gamma_{11} & \Gamma_{12} \\ \hline \Gamma_{20} & \Gamma_{21} & \Gamma_{22} \end{array}\right] = \left[\begin{array}{c|c|c} p_0\, a^{(0)} & p_1\, a^{(1)} & p_2\, a^{(1)} \\ \hline p_0\, a^{(1)} & p_1\, a^{(0)} & p_2\, a^{(2)} \\ \hline p_0\, a^{(2)} & p_1\, a^{(2)} & p_2\, a^{(0)} \end{array}\right]. \tag{A.1.16}$$

Alternatively, we may write the correlation matrix elements Γ_{ij} as $\Gamma_{ij} = p_j\, a^{(\tilde{\pi}_{ij})}$ where $\tilde{\pi} = (\tilde{\pi}_{ij})$ is a $d_S \times d_S$ matrix with entries $\tilde{\pi}_{ij} \in \{0, 1, \ldots, d_S - 1\}$. Unbiasedness requires that each element of $\{0, 1, \ldots, d_S - 1\}$ appears exactly once in each column of $\tilde{\pi}$, that is, $\{\tilde{\pi}_{ij}\}_{i=0,1,\ldots,d_S} = \{0, 1, \ldots, d_S - 1\}$. Achieving maximal correlation $C_{\max}$ for an unbiased measurement further fixes the diagonal of $\tilde{\pi}$, i.e., $\tilde{\pi}_{ii} = 0\ \forall\, i$. Finally, minimal energy of UMC measurements implies that the off-diagonal elements of $\tilde{\pi}$ are arranged such that the lowest values fill up the top rows (for lowest row index i) of $\tilde{\pi}$ first, that is, for a fixed j $\tilde{\pi}_{ij} = i + 1$ if $i < j$ and $\tilde{\pi}_{ij} = j$ if $i > j$. More details on minimal energy UMC measurements can be found in Ref. [14, Appendices A.8 and A.9].

A.1.d. Minimally Invasive UMC Measurements

As we have discussed in Sec. II (following Ref. [14]), non-ideal measurement procedures cannot be both unbiased and non-invasive. However, while one cannot construct an unbiased measurement that is non-invasive for all initial system states ρ_S, one can indeed construct unbiased measurements that leave (the diagonal of) certain system states invariant. One case that is of specific interest here (and goes beyond what is considered in Ref. [14]) is the case of UMC measurements, which leave the maximally mixed state $\rho_S = \mathbb{1}_S/d_S$ invariant. For this maximum entropy state, all initial probabilities are the same, $p_n = 1/d_S\ \forall\, n$. At the same time, we observe that the diagonal elements $\langle n|\,\mathrm{Tr}_P(\tilde{\rho}_{SP})\,|n\rangle$ of the post-measurement system state are obtained by taking the trace of the sum of the elements of the nth row of the correlation matrix Γ.

We thus observe that such a map, which we call *minimally invasive UMC* measurements, can be realized if each row of Γ features each superscript index i of $a^{(i)}$ exactly once. Together with the unbiasedness requirement we see that unbiased measurements are minimally invasive if and only if $\tilde{\pi}$ is a Latin square. That is, each row and each column of $\tilde{\pi}$ features each element once and only once. It is interesting to note that any minimal energy UMC measurement can thus be turned into a minimally invasive UMC measurement, and vice versa, by rearranging the entries of the Γ within its columns only. This implies that the additional energy cost for moving away from the energy minimum depends only on the spectrum of the system Hamiltonian, but not on the pointer Hamiltonian. At the same time, this can be done in such a way that the energy is only minimally increased with respect to the minimal energy UMC measurements, obtaining a *minimal energy minimally invasive UMC measurement*. For the example in $d_S = 3$ discussed above, the correlation matrix for such a measurement takes the form

$$\Gamma = \left[\begin{array}{c|c|c} \Gamma_{00} & \Gamma_{01} & \Gamma_{02} \\ \hline \Gamma_{10} & \Gamma_{11} & \Gamma_{12} \\ \hline \Gamma_{20} & \Gamma_{21} & \Gamma_{22} \end{array}\right] = \left[\begin{array}{c|c|c} p_0\, a^{(0)} & p_1\, a^{(2)} & p_2\, a^{(1)} \\ \hline p_0\, a^{(1)} & p_1\, a^{(0)} & p_2\, a^{(2)} \\ \hline p_0\, a^{(2)} & p_1\, a^{(1)} & p_2\, a^{(0)} \end{array}\right]. \tag{A.1.17}$$

A property of minimally invasive UMC measurements that follows directly from the form described above is that the conditional probabilities $q_{l|n}$ for the system to be left in the lth energy eigenstate given a pointer outcome n form a doubly stochastic matrix, i.e.,

$$\sum_l q_{l|n} = \sum_n q_{l|n} = 1. \tag{A.1.18}$$

As we will see in Appendix A.5, minimally invasive UMC measurements allow satisfying the Jarzynski relation.

Appendix A.2: The Non-Ideal TPM Scheme

The estimation of the work performed/extracted during the process $(\tau^{(0)}, H^{(0)}) \xrightarrow{\Lambda} (\rho^{(f)}, H^{(f)})$ can be calculated by means of the so called two projective measurement (TPM) process [20, 21], which consists of three steps. In the first, one performs a projective measurement on $\tau^{(0)}$ in the eigenbasis $\{|E_n^{(0)}\rangle\}_{n=0,\ldots,d_S-1}$ of the initial Hamiltonian $H^{(0)}$, obtaining an outcome n. Then one lets the resulting post-measurement system state evolve under the action of U_Λ.

Finally, a second projective measurement is performed in the eigenbasis $\{|E_n^{(\mathrm{f})}\rangle\}_{n=0,\dots,d_S-1}$ of the final Hamiltonian $H^{(\mathrm{f})}$, resulting in an outcome m. The aim of the protocol is to estimate the work performed (or extracted) by the process Λ based on the joint probability distribution $p(n,m)$, which itself is estimated by repeating the procedure sufficiently many times. For ideal projective measurements, the joint probability distribution can be written as [21]

$$p(n,m) = p_n^{(0)}\, p_{n\to m}, \tag{A.2.1}$$

where $p_n^{(0)} = \frac{1}{\mathcal{Z}^{(0)}}\exp(-\beta E_n^{(0)})$ are the diagonal elements of the initial thermal state $\tau^{(0)}$ of the system, $\mathcal{Z}^{(0)} = \sum_n \exp(-\beta E_n^{(0)})$ is the partition function w.r.t. the initial Hamiltonian and inverse temperature β. Note that the inverse system temperature β need not match the initial inverse temperature β_P of the pointer, which enters $C_{\max}$ from Eq. (A.1.10). The symbol $p_{n\to m} = |\langle E_m^{(\mathrm{f})}|U_\Lambda|E_n^{(0)}\rangle|^2$ denotes the transition probability from an initial energy eigenstate $|E_n^{(0)}\rangle$ to a final energy eigenstate $|E_m^{(\mathrm{f})}\rangle$. The work distribution, i.e., the probability density for performing (or extracting) the work W given the process Λ is defined as

$$P(W) = \sum_{m,n} p(n,m)\,\delta\left(E_m^{(\mathrm{f})} - E_n^{(0)} - W\right). \tag{A.2.2}$$

On average the work spent or extracted by the process Λ is then obtained by integration, i.e.,

$$\langle W\rangle = \int P(W)\,W\,dW, \tag{A.2.3}$$

where the integral is taken over all possible values of work. The average work can be written in terms of the joint probability $p(n,m)$ by inserting Eq. (A.2.2) into Eq. (A.2.3), resulting in the expression

$$\langle W\rangle = \sum_{m,n} p(n,m)\left(E_m^{(\mathrm{f})} - E_n^{(0)}\right). \tag{A.2.4}$$

Let us now consider the TPM scheme when the ideal projective measurements are replaced by non-ideal measurements, more specifically, minimal energy UMC measurements as described in Appendix A.1. We discuss each step of the process in detail below.

A.2.a. First Measurement

First, note that the initial system state for the TPM scheme is a Gibbs equilibrium state at the ambient temperature. That is, here the system state to be measured during the first measurement is $\rho_S = \tau^{(0)}(\beta)$, given by

$$\tau^{(0)}(\beta) = \frac{1}{\mathcal{Z}^{(0)}}\exp(-\beta H^{(0)}) = \frac{1}{\mathcal{Z}^{(0)}}\sum_n \exp(-\beta E_n^{(0)})|E_n^{(0)}\rangle\langle E_n^{(0)}|, \tag{A.2.5}$$

with $H^{(0)} = \sum_n E_n^{(0)}|E_n^{(0)}\rangle\langle E_n^{(0)}|$ at time t_0. Then, we assume that the first non-ideal measurement is performed in the eigenbasis of $H^{(0)}$. We assume that this measurement, though non-ideal, is unbiased [property (i) described in Sec. II.2], such that a measurement result n is obtained with probability $p_n^{(0)} = \langle E_n^{(0)}|\tau^{(0)}|E_n^{(0)}\rangle = \exp(-\beta E_n^{(0)})/\mathcal{Z}^{(0)}$. Moreover, from here on, we restrict our investigation to non-ideal unbiased measurements that achieve maximal correlation $C_{\max}$ (UMC measurements) and either have *minimal energy* or are *minimal energy minimally invasive measurements*, as described in Appendices A.1.c and A.1.d, respectively, such that the post-measurement state of the system is diagonal w.r.t. the measurement basis, i.e.,

$$\rho_n^{(0)} = \sum_l q_{ll|n}^{(0)}|E_l^{(0)}\rangle\langle E_l^{(0)}| \quad \forall\, n, \tag{A.2.6}$$

where $q_{ll|n}^{(0)}$ is the the probability to find the post-measurement system in the state $|E_l^{(0)}\rangle$ given the measurement result n. In particular, the probability to correctly guess the energy eigenstate $|E_n^{(0)}\rangle$, when the pointer shows n is given by $q_{nn|n}^{(0)}$, which has the value

$$q_{nn|n}^{(0)} = C_{\max} \tag{A.2.7}$$

for UMC measurements. The conditional probabilities $q_{ll|n}^{(0)}$ are functions of the inverse temperature β, and the pointer energies (since the initial pointer state is also a thermal state at ambient temperature $T = 1/k_{\mathrm{B}}\beta$), but because the measurement is unbiased, the conditional state $\rho_n^{(0)}$ is independent of the system state $\tau^{(0)}$ before the measurement.

A.2.b. Evolution and Second Measurement

After the initial non-ideal measurement, the post-measurement conditional state $\rho_n^{(0)}$ is evolving according to the unitary U_Λ corresponding to the process Λ. The unitary U_Λ can be written as time evolution from an initial time t_0 to a final time t_f, $U_\Lambda = \mathcal{T}_+ \exp\left(-i\int_{t_0}^{t_\mathrm{f}} H(\lambda_t)\right) dt$, where $\mathcal{T}_+$ denotes time-ordering, and λ_t is a control parameter for the time-dependent Hamiltonian such that $H(\lambda_{t_0}) = H^{(0)}$ and $H(\lambda_{t_\mathrm{f}}) = H^{(\mathrm{f})}$. The time-evolved state prior to the second measurement is thus

$$\rho_n^\Lambda = U_\Lambda \rho_n^{(0)} U_\Lambda^\dagger = \sum_l q_{ll|n}^{(0)} U_\Lambda |E_l^{(0)}\rangle\langle E_l^{(0)}| U_\Lambda^\dagger \,. \tag{A.2.8}$$

Finally, a second non-ideal measurement is performed on ρ_n^Λ w.r.t. the eigenbasis $\{|E_m^{(\mathrm{f})}\rangle\}$ of $H^{(\mathrm{f})}$. Once again, since the measurement is unbiased, the probability to obtain any particular outcome m only depends on the specific system state prior to the measurement. However, in this case this state is ρ_n^Λ from Eq. (A.2.8) and hence depends on the first outcome n. The conditional probability to obtain outcome m in the second measurement given outcome n in the first measurement is thus

$$p(m|n) = \langle E_m^{(\mathrm{f})} | \rho_n^\Lambda | E_m^{(\mathrm{f})} \rangle = \sum_l q_{ll|n}^{(0)} \langle E_m^{(\mathrm{f})} | U_\Lambda | E_l^{(0)} \rangle\langle E_l^{(0)} | U_\Lambda^\dagger | E_m^{(\mathrm{f})} \rangle = \sum_l q_{ll|n}^{(0)} p_{l\to m}, \tag{A.2.9}$$

where $p_{l\to m} = |\langle E_m^{(\mathrm{f})} | U_\Lambda | E_l^{(0)} \rangle|^2$. Consequently, the joint probability $p(n,m)$ is obtained by multiplying with the probability to obtain outcome n in the first measurement, i.e.,

$$p(n,m) = p(m|n)\, p_n^{(0)} = p_n^{(0)} \sum_l q_{ll|n}^{(0)} p_{l\to m}. \tag{A.2.10}$$

Meanwhile, the post-measurement state after the second measurement, conditioned on having obtained outcome n and m in the first and second measurement, respectively, is

$$\rho_{m|n}^{(\mathrm{f})} = \sum_k q_{kk|m}^{(\mathrm{f})} |E_k^{(\mathrm{f})}\rangle\langle E_k^{(\mathrm{f})}| \quad \forall\, m \,. \tag{A.2.11}$$

As in the first measurement, the conditional post-measurement state of the system is independent of the pre-measurement system state. In this case, this implies that $\rho_{m|n}^{(\mathrm{f})}$ is indeed independent of the outcome n of the first measurement.

Appendix A.3: Work Estimation in the Non-Ideal TPM Scheme

A.3.a. Non-Ideal Work Distribution and Estimate

To estimate the work in the TPM scheme based on non-ideal measurements (as before, we assume minimal energy UMC measurements as described in Appendix A.1), some energy must be spent on the measurement processes, where the precision in the estimate is directly dependent on the energy spent. Considering the joint probability in Eq. (A.2.10) and the definition in Eq. (A.2.2), the probability distribution for inferring the work value W is

$$P(W) = \sum_{m,n} p_n^{(0)} \sum_l q_{ll|n}^{(0)} |\langle E_m^{(\mathrm{f})} | U_\Lambda | E_l^{(0)} \rangle|^2 \, \delta\left((E_m^{(\mathrm{f})} - E_n^{(0)}) - W\right). \tag{A.3.1}$$

We can now calculate the average work,

$$\begin{aligned}\langle W\rangle_{\text{non-id}} &= \int dW\, P(W) W = \sum_{m,n} p_n^{(0)} \sum_l q_{ll|n}^{(0)} |\langle E_m^{(\mathrm{f})} | U_\Lambda | E_l^{(0)} \rangle|^2 \, (E_m^{(\mathrm{f})} - E_n^{(0)}) \,, \\ &= \sum_{m,n} q_{nn|n}^{(0)} p_n^{(0)} |\langle E_m^{(\mathrm{f})} | U_\Lambda | E_n^{(0)} \rangle|^2 \, (E_m^{(\mathrm{f})} - E_n^{(0)}) + \sum_{m,n} \sum_{l\neq n} q_{ll|n}^{(0)} p_n^{(0)} |\langle E_m^{(\mathrm{f})} | U_\Lambda | E_l^{(0)} \rangle|^2 \, (E_m^{(\mathrm{f})} - E_n^{(0)}) \,.\end{aligned} \tag{A.3.2}$$

Substituting for C_max in Eq. (A.2.7), we arrive at

$$\langle W\rangle_{\text{non-id}} = C_\mathrm{max} \sum_{m,n} p_n^{(0)} |\langle E_m^{(\mathrm{f})} | U_\Lambda | E_n^{(0)} \rangle|^2 \, (E_m^{(\mathrm{f})} - E_n^{(0)}) + \sum_{m,n} \sum_{l\neq n} q_{ll|n}^{(0)} p_n^{(0)} |\langle E_m^{(\mathrm{f})} | U_\Lambda | E_l^{(0)} \rangle|^2 \, (E_m^{(\mathrm{f})} - E_n^{(0)}) \,. \tag{A.3.3}$$

Since $\langle W\rangle_\Lambda = \sum_{m,n} p_n^{(0)} |\langle E_m^{(\mathrm{f})} | U_\Lambda | E_n^{(0)} \rangle|^2 \, (E_m^{(\mathrm{f})} - E_n^{(0)})$ we can write the non-ideal work estimate as

$$\langle W\rangle_{\text{non-id}} = C_\mathrm{max} \langle W\rangle_\Lambda + \sum_{m,n} \sum_{l\neq n} q_{ll|n}^{(0)} p_n^{(0)} |\langle E_m^{(\mathrm{f})} | U_\Lambda | E_l^{(0)} \rangle|^2 \, (E_m^{(\mathrm{f})} - E_n^{(0)}) \,. \tag{A.3.4}$$

A.3.b. Ideal versus Non-Ideal Work Estimate

Before moving on, let us discuss more about the difference between the work estimates based on ideal and non-ideal (UMC) measurements. Noting that $\langle W\rangle_\Lambda = \Delta E_\Lambda = \mathrm{Tr}(U_\Lambda \tau^{(0)} U_\Lambda^\dagger H^{(\mathrm{f})}) - \mathrm{Tr}(\tau^{(0)} H^{(0)})$ and $\mathrm{Tr}(\tau^{(0)} H^{(0)}) = \sum_n p_n^{(0)} E_n^{(0)}$, we can use the expression of $\langle W\rangle_{\text{non-id}}$ from Eq. (A.3.2) to write

$$\langle W\rangle_{\text{non-id}} - \langle W\rangle_\Lambda = \sum_{m,n} p_n^{(0)} \sum_l q_{ll|n}^{(0)} |\langle E_m^{(\mathrm{f})} | U | E_l^{(0)} \rangle|^2 E_m^{(\mathrm{f})} - \mathrm{Tr}(U_\Lambda \tau^{(0)} U_\Lambda^\dagger H^{(\mathrm{f})}). \tag{A.3.5}$$

The first term on the right-hand side of Eq. (A.3.5) can be rewritten as

$$\sum_{m,n} p_n^{(0)} \sum_l q_{ll|n}^{(0)} |\langle E_m^{(\mathrm{f})} | U_\Lambda | E_l^{(0)} \rangle|^2 E_m^{(\mathrm{f})} = \mathrm{Tr}\Big(\sum_n p_n^{(0)} \sum_l q_{ll|n}^{(0)} U_\Lambda |E_l^{(0)}\rangle\langle E_l^{(0)}| U_\Lambda^\dagger H^{(\mathrm{f})}\Big) = \mathrm{Tr}\Big(U_\Lambda \tilde{\rho}_S^{(0)} U_\Lambda^\dagger H^{(\mathrm{f})}\Big), \tag{A.3.6}$$

where we have denoted the unconditional post-measurement state after the first measurement as $\tilde{\rho}^{(0)} := \sum_n p_n^{(0)} \rho_n^{(0)}$ and we have used $\rho_n^{(0)} = \sum_l q_{ll|n}^{(0)} |E_l^{(0)}\rangle\langle E_l^{(0)}|$. With this, we can rewrite Eq. (A.3.5) as

$$\langle W\rangle_{\text{non-id}} - \langle W\rangle_\Lambda = \mathrm{Tr}\left(U_\Lambda \big(\tilde{\rho}^{(0)} - \tau^{(0)}\big) U_\Lambda^\dagger H^{(\mathrm{f})}\right). \tag{A.3.7}$$

If $T \to 0$, then $\tilde{\rho}^{(0)} = \tau^{(0)}$ for any process Λ, which means that $\langle W\rangle_{\text{non-id}} = \langle W\rangle_\Lambda$, i.e., one obtains ideal projective measurements. On the other hand if T is nonzero, $\langle W\rangle_{\text{non-id}} = \langle W\rangle_\Lambda$ can be achieved only for specific processes Λ. For example, for the unitary U_Λ of the form

$$U_\Lambda = \frac{1}{\sqrt{d_S}} \sum_{j,k} e^{-\frac{2\pi i}{d_S} jk} |E_k^{(0)}\rangle\langle E_j^{(0)}|, \tag{A.3.8}$$

and $[H^{(0)}, H^{(\mathrm{f})}] = 0$ we have $|\langle E_m^{(\mathrm{f})} | U_\Lambda | E_l^{(0)} \rangle|^2 = \frac{1}{d_S}$. It is then simple to check that

$$\mathrm{Tr}\left(U_\Lambda \tilde{\rho}^{(0)} U_\Lambda^\dagger H^{(\mathrm{f})}\right) = \mathrm{Tr}\left(U_\Lambda \tau^{(0)} U_\Lambda^\dagger H^{(\mathrm{f})}\right) = \frac{1}{d_S} \sum_m E_m^{(\mathrm{f})}. \tag{A.3.9}$$

Consequently, one finds $\langle W\rangle_{\text{non-id}} = \langle W\rangle_\Lambda$. The intuition behind this example is the following. The disturbances due to non-ideal UMC measurements do not generate coherences (off-diagonal elements) between states with different energies in the initial thermal state. Consequently, the unconditional state upon which the process Λ acts is diagonal in the energy eigenbasis. For any such state, the process chosen in our example results in a state whose diagonal elements (w.r.t. the energy eigenbasis) are all equal to $1/d_S$. As a result, the average energy at the end of the protocol is independent of the disturbance induced by the first measurement. We thus see that there exist processes such that $\langle W\rangle_\Lambda$ can be precisely estimated independently of the temperature of the pointer.

Let us now bound the deviation of the non-ideal work estimate $\langle W\rangle_{\text{non-id}}$ from the ideal estimate $\langle W\rangle_\Lambda$. Starting from the expression in Eq. (A.3.7), we can use Hölder's inequality $|\mathrm{Tr}(XY^\dagger)| \le \|X\|_1 \|Y\|_\infty$ to write

$$|\langle W\rangle_{\text{non-id}} - \langle W\rangle_\Lambda| \le \left\|U_\Lambda \big(\tilde{\rho}^{(0)} - \tau^{(0)}\big) U_\Lambda^\dagger\right\|_1 \|H^{(\mathrm{f})}\|_\infty = \|\tilde{\rho}^{(0)} - \tau^{(0)}\|_1 \|H^{(\mathrm{f})}\|_\infty, \tag{A.3.10}$$

where in last step we have applied the trace invariance under unitary. For details about the properties of the trace distance we refer the reader to, e.g., Ref. [73]. Inserting $\tilde{\rho}^{(0)} := \sum_n p_n^{(0)} \rho_n^{(0)}$ and $\tau^{(0)} = \sum_n p_n^{(0)} |E_n^{(0)}\rangle\langle E_n^{(0)}|$, we can further write

$$\|\tilde{\rho}^{(0)} - \tau^{(0)}\|_1 = \Big\|\sum_n p_n^{(0)} \big(\rho_n^{(0)} - |E_n^{(0)}\rangle\langle E_n^{(0)}|\big)\Big\|_1 \le \sum_n \Big\|p_n^{(0)} \big(\rho_n^{(0)} - |E_n^{(0)}\rangle\langle E_n^{(0)}|\big)\Big\|_1 = \sum_n p_n^{(0)} \Big\|\rho_n^{(0)} - |E_n^{(0)}\rangle\langle E_n^{(0)}|\Big\|_1, \tag{A.3.11}$$

where we have used the triangle inequality in the second step. Finally, we recall that the 1-norm coincides with the trace norm for the operators we consider here, and we insert from Eq. (A.1.12) before using $\sum_n p_n^{(0)} = 1$ to arrive at

$$|\langle W\rangle_{\text{non-id}} - \langle W\rangle_\Lambda| \le (1 - C_{\max}) \|H^{(\mathrm{f})}\|_\infty. \tag{A.3.12}$$

Appendix A.4: Energy Variation in Non-Ideal Work Estimation

In the scenario in which ideal projective measurements are not assumed, the change in the average energy of the system in the TPM process is also different from the work estimate. Once again, we therefore consider non-ideal minimal energy UMC measurements as described in Appendix A.1. To express the work estimate in this case, let us first define the difference between the energy $\Delta E_{\text{non-id}}$ at the beginning and at the end of the TPM process

$$\Delta E_{\text{non-id}} = \sum_k p_k^{(\mathrm{f})} E_k^{(\mathrm{f})} - \sum_n p_n^{(0)} E_n^{(0)}, \tag{A.4.1}$$

where $p_k^{(\mathrm{f})}$ is the probability to find the post-measurement system after the second measurement in the eigenstate $|E_k^{(\mathrm{f})}\rangle$ of $H^{(\mathrm{f})}$. This probability can be expressed as

$$p_k^{(\mathrm{f})} = \sum_{m,n} q_{kk|m}^{(\mathrm{f})} p(m|n)\, p_n^{(0)}, \tag{A.4.2}$$

where $q_{kk|m}^{(\mathrm{f})}$ is the conditional probability to find the system in the final energy eigenstate $|E_k^{(\mathrm{f})}\rangle$ given the measurement result m. The conditional probability $p(m|n) = \langle E_m^{(\mathrm{f})}|U_\Lambda\, \rho_n^{(0)}\, U_\Lambda^\dagger|E_m^{(\mathrm{f})}\rangle$ is the probability to obtain outcome m in the second measurement given that the result of the first measurement was n, as in Eq. (A.2.8). Collecting all these expressions, we can rewrite the average energy difference between the initial system state and the system state after the non-ideal TPM scheme as

$$\Delta E_{\text{non-id}} = \sum_{m,n,k} q_{kk|m}^{(\mathrm{f})} p(m|n)\, p_n^{(0)} \left(E_k^{(\mathrm{f})} - E_n^{(0)}\right) = \sum_{m,n} p_n^{(0)} \sum_{k,l} q_{ll|n}^{(0)} q_{kk|m}^{(\mathrm{f})} |\langle E_m^{(\mathrm{f})}|U_\Lambda|E_l^{(0)}\rangle|^2 \left(E_k^{(\mathrm{f})} - E_n^{(0)}\right). \tag{A.4.3}$$

We then split this expression into several sums, where the first collects all terms for which $n = l$ and $m = k$, i.e.,

$$\begin{aligned}\Delta E_{\text{non-id}} &= \sum_{m,n} p_n^{(0)} \Big[q_{nn|n}^{(0)} |\langle E_m^{(\mathrm{f})}|U_\Lambda|E_n^{(0)}\rangle|^2 + \sum_{l\neq n} q_{ll|n}^{(0)} |\langle E_m^{(\mathrm{f})}|U_\Lambda|E_l^{(0)}\rangle|^2\Big]\Big[q_{mm|m}^{(\mathrm{f})}\left(E_m^{(\mathrm{f})} - E_n^{(0)}\right) + \sum_{k\neq m} q_{kk|m}^{(\mathrm{f})}\left(E_k^{(\mathrm{f})} - E_n^{(0)}\right)\Big] \\ &= C_{\max}^2 \langle W\rangle_\Lambda + C_{\max} \sum_{m,n} p_n^{(0)} \Big[\sum_{k\neq m} q_{kk|m}^{(\mathrm{f})} p_{n\to m}\left(E_k^{(\mathrm{f})} - E_n^{(0)}\right) + \sum_{l\neq n} q_{ll|n}^{(0)} p_{l\to m}\left(E_m^{(\mathrm{f})} - E_n^{(0)}\right)\Big] \\ &\quad + \sum_{\substack{m,n\\ l\neq n\\ k\neq m}} p_n^{(0)}\, q_{ll|n}^{(0)}\, p_{l\to m}\, q_{kk|m}^{(\mathrm{f})} \left(E_k^{(\mathrm{f})} - E_n^{(0)}\right).\end{aligned} \tag{A.4.4}$$

where we have used the shorthand $p_{l\to m} = |\langle E_m^{(\mathrm{f})}|U_\Lambda|E_l^{(0)}\rangle|^2$ and we note that $q_{nn|n}^{(0)} = q_{mm|m}^{(\mathrm{f})} = C_{\max}$ for all n and m, since we are considering UMC measurements. For ideal projective measurements, $C_{\max} \to 1$ and consequently $q_{ll|n}^{(0)} = \delta_{l,n}$, and therefore one can notice that $\Delta E_{\text{non-id}}(C_{\max} \to 1) = \langle W\rangle_\Lambda$.

Appendix A.5: Jarzynski Equality for Non-Ideal Projective Measurements

A.5.a. Characteristic Function and Jarzynski Equality

In [21] it was shown that by taking the Fourier transform of the work probability distribution $P(W)$ one can define a following characteristic function, which we parametrise by u

$$G(u) = \int \mathrm{d}W P(W) \exp(iuW) = \sum_{n,m} \exp\left(iu(E_m^{(\mathrm{f})} - E_n^{(0)})\right) p(n,m). \tag{A.5.1}$$

We can calculate this function explicitly for the non-ideal projective measurement by substituting for the probabilities $p(n,m)$ from Eq. (A.2.10):

$$G(u) = \sum_{m,n} \sum_l p_n^{(0)} q_{ll|n}^{(0)} |\langle E_m^{(\mathrm{f})}|U_\Lambda|E_l^{(0)}\rangle|^2 \tag{A.5.2}$$

$$= \sum_m \langle E_m^{(\mathrm{f})}|\exp(iuH^{(\mathrm{f})}) \sum_{n,l} \exp(-iuE_n^{(0)}) p_n^{(0)} q_{ll|n}^{(0)} U_\Lambda|E_l^{(0)}\rangle \langle E_l^{(0)}|U_\Lambda^\dagger|E_m^{(\mathrm{f})}\rangle \tag{A.5.3}$$

$$= \sum_m \langle E_m^{(\mathrm{f})}|\exp(iuH^{(\mathrm{f})}) \sum_n \exp(-iuE_n^{(0)}) p_n^{(0)} U_\Lambda \rho_n^{(0)} U_\Lambda^\dagger|E_m^{(\mathrm{f})}\rangle, \tag{A.5.4}$$

where $\sum_l q^{(0)}_{ll|n} \,|\, E^{(0)}_l \,\rangle\langle\, E^{(0)}_l \,| = \rho^{(0)}_n$ is the post measurement state of the pointer that indicates outcome n with probability $p^{(0)}_n = \exp\left(-\beta E^{(0)}_n\right)/\mathcal{Z}^{(0)}$. Here, we have again restricted our analysis to minimal energy UMC measurements as described in Appendix A.1.

Let us then write

$$\sigma(u) = \sum_n \exp\left(-iuE^{(0)}_n\right) p^{(0)}_n \rho^{(0)}_n = \frac{1}{\mathcal{Z}^{(0)}} \sum_n \exp\left(-(iu+\beta)E^{(0)}_n\right)\rho^{(0)}_n, \tag{A.5.5}$$

which we then use to obtain the characteristic function

$$G(u) = \mathrm{Tr}\left(\exp\left(iuH^{(\mathrm{f})}\right) U_\Lambda \sigma(u) U^\dagger_\Lambda\right). \tag{A.5.6}$$

To calculate the work average $\langle \exp(-\beta W)\rangle$, we further calculate $G(u = i\beta)$

$$\langle \exp(-\beta W)\rangle = G(u = i\beta) = \mathrm{Tr}\left[\exp\left(-\beta H^{(\mathrm{f})}\right) U_\Lambda \sigma(i\beta) U^\dagger_\Lambda\right], \tag{A.5.7}$$

where

$$\sigma(i\beta) = \frac{1}{\mathcal{Z}^{(0)}} \sum_n \exp\left(-(-\beta+\beta)E^{(0)}_n\right) \sum_l q^{(0)}_{ll|n} |E^{(0)}_l\rangle\langle E^{(0)}_l| = \frac{1}{\mathcal{Z}^{(0)}} \sum_l \left(\sum_n q^{(0)}_{ll|n}\right) |E^{(0)}_l\rangle\langle E^{(0)}_l|. \tag{A.5.8}$$

Consequently, Jarzynski's equality is satisfied if $\sum_n q^{(0)}_{ll|n} = 1$, i.e.,

$$\langle \exp(-\beta W)\rangle = \exp\left(-\beta \Delta F\right), \tag{A.5.9}$$

where ΔF is the free energy $\Delta F = \frac{1}{\mathcal{Z}^{(0)}} \mathrm{Tr}\left(\exp(-\beta H^{(\mathrm{f})})\right) = \frac{\mathcal{Z}^{(\mathrm{f})}}{\mathcal{Z}^{(0)}}$. The condition $\sum_n q^{(0)}_{ll|n} = 1$ is not generally met for all unbiased measurements, and in particular not by any minimal energy UMC measurements beyond dimension[6] $d_S = 2$. However, minimally invasive UMC measurements discussed in Appendix A.1.d satisfy exactly the desired condition, thus allowing to satisfy the Jarzynski equality, while providing the correct (unbiased) measurement statistics and achieving maximal correlation $C_{\max}$ between the pointer outcomes and post-measurement system states.

Appendix A.6: Crook's Relation in the Presence of Non-Ideal Measurements

A.6.a. Backward Process for Non-Ideal TPM Scheme

Crook's theorem [60] quantifies the relation between the probability of observing a work value during a realisation of the two projective measurement scheme for a given process Λ with the probability of observing the same amount of work for the time-reversed process $\tilde{\Lambda}$. The time-reverse is defined in an operational sense, meaning that the sequence of external operations (driving, measurements and so forth) used to bring the system out-of-equilibrium during the original process is inverted in the time-reversed process. A prerequisite for obtaining Crook's relation (as well as the Jarzynski equalit [59]), is that both the forward and backward processes start with the system in equilibrium at some given inverse temperature β. Therefore, the first step in studying whether Crook's relation holds in the non-ideal projective measurement setting (more specifically, restrict our analysis to minimal energy UMC measurements described in Appendix A.1), is to define a meaningful time-reversed (backward) process. This is achieved in three steps.

1. We start with a system with Hamiltonian $H^{(\mathrm{f})} = \sum_m E^{(\mathrm{f})}_m \,|\, E^{(\mathrm{f})}_m \,\rangle\langle\, E^{(\mathrm{f})}_m \,|$ that is in equilibrium at inverse temperature β. The initial state of the backward process, which is to be transformed according to

$$\left(\tau^{(\mathrm{f})}, H^{(\mathrm{f})}\right) \xrightarrow{\tilde{\Lambda}} \left(\rho^{(0)}_{\mathrm{B}}, H^{(0)}\right), \tag{A.6.1}$$

reads $\tau^{(\mathrm{f})} = e^{-\beta H^{(\mathrm{f})}}/\mathcal{Z}^{(\mathrm{f})} = \sum_m p^{(\mathrm{f})}_m \,|\, E^{(\mathrm{f})}_m \,\rangle\langle\, E^{(\mathrm{f})}_m \,|$. Prior to this transformation, the first non-ideal measurement is performed in the eigenbasis $\{|\, E^{(\mathrm{f})}_m \,\rangle\}_m$ of $H^{(\mathrm{f})}$. Given that outcome m is obtained, which occurs with probability $p^{(\mathrm{f})}_m$, the post-measurement state reads

$$\rho^{(\mathrm{f})}_m = \sum_k q^{(\mathrm{f})}_{kk|m} \,|\, E^{(\mathrm{f})}_k \,\rangle\langle\, E^{(\mathrm{f})}_k \,|. \tag{A.6.2}$$

[6] For the special case $d_S = 2$, the unbiasedness condition already leaves no choice but to make the matrix $\tilde{\pi}$ a Latin square, and hence the measurement can be a minimal energy UMC measurement that is also minimally invasive.

2. The second step of the backward process consists of driving the system according to the time-reversed protocol parameterized by $\{\tilde{\lambda}_t = \lambda_{t_f - t}; 0 \le t \le t_f\}$, where $\{\lambda_t; 0 \le t \le t_f\}$ is a parameterizsation of the forward process. In the time-reversed protocol, the system thus evolves according to

$$\rho_B^{(0)} = U_{\tilde{\Lambda}} \rho_B^{(f)} U_{\tilde{\Lambda}}^\dagger, \qquad U_{\tilde{\Lambda}}(t_f, 0) = \mathcal{T}_+ \exp\left(-i \int_0^{t_f} H(\tilde{\lambda}_t)\, dt\right), \tag{A.6.3}$$

where $\rho_B^{(f)} = \sum_n p_n^{(f)} \rho_n^{(f)}$ is the average post-measurement state after the first measurement in the backward process.

3. The last step consists of the second non-ideal measurement in the eigenbasis $\{|E_n^{(0)}\rangle\}_n$ of $H^{(0)}$. We are then interested in determining the joint probability $P_B(m,n)$ to obtain outcome m in the first and outcome n in the second non-ideal measurement, before and after the backward process, respectively. Given outcome m in the first measurement, the probability for the system to be in an energy eigenstate $|E_k^{(f)}\rangle$ is $q_{kk|m}^{(f)}$, and the probability for the backward process to further map the system from the state $|E_k^{(f)}\rangle$ to $|E_n^{(0)}\rangle$ is $\tilde{p}_{k\to n} = |\langle E_n^{(0)}|U_{\tilde{\Lambda}}|E_k^{(f)}\rangle|^2$. Consequently, we obtain the joint probability

$$P_B(n,m) = \sum_k \tilde{p}_{k\to n}\, q_{kk|m}^{(f)}\, p_m^{(f)}. \tag{A.6.4}$$

The micro-reversibility principle for non-autonomous systems implies $U_{\tilde{\Lambda}} = \Theta U_\Lambda^\dagger \Theta^\dagger$ [21], where Θ is the anti-unitary time-reversal operator satisfying $\Theta i = -i\Theta$ and $\Theta\Theta^\dagger = \Theta^\dagger\Theta = \mathbb{1}$. Micro-reversibility, together with a time-reversal symmetric Hamiltonian, $\Theta H^{(k)}\Theta^\dagger = H^{(k)}$ for $k = 0, f$, leads to the relationship $|\langle E_n^{(0)}|U_{\tilde{\Lambda}}|E_k^{(f)}\rangle|^2 = |\langle E_k^{(f)}|U_\Lambda|E_n^{(0)}\rangle|^2$, that is $\tilde{p}_{k\to n} = p_{n\to k}$.

For non-ideal projective measurements, Crook's relation can be satisfied if we make some changes to the probability distributions, as we shall explain now. To begin, let us consider the work probabilities of the forward and backward process, respectively, i.e.,

$$P_F(W) = \sum_{m,n} P_F(n,m)\delta\left((E_m^{(f)} - E_n^{(0)}) - W\right), \tag{A.6.5}$$

$$P_B(-W) = \sum_{m,n} P_B(n,m)\delta\left((E_n^{(0)} - E_m^{(f)}) + W\right), \tag{A.6.6}$$

where $P_F(n,m)$ is given in Eq. (A.2.10). We then assume that the measurement apparatus in the forward and backward process operates in the same way. More specifically, this means that the pointer is prepared in the same initial state and the same unitary U_{meas} is used to couple system and pointer. In this case, we have $q_{mm|n}^{(0)} = q_{mm|n}^{(f)} \equiv q_{mm|n}$. As in Ref. [74], we write the ratio between the joint probabilities of a given transition between $E_n^{(0)}$ to $E_m^{(f)}$ in the forward and backward process as

$$e^{-\sigma(n,m)} := \frac{P_B(n,m)}{P_F(n,m)}. \tag{A.6.7}$$

The average of the quantity $\sigma(n,m)$ defined by this ratio can be seen to be precisely the relative entropy between the probability of the forwards and backwards processes

$$\langle\sigma\rangle = \sum_{m,n} P_F(m,n)\sigma(m,n) = \sum_{m,n} P_F(m,n)\log\left(\frac{P_F(m,n)}{P_B(m,n)}\right) = D(P_F\|P_B), \tag{A.6.8}$$

where the relative entropy of two random variables Q and P is defined $D(P\|Q) := \sum_x P(x)\log\left(\frac{P(x)}{Q(x)}\right)$.

In order to express the relation between performing or extracting the same amount of work in the forward and backward process, one can substitute (A.6.7) into (A.6.6)

$$P_B(-W) = \sum_{m,n} e^{-\sigma(n,m)} P_F(n,m)\,\delta\left((E_n^{(0)} - E_m^{(f)}) + W\right). \tag{A.6.9}$$

It is straightforward to see that

$$\exp\left(-\sigma(n,m)\right) = \frac{p_m^{(f)}}{p_n^{(0)}}\frac{P_B(n|m)}{P_F(m|n)} = \exp\left(-\beta(E_m^{(f)} - E_n^{(0)} - \Delta F)\right)\frac{P_B(n|m)}{P_F(m|n)} \tag{A.6.10}$$

where $\Delta F = \beta^{-1}\log(\mathcal{Z}^{(0)}/\mathcal{Z}^{(f)})$ is the equilibrium free energy difference, and the conditional probability to find the state with energy n (m) after the backward (forward) process if initially it was m (n) is $P_B(n|m) = \sum_k p_{k\to n} q_{kk|m}$ ($P_F(m|n) = \sum_l p_{l\to m} q_{ll|n}$). Therefore, a Crook's-like relation for non-ideal projective measurements can be written as

$$P_B(-W) = e^{-\beta(W-\Delta F)}\tilde{P}_F(W), \tag{A.6.11}$$

with

$$\tilde{P}_F(W) = \sum_{m,n} \exp\left(-\gamma(m,n)\right) P_F(m,n)\delta\left((E_m^{(f)} - E_n^{(0)}) - W\right), \tag{A.6.12}$$

where $\gamma(m,n) = \log\left(P_F(m|n)/P_B(n|m)\right)$. If ideal projective measurements are assumed in the estimation process (in other words, if an infinite amount of resources is available) no disturbance is created by the measurements on the system in the forward and backward processes, which implies that $q_{mm'|n} = \delta_{m,m'}\delta_{m,n}$, and $\gamma(m,n) = 0$. Therefore Eq. (A.6.11) results in the well known Crook's relation

$$P_B(-W) = e^{-\beta(W-\Delta F)}P_F(W). \tag{A.6.13}$$

A.6.b. Irreversibility and Dissipation

Consider a thermal state $\tau^{(0)}(\beta)$ of a system with Hamiltonian $H^{(0)}$ at time t_0 that is driven out of equilibrium by means of the unitary

$$U_\Lambda(t_f, 0) = \mathcal{T}_+ \exp\left(-i\int_0^{t_f} H(\lambda_t)\,\mathrm{d}t\right). \tag{A.6.14}$$

At time t_f, the system is in the (out-of-equilibrium) state $\rho^{(f)} = U_\Lambda \tau^{(0)} U_\Lambda^\dagger$, and the Hamiltonian of the system is $H^{(f)}$. Then the system is coupled again to a thermal bath at temperature $T = 1/\beta$, and left to thermalise to the equilibrium state $\tau^{(f)}(\beta)$.

The work dissipated in the driving process, defined as $\langle W\rangle_\Lambda - \Delta F$, is the extra amount of energy that is transferred to the bath in the final thermalisation step, leading to entropy production. This energy cannot be reversibly recovered by reversing the protocol. A closed-form expression for this amount of work presented in Ref. [23] is

$$\langle W\rangle_\Lambda - \Delta F = k_B T D(\rho_F(t)\|\Theta^\dagger\rho_B(t_f - t)\Theta), \tag{A.6.15}$$

where $D(\rho_F(t)\|\Theta^\dagger\rho_B(t_f-t)\Theta)$ is the relative entropy between the state $\rho_F(t) = U_\Lambda(t,0)\tau^{(0)}U_\Lambda^\dagger(t,0)$ out of equilibrium at time t and $\rho_B(t_f-t) = U_{\tilde{\Lambda}}(t_f-t,0)\tau^{(f)}U_{\tilde{\Lambda}}^\dagger(t_f-t,0)$ is the state of the system in the backward process. Here $U_{\tilde{\Lambda}}(t_f-t,0)$ is the unitary evolution generated by the time-reversed protocol $\tilde{\Lambda}$. If the process is reversible, $\rho_F(t) = \Theta^\dagger\rho_B(t_f-t)\Theta$ for any $0 \le t \le t_f$, and there is no work being dissipated in the process. In particular, the equilibrium state is reached at time t_f, that is, $\rho^{(f)} = \tau^{(f)}$.

Now, consider non-ideal projective measurements used for work estimation, more specifically, minimal energy UMC measurements described in Appendix A.1. Here, we see that more energy is dissipated in the process because some entropy is produced during the measurement process.

Proposition A.6.1. *For work estimation based on non-ideal minimal energy UMC measurements measurements, the work dissipated in the driving process is*

$$\langle W\rangle_{\text{non-id}} - \Delta F = k_B T\left[D(\tilde{\rho}^{(f)}\|\tau^{(f)}) + \Delta S_0\right], \tag{A.6.16}$$

where $\tilde{\rho}^{(f)} = U_\Lambda\left(\sum_n p_n^{(0)}\sum_l q_{ll|n}^{(0)}|E_l^{(0)}\rangle\langle E_l^{(0)}|\right)U_\Lambda^\dagger$ *and* $\Delta S_0 = S(\tilde{\rho}^{(0)}) - S(\tau^{(0)})$ *is the entropy change in the system due to the first non-ideal measurement.*

Proof. For the TPM scheme with non-ideal measurements, the work estimate is as in Eq. (A.3.2), i.e.,

$$\langle W\rangle_{\text{non-id}} = \int dW P(W) W = \sum_{m,n} p_n^{(0)}\sum_l q_{ll|n}^{(0)} p_{l\to m}\left(E_m^{(f)} - E_n^{(0)}\right), \tag{A.6.17}$$

where $p_{l\to m} = |\langle E_m^{(f)}|U_\Lambda|E_l^{(0)}\rangle|^2$. We then note that the thermal states with respect to the initial and final Hamiltonians are

$$\tau^{(0)} = \exp(-\beta H^{(0)})/\mathcal{Z}^{(0)} = \sum_n p_n^{(0)}|E_n^{(0)}\rangle\langle E_n^{(0)}|, \tag{A.6.18a}$$

$$\tau^{(f)} = \exp(-\beta H^{(f)})/\mathcal{Z}^{(f)} = \sum_m p_m^{(f)}|E_m^{(f)}\rangle\langle E_m^{(f)}|, \tag{A.6.18b}$$

where the partition functions are $\mathcal{Z}^{(0)} = \mathrm{Tr}\left(\exp(-\beta H^{(0)})\right)$ and $\mathcal{Z}^{(f)} = \mathrm{Tr}\left(\exp(-\beta H^{(f)})\right)$, and the probabilities for the individual energy eigenstates are $p_n^{(0)} = \exp(-\beta E_n^{(0)})/\mathcal{Z}^{(0)}$ and $p_m^{(f)} = \exp(-\beta E_m^{(f)})/\mathcal{Z}^{(f)}$. The logarithms of the probabilities above are $\log p_n^{(0)} = -\beta E_n^{(0)} - \log\mathcal{Z}^{(0)}$ and $\log p_m^{(f)} = -\beta E_m^{(f)} - \log\mathcal{Z}^{(f)}$. With this, we can rewrite the factor $(E_m^{(f)} - E_n^{(0)})$ in $\langle W\rangle_{\text{non-id}}$ and obtain

$$\beta\langle W\rangle_{\text{non-id}} = \sum_{m,n} p_n^{(0)} \sum_l p_{l\to m}\, q_{ll|n}^{(0)} \left((-\log p_m^{(f)} - \log\mathcal{Z}^{(f)}) - (-\log p_n^{(0)} - \log\mathcal{Z}^{(0)})\right). \tag{A.6.19}$$

We then note that $\sum_m\sum_l p_{l\to m}\, q_{ll|n}^{(0)} = 1$ and identify the free energy $\Delta F = \frac{1}{\beta}(\log\mathcal{Z}^{(0)} - \log\mathcal{Z}^{(f)})$, as well as the initial thermal state entropy $S(\tau^{(0)}) = -\sum_n p_n^{(0)}\log p_n^{(0)}$ to write

$$\begin{aligned}\beta\left(\langle W\rangle_{\text{non-id}} - \Delta F\right) &= -S(\tau^{(0)}) - \sum_{m,n} p_n^{(0)}\sum_l p_{l\to m}\, q_{ll|n}^{(0)}\log p_m^{(f)} = -S(\tau^{(0)}) - \mathrm{Tr}\left(U_\Lambda\left(\sum_n p_n^{(0)}\sum_l q_{ll|n}^{(0)}|E_l^0\rangle\langle E_l^0|\right)U_\Lambda^\dagger\log\tau^{(f)}\right)\\ &= -S(\tau^{(0)}) - \mathrm{Tr}\left(U_\Lambda\tilde{\rho}^{(0)}U_\Lambda^\dagger\,\log\tau^{(f)}\right),\end{aligned} \tag{A.6.20}$$

where we have recognised the conditional post-measurement state $\rho_n^{(0)} = \sum_l q_{ll|n}^{(0)}|E_l^0\rangle\langle E_l^0|$, and we have denoted the unconditional post-measurement state as $\tilde{\rho}^{(0)} = \sum_n p_n^{(0)}\rho_n^{(0)}$. We can then add and subtract the entropy of $\tilde{\rho}^{(f)} = U_\Lambda\tilde{\rho}^{(0)}U_\Lambda^\dagger$ and use the invariance of the von Neumann entropy under unitaries, in particular, $S(\tilde{\rho}^{(f)}) = S(\tilde{\rho}^{(0)})$ to arrive at

$$\beta\left(\langle W\rangle_{\text{non-id}} - \Delta F\right) = D(\tilde{\rho}^{(f)}\|\tau^{(f)}) + S(\tilde{\rho}^{(0)}) - S(\tau^{(0)}) \;=\; D(\tilde{\rho}^{(f)}\|\tau^{(f)}) + \Delta S_0. \tag{A.6.21}$$

□

This result can be seen as a version of Eq. (A.6.15) applicable to non-ideal projective measurement when taking $t = t_f$, which expresses the amount of irreversible work in the process Λ calculated by means of the non-ideal TPM scheme. ΔS_0 represents the entropy change in the system due to the initial non-ideal measurement process, which may be either positive or negative in general. However, for unital measurements, the entropy of the pointer does not change [64] and, as a consequence, the second law implies $\Delta S_0 \geq 0$, which can now be interpreted as the entropy production in the measurement process.

Proposition A.6.2. *Consider a system of dimension d_S, and an interaction between system and pointer such that their correlations are maximal, $C(\tilde{\rho}_{SP}) = (C_{max})$, for instance, any UMC measurement. Then the entropy produced in the measurement process satisfies*

$$\Delta S_0 \leq (1 - C_{max})\log(d_S - 1) + H_2(C_{max}), \tag{A.6.22}$$

where $H_2(x) = -x\log x - (1-x)\log(1-x)$ is the binary entropy of the random variable x with $0 \leq x \leq 1$.

Proof. This inequality comes directly from the Fannes-Audenaert inequality in Refs. [75, 76]

$$S(\tilde{\rho}^{(0)}) - S(\tau_0) \leq D(\tilde{\rho}^{0}, \tau_0)\log(d_S - 1) + H_2(D(\tilde{\rho}^{(0)}, \tau_0^{(0)})), \tag{A.6.23}$$

where $D(\tilde{\rho}^{(0)}, \tau^{(0)}) = \frac{1}{2}\|\tilde{\rho}^{(0)} - \tau^{(0)}\|_1$ is the trace distance. To obtain Eq. (A.6.22), the trace distance must be $D(\tilde{\rho}^{(0)}, \tau^{(0)}) \leq 1$. Since the trace distance is convex and monotonic:

$$D(\tilde{\rho}^{(0)}, \tau^{(0)}) = \frac{1}{2}\|\tilde{\rho}^{(0)} - \tau^{(0)}\|_1 \;\leq\; \frac{1}{2}\sum_n p_n^{(0)}\,\|\rho_n^{(0)} - |E_n^{(0)}\rangle\langle E_n^{(0)}|\|_1 \;=\; 1 - C_{max}, \tag{A.6.24}$$

where the last equality comes from Eq. (A.1.12). □

Proposition A.6.3. *Assuming non-ideal (minimal energy UMC) projective measurements for work estimation, the work dissipated in the driving process can be written as*

$$\langle W\rangle_{\text{non-id}} - \Delta F = k_{\text{B}}T\left[\Delta S_0 + \Delta D_{\text{f}} + D(\rho_{\text{F}}(t)\|\Theta^{\dagger}\rho_{\text{B}}(t_{\text{f}}-t)\Theta)\right], \tag{A.6.25}$$

where $\rho_{\text{F}}(t) = U_{\Lambda}(t,0)\tilde{\rho}^{(\text{f})}U_{\Lambda}^{\dagger}(t,0)$ *is the system state at intermediate time* $0 \le t \le t_{\text{f}}$ *in the forward process,* $\rho_{\text{B}}(t_{\text{f}}-t) = U_{\tilde{\Lambda}}(t_{\text{f}}-t,0)\rho_{\text{B}}^{(\text{f})}U_{\tilde{\Lambda}}^{\dagger}(t_{\text{f}}-t,0)$ *is the (inverted) state of the system at the same instant of time in the backward process, and we introduced the correction term*

$$\Delta D_{\text{f}} = D(\tilde{\rho}^{(\text{f})}\|\tau^{(\text{f})}) - D(\tilde{\rho}^{(\text{f})}\|\rho_{\text{B}}^{(\text{f})}) = \text{Tr}[\tilde{\rho}^{(\text{f})}\left(\log\rho_{\text{B}}^{(\text{f})} - \log\tau^{(\text{f})}\right)]. \tag{A.6.26}$$

Proof. We start from Eq. (A.6.16), adding and subtracting the quantity $D(\tilde{\rho}^{(\text{f})}\|\rho_{\text{B}}^{(\text{f})})$ to arrive at

$$\langle W\rangle_{\text{non-id}} - \Delta F = k_{\text{B}}T\left[\Delta S_0 + \Delta D_{\text{f}} + D(\tilde{\rho}^{(\text{f})}\|\rho_{\text{B}}^{(\text{f})})\right]. \tag{A.6.27}$$

Then we use the properties of the quantum relative entropy and the unitary evolution (A.6.14) to write

$$\begin{aligned} D(\tilde{\rho}^{(\text{f})}\|\rho_{\text{B}}^{(\text{f})}) &= D(U_{\Lambda}(t_{\text{f}},t)U_{\Lambda}(t,0)\tilde{\rho}^{(0)}U_{\Lambda}^{\dagger}(t,0)U_{\Lambda}^{\dagger}(t_{\text{f}},t)\|\rho_{\text{B}}^{(\text{f})}) \\ &= D(U_{\Lambda}(t,0)\tilde{\rho}^{(0)}U_{\Lambda}^{\dagger}(t,0)\|U_{\Lambda}^{\dagger}(t_{\text{f}},t)\rho_{\text{B}}^{(\text{f})}U_{\Lambda}(t_{\text{f}},t)) \\ &= D(U_{\Lambda}(t,0)\tilde{\rho}^{(0)}U_{\Lambda}^{\dagger}(t,0)\|\Theta^{\dagger}U_{\tilde{\Lambda}}(t_{\text{f}}-t,0)\rho_{\text{B}}^{(\text{f})}U_{\tilde{\Lambda}}^{\dagger}(t_{\text{f}}-t,0)\Theta) = D(\rho_{\text{F}}(t)\|\Theta^{\dagger}\rho_{\text{B}}(t_{\text{f}}-t)\Theta), \end{aligned} \tag{A.6.28}$$

where we have used the micro-reversibility principle for non-autonomous systems [21] in the last line , i.e., $U_{\Lambda}^{\dagger}(t_{\text{f}},t) = \Theta^{\dagger}U_{\tilde{\Lambda}}(t_{\text{f}}-t,0)\Theta$, and we have identified the expressions for $\rho_{\text{F}}(t)$ and $\rho_{\text{B}}(t_{\text{f}}-t)$. Insering Eq. (A.6.28) into Eq. (A.6.27), we directly obtain Eq. (A.6.25). □

Proposition A.6.4. *In the case of open quantum systems, Eq.* (A.6.25) *becomes the inequality*

$$\langle W\rangle_{\text{non-id}} - \Delta F \ge k_{\text{B}}T\left[\Delta S_0 + \Delta D_{\text{f}} + D(\rho_{\text{F}}(t)\|\Theta^{\dagger}\rho_{\text{B}}(t_{\text{f}}-t)\Theta)\right]. \tag{A.6.29}$$

Proof. Let us consider our system of interest as before along with a thermal bath to which it is coupled. The joint system can be considered to be closed, and Eq. (A.6.25) hence holds for the joint system, i.e.,

$$\langle W\rangle_{\text{non-id}} - \Delta F = k_{\text{B}}T\left[\Delta S_0 + \Delta D_{\text{f}} + D(\rho'_{\text{F}}(t)\|\Theta^{\dagger}\rho'_{B}(t_{\text{f}}-t)\Theta)\right] \le k_{\text{B}}T\left[\Delta S_0 + \Delta D_{\text{f}} + D(\rho_{\text{F}}(t)\|\Theta^{\dagger}\rho_{\text{B}}(t_{\text{f}}-t)\Theta)\right]. \tag{A.6.30}$$

The left-hand side is the same as before as long as work is performed by implementing the protocol Λ only involving system degrees of freedom, whereas the primed quantities on the right-hand side correspond to the global state of system and bath. Notice that assuming that non-ideal measurements are only performed on the system implies that we recover the same terms ΔS_0 and ΔD_{f}. Finally, applying monotonicity of the relative entropy under the partial trace, i.e., $S(\rho|\sigma) \ge S(\rho'\|\sigma')$ for any $\rho = \text{Tr}_{\text{bath}}[\rho']$ and $\sigma = \text{Tr}_{\text{bath}}[\sigma']$, the last inequality is obtained. □

Appendix A.7: Two-level system driven by a classical field

To illustrate the formalism presented in this work, let us consider a two-level atom driven out of equilibrium by a classical field, described by the Hamiltonian $H = H_S + H_F$, where $H_S = -\frac{E_S}{2}\sigma_z$ is the atomic Hamiltonian with $\sigma_z = |0\rangle\langle 0| - |1\rangle\langle 1|$, and the interaction Hamiltonian is $H_F = -\mathbf{D}\cdot\mathbf{E}_F$, where $\mathbf{E}_F = i\varepsilon_F\left(\boldsymbol{\epsilon}_F e^{-i\omega_F t}e^{-i\phi_0} + \boldsymbol{\epsilon}_F^{*}e^{i\omega_F t}e^{i\phi_0}\right)$ is a classical field with real-amplitude ε_F, polarization vector $\boldsymbol{\epsilon}_F$, phase ϕ_0 and angular frequency ω_F. The atomic dipole is $\mathbf{D} = d(\boldsymbol{\epsilon}_S^{*}\sigma_+ + \boldsymbol{\epsilon}_S\sigma_-)$, with d being the dipole strength, and the vector $\boldsymbol{\epsilon}_S$ describes the atomic transition polarization given the energy transition operators $\sigma_+ = |1\rangle\langle 0|$ and $\sigma_- = |0\rangle\langle 1|$. After applying the Rotating Wave Approximation (RWA), the Hamiltonian describes a Rabi oscillation with angular frequency $\tilde{\Omega}_F$, i.e.,

$$H \approx \frac{\tilde{\Omega}_F}{2}\boldsymbol{\sigma}\cdot\mathbf{n}, \tag{A.7.1}$$

where $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ and $\mathbf{n} = (\Delta_F\mathbf{u}_z + \Omega_F\mathbf{u}_y)/\tilde{\Omega}_F$ with corresponding unit vectors $(\mathbf{u}_x, \mathbf{u}_y, \mathbf{u}_z)$, and we use units where $\hbar = 1$. The angular frequency is defined in terms of atom and field variables as

$$\tilde{\Omega}_F^2 = \Delta_F^2 + \Omega_F^2, \tag{A.7.2}$$

where $\Delta_F = E_S - \omega_F$ is the difference between the angular frequency of the field and the energy gap of the atom (recall that $\hbar = 1$), and $\Omega_F = 2d\varepsilon_F\left(\boldsymbol{\epsilon}_F \cdot \boldsymbol{\epsilon}_S^*\right)e^{i\phi_0}$ is the classical Rabi frequency of the atom-field interaction. The transformation generated by the Hamiltonian in Eq. (A.7.1) is a unitary of the form

$$U(\theta) = \exp\left(-i\frac{\theta}{2}\boldsymbol{\sigma}\cdot\mathbf{n}\right) = \cos\left(\frac{\theta}{2}\right)\mathbb{I} - i\sin\left(\frac{\theta}{2}\right)\boldsymbol{\sigma}\cdot\mathbf{n}, \tag{A.7.3}$$

for $\theta = \tilde{\Omega}_F t$, and t is the duration of the transformation. For the purpose of illustration, let us restrict our further analysis to the resonant case, where $\Delta_F \approx 0$. This implies $\mathbf{n} = \mathbf{u}_y$ and hence that the transformation $U(\theta)$ is a rotation around the y axis and $\tilde{\Omega}_F = \Omega_F$. Further details about this physical system, and a more complete and general scenario can be found in Ref. [77].

Let us now consider a process in which the atom is initially prepared in a thermal state at inverse temperature $\beta_S = 1/(k^{\mathrm{B}}T_S)$ with respect to the initial system Hamiltonian $H_S^{(0)} = -E_S\sigma_z/2$, i.e.,

$$\rho_S^{(0)} = \tau_S^{(0)} = \frac{\exp\left(-\beta_S H_S^{(0)}\right)}{\mathcal{Z}_S^{(0)}}. \tag{A.7.4}$$

We then consider the TPM scheme with non-ideal measurements to estimate the work that is performed on the system by the transformation $U(\theta)$, where we assume the measurements to be non-ideal but minimal energy UMC measurements (see Appendix A.1.c). After the first measurement, the interaction with the field (H_F) is instantaneously switched on at $t = 0$, and the field evolves until time $t = t_{\mathrm{f}}$ to an out-of-equilibrium state given by $\rho_S^{(\mathrm{f})}(\theta) = U(\theta)\rho_S^{(0)}U(\theta)^\dagger$, with $\theta = t_{\mathrm{f}}\Omega_F$. At the time $t = t_{\mathrm{f}}$ the interaction with the field (H_F) is instantaneously switched off, such that $H_S^{(\mathrm{f})} = H_S^{(0)}$, and the second non-ideal measurement is performed.

The work done by the field by means of $U(\theta)$ is the energy difference between the initial and final configurations,

$$\langle W\rangle_\Lambda = \mathrm{Tr}\left(\rho_S^{(\mathrm{f})}(\theta)H_S^{(\mathrm{f})}\right) - \mathrm{Tr}\left(\rho_S^{(0)}H_S^{(0)}\right) = -\frac{E_S}{2}\mathrm{Tr}\left(\sigma_z(\rho_S^{(\mathrm{f})}(\theta) - \rho_S^{(0)})\right) = E_S\sin^2\left(\tfrac{\theta}{2}\right)\tanh\left(\tfrac{\beta_S E_S}{2}\right). \tag{A.7.5}$$

However, when we estimate this work using non-ideal measurements, we obtain a different value $\langle W\rangle_{\mathrm{non-id}}$. For instance, let us consider minimal energy UMC measurements using a three-qubit pointer that is prepared in a thermal state

$$\tau(\beta_P) = \left(\frac{\exp\left(-\beta_P H_P\right)}{\mathcal{Z}_P}\right)^{\otimes 3}, \tag{A.7.6}$$

where $H_P = -\frac{E_P}{2}\sigma_z = \frac{E_P}{2}\left(|1\rangle\langle 1| - |0\rangle\langle 0|\right)$ is the Hamiltonian of each single-qubit subsystem of the pointer and $\beta_P = 1/(k_{\mathrm{B}}T_P)$. As discussed before, the maximum correlation created between system and pointer depends only on the preparation of the measurement apparatus, which in this example is the sum of the $d_P/d_S = 4$ biggest eigenvalues of the pointer, i.e.,

$$C_{\max} = \frac{\left(1 + 3\exp\left(-\beta_P E_P\right)\right)}{\mathcal{Z}_P^3}. \tag{A.7.7}$$

Following the approach developed in Sec. A.3, the non-ideal work estimate can be written as

$$\langle W\rangle_{\mathrm{non-id}} = \mathrm{Tr}\left(U(\theta)\,\tilde{\rho}_S^{(0)}\,U(\theta)^\dagger\,H_S^{(\mathrm{f})}\right) - \mathrm{Tr}\left(\rho_S^{(0)}H_S^{(0)}\right), \tag{A.7.8}$$

where the unconditional post-measurement system state is $\tilde{\rho}_S^{(0)} := \sum_n p_n^{(0)}\rho_n^{(0)}$. The conditional post-measurement system states are $\rho_n^{(0)} = \sum_l q_{ll|n}^{(0)}|l\rangle\langle l|$ and $p_0^{(0)} = 1/\mathcal{Z}_S$ and $p_1^{(0)} = \exp\left(-\beta_S E_S\right)/\mathcal{Z}_S$ and $\mathcal{Z}_S = 1 + \exp\left(-\beta_S E_S\right)$. The probability $q_{ll|n}^{(0)}$ to find the post-measurement state $\rho_n^{(0)}$ in a specific eigenstate $|l\rangle$ of the system, depends only on the amount of correlation created between system and pointer, that is,

$$q_{nn|n}^{(0)} = C_{\max} = \frac{1 + 3\exp\left(-\beta_P E_P\right)}{\mathcal{Z}_P^3} \quad \text{for} \quad n = 0, 1, \tag{A.7.9}$$

$$q_{00|1}^{(0)} = q_{11|0}^{(0)} = 1 - C_{\max} = \frac{\exp\left(-3\beta_P E_P\right) + 3\exp\left(-2\beta_P E_P\right)}{\mathcal{Z}_P^3}. \tag{A.7.10}$$

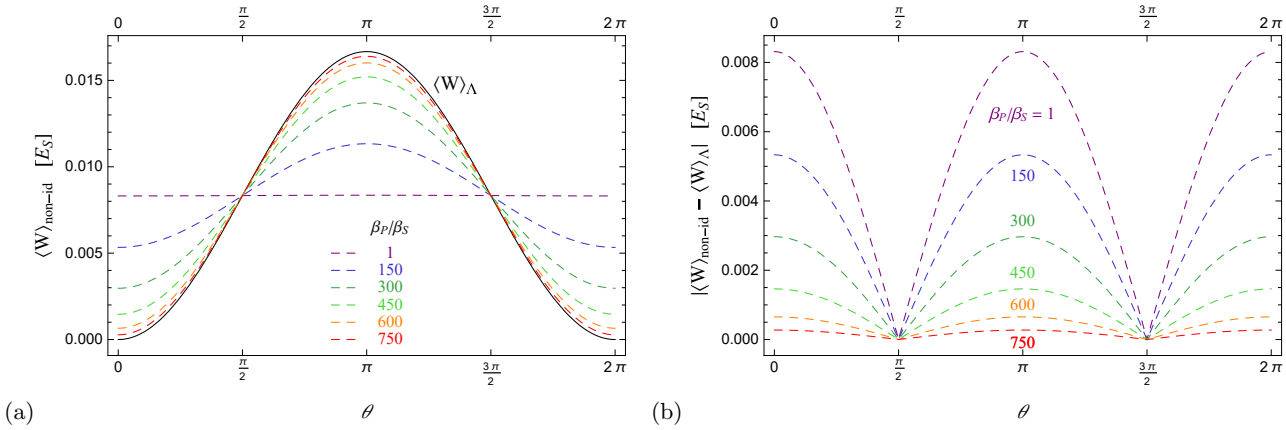

Figure A.2. The non-ideal work estimate $\langle W\rangle_{\text{non-id}}$ and its deviation from the ideal work estimate $\langle W\rangle_\Lambda$ are shown in (a) and (b), respectively, as functions of $\theta = \Omega_F t \in [0, 2\pi]$, for selected initial temperatures of the pointer, represented by different ratios of the system and pointer temperature, i.e., $\beta_P/\beta_S = 1$, and from $\beta_P/\beta_S = 150$ to 750 in steps of 150. For $\theta = \pi/2$ and $\theta = 3\pi/2$ the non-ideal work estimate coincides with the work performed by the process $U(\theta)$, independently of the temperature of the pointer. For the purpose of the this illustration, the initial temperature of the system has been chosen to be room temperature $T_S = 300$ K for an energy gap in the microwave regime such that $\beta_S E_S \approx 1/30$, and $E_P = E_S/10$.

Given the initial state of the atom in Eq. (A.7.4), the unconditional post-measurement system state evolves as

$$U(\theta)\,\tilde{\rho}_S^{(0)}\,U(\theta)^\dagger = \frac{1}{\mathcal{Z}_S}\left(U(\theta)\,\rho_{n=0}^{(0)}\,U(\theta)^\dagger + \exp\left(-\beta_S E_S\right)U(\theta)\,\rho_{n=1}^{(0)}\,U(\theta)^\dagger\right). \tag{A.7.11}$$

Figure A.2 (a) shows $\langle W\rangle_{\text{non-id}}$ as a function of $\theta = \Omega_F t$ for different temperatures of the pointer, where each of the three pointer qubits is assumed to have an energy gap $E_P = E_S/10$. The system is taken to be at room temperature $T_S = 300K$ initially, with an energy gap in the microwave regime, such that $\beta_S E_S \approx 1/30$. The pointer is initially at the same temperature as the system, but the pointer can be cooled in order to obtain a better precision in the work estimation as illustrated in Fig. A.2 (b).

For $\theta = \Omega_F t = \pi/2$ and $\theta = 3\pi/2$, the non-ideal work estimate coincides with the work realised by the process $U(\theta)$ independently of the temperature of the pointer. However, for $\theta = \pi$ one notices that the *deviation* $|\langle W\rangle_\Lambda - \langle W\rangle_{\text{non-id}}|$ of the work-estimate from its ideal value $\langle W\rangle_\Lambda$ has the same order of magnitude as $\langle W\rangle_\Lambda$ for a pointer at temperature close to room temperature, $\beta_P/\beta_S = 1$, where we find that $|\langle W\rangle_\Lambda - \langle W\rangle_{\text{non-id}}|/\langle W\rangle_\Lambda = 0.49875$. Indeed, for values of θ close to integer multiples of 2π, the non-ideal estimate can even be arbitrarily far away from the ideal estimate (which vanishes at these points) in the sense that $|\langle W\rangle_\Lambda - \langle W\rangle_{\text{non-id}}|/\langle W\rangle_\Lambda$ diverges as $\theta \to 2\pi$. In other words, if the temperature of the pointer is not taken into account in the work-estimation, the imprecision of the estimate can be as bigger than the work performed or extracted by the process $U(\theta)$. In these cases $U(\theta)$ satisfies the condition in Eq. (A.3.9). At the same time, we note that, since we consider the special case of a qubit system, the minimal energy UMC measurements we consider are also minimally invasive, meaning that Jarzynski's relation is satisfied for all θ and β_P/β_S.

Let us now consider the energy spent to perform the measurements in the TPM process in the first place, which is given by $\Delta E_{\text{TPM}} := \Delta E_{\text{meas}}^{(0)} + \Delta E_{\text{meas}}^{(\text{f})}$. The contributions from the two respective measurements are

$$\Delta E_{\text{meas}}^{(0)} = \text{Tr}\Big[\big(H_S^{(0)} + \sum_{i=1,2,3} H_{P_i}\big)\big(\tilde{\rho}_{SP}^{(0)} - \rho_S^{(0)} \otimes \tau_P\big)\Big], \tag{A.7.12}$$

$$\Delta E_{\text{meas}}^{(\text{f})} = \text{Tr}\Big[\big(H_S^{(\text{f})} + \sum_{i=1,2,3} H_{P_i}\big)\big(\tilde{\rho}_{SP}^{(\text{f})} - \rho_S^{(\text{f})} \otimes \tau_P\big)\Big], \tag{A.7.13}$$

where H_{P_i} is the Hamiltonian for qubit i (for $i = 1, 2, 3$) and we have assumed that the pointer is prepared in the same initial state for both measurements. The states $\tilde{\rho}_{SP}^{(0)}$ and $\tilde{\rho}_{SP}^{(\text{f})}$ are the joint system-pointer post-measurement states after the two respective measurements, as in Eq. (A.1.14), for the initial state $\rho_S^{(0)}$ at $t = 0$, and the final state at $t = t_\text{f}$ is $\rho_S^{(\text{f})} = \rho_S(\theta)$.

In Fig. A.3 we plot ΔE_{TPM} as a function of the initial temperature of the pointer for two distinct durations of the driving protocol. We notice that there is no discernible dependence of ΔE_{TPM} on the duration of the protocol. Furthermore, we calculate the energy cost of cooling the pointer within the single-qubit refrigerator paradigm [33, 34].

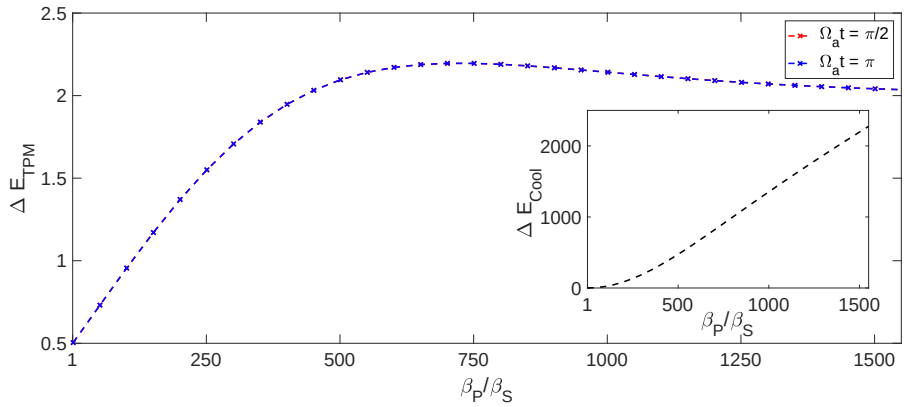

Figure A.3. The energy cost ΔE_{TPM} to perform the (non-ideal) TPM measurements is shown in units of E_S as a function of the ratio between the temperatures of the system and pointer $\beta_P/\beta_S = T_S/T_P$, where the system is assumed to be at room temperature $T_S = 300K$ initially, for two exemplary durations $\theta = \Omega_F t = \pi/2$ and $\theta = \Omega_F t = \pi$ of the driving protocol. We see that there is no discernible dependence on $\theta = \Omega_F t$. More importantly, the inset plot shows the energy cost ΔE_{Cool} (in units of E_S) for cooling the pointer from $1/\beta_S$ to $1/\beta_P \leq 1/\beta_S$ as a function of the ratio β_P/β_S.

For a refrigerator with an energy gap E_{F}, the energy needed to cool the pointer from a temperature $1/\beta_S$ to the lower temperature $1/\beta_P$ is at least

$$\Delta E_{\mathrm{Cool}} = N(E_{\mathrm{F}} - 1)\left(\frac{1}{e^{-\beta_S E_{\mathrm{F}}} + 1} - \frac{1}{e^{-\beta_S E_P} + 1}\right), \tag{A.7.14}$$

where N is the number of qubits to be cooled, and $E_{\mathrm{F}} = E_P \beta_P/\beta_S$. In the example we consider $N = 3$ for each measurement and $E_P = E_S/10$. From Fig. A.2, we see that for $\theta = \pi$ and $\beta_P/\beta_S \geq 750$, the deviation of the non-ideal work-estimate from the ideal one is nearly half as big as $\langle W\rangle_\Lambda$, but $|\langle W\rangle_\Lambda - \langle W\rangle_{\mathrm{non-id}}| < 0.01 E_S$. However, as illustrated in Fig. A.3, the energy cost for cooling the pointer such that $\beta_P/\beta_S = 750$ is more than two times E_S. In other words, the energy cost of cooling the pointer to the required temperature can outweigh $\langle W\rangle_\Lambda$ by orders of magnitude. That is, the cost of estimating the work done by a physical process can by far exceed the amount of work that is done or consumed during the process.

Curriculum Vitae — Nicolai Friis

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Academic Career

Since **2018** **Project Leader**: FWF Stand-Alone Project *"Resources for flexible quantum information processing"* at Institute for Quantum Optics & Quantum Information Vienna

Since **2017** **Senior Postdoc**: Institute for Quantum Optics & Quantum Information Vienna in the group of Dr. Marcus Huber

2015 – 2017 **Postdoc** (Universitätsassistent): University of Innsbruck, Institute for Theoretical Physics, in the group of Univ.-Prof. Dr. Hans J. Briegel

2013 – 2014 **Postdoc**: Institute for Quantum Optics & Quantum Information (IQOQI) Innbruck, Austrian Academy of Sciences, in the group of Univ.-Prof. Dr. Hans J. Briegel

Education

2010 – 2013 **PhD**: University of Nottingham, School of Mathematical Sciences,
Thesis: Cavity mode entanglement in relativistic quantum information
Supervisors: Prof. Dr. Ivette Fuentes and Prof. Dr. Gerardo Adesso
Viva voce examination: 4th November 2013 (thesis accepted without corrections)

2010 – 2010 **Lecturer** (part time): University of Vienna, Department of Physics

2004 – 2010 **Undergraduate degree**: University of Vienna, Physics Diploma (Mag. rer. nat.)
Thesis: Relativistic Effects in Quantum Entanglement
Supervisor: Ao. Univ.-Prof. Dr. Reinhold A. Bertlmann
Graduation with *distinction* ("mit Auszeichnung"), 24th February 2010

Grants & Awards

2018 *FWF Stand-Alone Project "Resources for flexible quantum information processing"*

2018 *Entropy Young Investigator Award 2018*

2014 *Václav Votruba Prize* 2014 for best PhD thesis in theoretical physics

2012 University of Nottingham Graduate School *Universitas 21 Prize Scholarship*

2007 – 2009 Award of three *Performance Scholarships* of the University of Vienna

Commissions of Trust

Since **2020** **Editor of Quantum Views**: Copy-editing of perspective articles for the open--access peer-reviewed journal Quantum

Since **2016** **Deputy President**: "Verein zur Förderung des Open Access Publizierens in den Quantenwissenschaften" (legal body of the journal Quantum)

Since **2017** **International Editorial Board Member** for J. Phys. Commun. (IoP Publishing)

2016 **Invited Perspective Article** in New J. Phys. **18**, 061001 (2016)

Reviewer

since **2014** **Reviewer**: 90 verified reviews in 27 different journals (see publons profile) including: Nat. Phys.; Nat. Commun.; Phys. Rev. (Lett., X, Appl., A, D, & E); npj Quant. Inf.; Quantum; New. J. Phys.; J. Phys. A; Phys. Lett. A & B; Quant. Inf. Comput.;

2020 **IOP Trusted Reviewer Status** for 'exceptionally high level of peer review competency'

2019 **PhD Thesis Examiner** for Sebastian Kish, University of Queensland, Australia

2020 **Programme Committee Member** for Q-Turn 2020 (held online due to Covid-19)

2019 **Programme Committee Member** for YQIS, Sopot Poland, September 2019

2018 **Programme Committee Member** for YQIS Vienna, September 2018

2018 **Programme Committee Member** for Q-Turn, Florianópolis, Brazil, November 2018

Invited Talks

2021 *Entanglement in Quantum Fields* (June 2021, IWH Heidelberg, Germany + online, organizers S. Floerchinger, M. Gärttner, H. Strobel)

2018 *APCTP-KIAS workshop on Motors and Engines* (June 25-27, 2018, Korea Institute for Advanced Study (KIAS), organizers J. Yi, P. Talkner, H. Park, Y. W. Kim)

2016 Workshop *Recent Advances in Continuous-variable Quantum Information Theory* (Universitat Autònoma de Barcelona, organizers A. Winter, K. Sabapathy, M. Huber)

2015 Invited talk on the occasion of the celebration of Reinhold Bertlmann's 70th birthday (University of Vienna, organized by M. Arndt)

2014 Workshop *Quantum Mechanics Tests in Particle, Atomic, Nuclear and Complex Systems* (ECT*, Trento, Italy, organizers B. C. Hiesmayr, C. Curceanu and A. Buchleitner)

2012 *9th Vienna Central European Seminar on Particle Physics and Quantum Field Theory* (University of Vienna, organized by H. Hüffel)

2012 *Workshop on Effective Gravity in Fluids and Superfluids* (Abdus Salam International Centre for Theoretical Physics (ICTP), Trieste, organized by S. Weinfurtner)

Organization of Conference & Workshops

2017 *Organizer* of the December 2017 *Nonlocal Seminar* at IQOQI Vienna

2016 Summer School *Introductory Course on Quantum Information*, University of Innsbruck

2012 – 2013 *Organizing Committee* for the conference *RQI-N 2013* at the University of Nottingham

2012 – 2013 *Organizer* of the workshop "QFGI 2013" at the University of Nottingham

2011 *Local Organizing Committee* for the *ESF PESC Strategic Workshop* on "Signatures of Quantumness in Complex Systems" in Nottingham

Outreach and Media Coverage

Since **2010** **Conference Talks** (excl. invited): **22**

Conference Posters: **8**

Short-term Scientific Visits with Invited Seminar Talks: **21**

2021 News coverage of Nature **591**, 229–233 (2021) in e.g., standard.at (Austrian news, online version, in German); phys.org and New Scientist

News coverage of Phys. Rev. X **11**, 011046 (2021) in science.orf.at (Austrian news)

News coverage of Nature **589**, 220–224 (2021) in e.g., science.orf.at (Austrian news), standard.at (Austrian newspaper, online version); phys.org; physicsworld.com

News coverage of Quantum **4**, 376 (2020) in standard.at (Austrian news, online)

2018 News coverage of Nat. Phys. **14**, 1032 (2018) in e.g., science.orf.at (Austrian news), standard.at (Austrian newspaper, online version) and phys.org

2018 News coverage of Phys. Rev. X **8**, 021012 (2018) in e.g., science.orf.at (Austrian news), standard.at (Austrian newspaper, online version), phys.org, IEEE Spectrum

2017 News coverage of Nat. Commun. **8**, 1321 (2017) in e.g., tirol.orf.at (Austrian news), standard.at (Austrian newspaper, online version), phys.org, IEEE Spectrum

Teaching Experience

Total: 60+ ECTS as lecturer ("Lehrveranstaltungsleiter")

Lecturer at the University of Vienna:

2020 "Quantum Information" (Lecture, VO, 4 ECTS)

2019 – 2020 "Entanglement beyond qubits and the foundation of thermodynamics" (Seminar, SE, 5 ECTS, jointly w. M. Huber)

2019 SE "High-dimensional QI and thermodynamics" (5 ECTS, jointly w. M. Huber)

2018 – 2019 "Quantum Thermodynamics II" (Lecture + problem class., VO+UE, 2.5 ECTS each, jointly with M. Huber & M. Lock)

2018 "Quantum Thermodynamics I" (Lecture, VO, 2.5 ECTS, jointly w. M. Huber)

2010 "Exercises for Theoretical Physics II - Quantum Mechanics" (3 ECTS)

Lecturer at the University of Innsbruck:

2016 – 2017 Proseminar (PS) "Theoretical Quantum Information" (5 ECTS)

2016 PS "Quantentheorie" (3 groups, 4 ECTS each)

2015 – 2016 PS "Relativiy" (3 ECTS) & "Theoretical Quantum Information" (5 ECTS)

2015 PS "Quantentheorie" (2 groups, 4 ECTS each)

2014 – 2015 PS "Relativity" (3 ECTS)

Supervision

2020 – 2021 Markus Miethlinger (BSc. student, University of Vienna)

since **2019** Simon Morelli (PhD student, University of Vienna, jointly with M. Huber)

2017 – 2019 Beniamin Jablonski (MSc., University of Vienna, jointly with M. Huber)

2016 Leonhard Czarnetzki (BSc., University of Innsbruck, jointly with H. J. Briegel)

List of Publications

August 12, 2021

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Total Citations: 1745 (google scholar) **ORCID:** 0000-0003-1950-8640

h-index: 26 **i10-index:** 35

Peer-Reviewed Publications (in reverse chronological order)

(1) Matteo Fadel, Ayaka Usui, Marcus Huber, Nicolai Friis, and Giuseppe Vitagliano
Entanglement Quantification in Atomic Ensembles
Phys. Rev. Lett. **127**, 010401 (2021)
Preprint: arXiv:2103.15730 [quant-ph]

(2) Valeria Saggio, Beate Asenbeck, Arne Hamann, Teodor Strömberg, Peter Schiansky, Vedran Dunjko, Nicolai Friis, Nicholas C. Harris, Michael Hochberg, Dirk Englund, Sabine Wölk, Hans J. Briegel, and Philip Walther
Experimental quantum speed-up in reinforcement learning agents
Nature **591**, 229–233 (2021)
Preprint: arXiv:2103.06294 [quant-ph]

(3) Emanuel Schwarzhans, Maximilian Lock, Paul Erker, Nicolai Friis, and Marcus Huber
Autonomous Temporal Probability Concentration: Clockworks and the Second Law of Thermodynamics
Phys. Rev. X **11**, 011046 (2021)
Preprint: arXiv:2007.01307 [quant-ph]

(4) Simon Morelli, Ayaka Usui, Elizabeth Agudelo, and Nicolai Friis
Bayesian parameter estimation using Gaussian states and measurements
Quantum Sci. Technol. **6**, 025018 (2021)
Preprint: arXiv:2009.03709 [quant-ph]

(5) Alexander Erhard*, Hendrik Poulsen Nautrup*, Michael Meth, Lukas Postler, Roman Stricker, Martin Stadler, Vlad Negnevitsky, Martin Ringbauer, Philipp Schindler, Hans J. Briegel, Rainer Blatt, Nicolai Friis‡, and Thomas Monz‡
(*These authors contributed equally to this work. ‡ Corresponding authors.)
Entangling logical qubits with lattice surgery
Nature **589**, 220–224 (2021)
Preprint: arXiv:2006.03071 [quant-ph]

(6) Natalia Herrera Valencia, Vatshal Srivastav, Matej Pivoluska, Marcus Huber, Nicolai Friis, Will McCutcheon, and Mehul Malik
High-dimensional Pixel Entanglement Efficient Generation and Certification
Quantum **4**, 376 (2020)
Preprint: arXiv:2004.04994 [quant-ph]

(7) Tiago Debarba, Fernando Iemini, Geza Giedke, and Nicolai Friis
Teleporting quantum information encoded in fermionic modes
Phys. Rev. A **101**, 052326 (2020)
Preprint: arXiv:2002.08201 [quant-ph]

(8) Yelena Guryanova, Nicolai Friis, and Marcus Huber
Ideal Projective Measurements Have Infinite Resource Costs
Quantum **4**, 222 (2020)
Preprint: arXiv:1805.11899 [quant-ph]

(9) Hendrik Poulsen Nautrup, Nicolas Delfosse, Vedran Dunjko, Hans J. Briegel, and Nicolai Friis
Optimizing Quantum Error Correction Codes with Reinforcement Learning
Quantum **3**, 215 (2019)
Preprint: arXiv:1812.08451 [quant-ph]

(10) Tiago Debarba, Gonzalo Manzano, Yelena Guryanova, Marcus Huber, and Nicolai Friis
Work estimation and work fluctuations in the presence of non-ideal measurements
New J. Phys. **21**, 113002 (2019)
Preprint: arXiv:1902.08568 [quant-ph]

(11) Faraj Bakhshinezhad, Fabien Clivaz, Giuseppe Vitagliano, Paul Erker, Ali Rezakhani, Marcus Huber, and Nicolai Friis
Thermodynamically optimal creation of correlations
J. Phys. A Math. Theor. **52**, 465303 (2019)
Preprint: arXiv:1904.07942 [quant-ph]

(12) Nicolai Friis, Giuseppe Vitagliano, Mehul Malik, and Marcus Huber
Entanglement Certification From Theory to Experiment
Nat. Rev. Phys. **1**, 72–87 (2019)
Preprint: arXiv:1906.10929 [quant-ph]

(13) Theeraphot Sriarunothai, Sabine Wölk, Gouri Shankar Giri, Nicolai Friis, Vedran Dunjko, Hans J. Briegel, and Christof Wunderlich
Speeding-up the decision making of a learning agent using an ion trap quantum processor
Quant. Sci. Techn. **4**, 015014 (2019)
Preprint: arXiv:1709.01366 [quant-ph]

(14) Jessica Bavaresco, Natalia Herrera Valencia, Claude Klöckl, Matej Pivoluska, Paul Erker, Nicolai Friis, Mehul Malik, and Marcus Huber
Measurements in two bases are sufficient for certifying high-dimensional entanglement
Nat. Phys. **14**, 1032–1037 (2018)
Preprint: arXiv:1709.07344 [quant-ph]

(15) Nicolai Friis and Marcus Huber
Precision and Work Fluctuations in Gaussian Battery Charging
Quantum **2**, 61 (2018)
Preprint: arXiv:1708.00749 [quant-ph]

(16) Nicolai Friis*, Oliver Marty*, Christine Maier, Cornelius Hempel, Milan Holzäpfel, Petar Jurcevic, Martin B. Plenio, Marcus Huber, Christian Roos, Rainer Blatt, and Ben Lanyon (*These authors have contributed equally to this work.)
Observation of Entangled States of a Fully Controlled 20-Qubit System
Phys. Rev. X **8**, 021012 (2018)
Preprint: arXiv:1711.11092 [quant-ph]

(17) Hendrik Poulsen Nautrup, Nicolai Friis, and Hans J. Briegel
Fault-tolerant interface between quantum memories and quantum processors
Nat. Commun. **8**, 1321 (2017)
Preprint: arXiv:1609.08062 [quant-ph]

(18) Nicolai Friis, Davide Orsucci, Michalis Skotiniotis, Pavel Sekatski, Vedran Dunjko, Hans J. Briegel, and Wolfgang Dür
Flexible resources for quantum metrology
New J. Phys. **19**, 063044 (2017)
Preprint: arXiv:1610.09999 [quant-ph]
(selected for inclusion in the 'Highlights of 2017' by the editors of New J. Phys.)

(19) Nicolai Friis, Sridhar Bulusu, and Reinhold A. Bertlmann
Geometry of two-qubit states with negative conditional entropy
J. Phys. A: Math. Theor. **50**, 125301 (2017)
Preprint: arXiv:1609.04144 [quant-ph]

(20) Eric G. Brown, Nicolai Friis, and Marcus Huber
Passivity and practical work extraction using Gaussian operations
New J. Phys. **18**, 113028 (2016)
Preprint: arXiv:1608.04977 [quant-ph]

(21) Nicolai Friis, Marcus Huber, and Martí Perarnau-Llobet
Energetics of correlations in interacting systems
Phys. Rev. E **93**, 042135 (2016)
Preprint: arXiv:1511.08654 [quant-ph]

(22) Nicolai Friis
Reasonable fermionic quantum information theories require relativity
New J. Phys. **18**, 033014 (2016)
Preprint: arXiv:1502.04476 [quant-ph]

(23) Nicolai Friis, Alexey A. Melnikov, Gerhard Kirchmair, and Hans J. Briegel
Coherent controlization using superconducting qubits
Sci. Rep. **5**, 18036 (2015)
Preprint: arXiv:1508.00447 [quant-ph]

(24) Nicolai Friis, Michalis Skotiniotis, Ivette Fuentes, and Wolfgang Dür
Heisenberg scaling in Gaussian quantum metrology
Phys. Rev. A **92**, 022106 (2015)
Preprint: arXiv:1502.07654 [quant-ph]

(25) David E. Bruschi*, Martí Perarnau-Llobet*, Nicolai Friis*, Karen V. Hovhannisyan, and Marcus Huber (*These authors contributed equally to this work.)
The thermodynamics of creating correlations: Limitations and optimal protocols
Phys. Rev. E **91**, 032118 (2015)
Preprint: arXiv:1409.4647 [quant-ph]

(26) Vedran Dunjko, Nicolai Friis, and Hans J. Briegel
Quantum-enhanced deliberation of learning agents using trapped ions
New J. Phys. **17**, 023006 (2015)
Preprint: arXiv:1407.2830 [quant-ph]

(27) Nicolai Friis, Vedran Dunjko, Wolfgang Dür, and Hans J. Briegel
Implementing quantum control for unknown subroutines
Phys. Rev. A **89**, 030303(R) (2014)
Preprint: arXiv:1401.8128 [quant-ph]

(28) David E. Bruschi, Nicolai Friis, Ivette Fuentes, and Silke Weinfurtner
On the robustness of entanglement in analogue gravity systems
New J. Phys. **15**, 113016 (2013)
Preprint: arXiv:1305.3867 [quant-ph]

(29) Nicolai Friis, Antony R. Lee, and Jorma Louko
Scalar, spinor, and photon fields under relativistic cavity motion
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Preprint: arXiv:1307.1631 [quant-ph]

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(i) Philip Taranto*, Faraj Bakhshinezhad*, Andreas Bluhm†, Ralph Silva†, Nicolai Friis, Maximilian P. E. Lock, Giuseppe Vitagliano, Felix C. Binder, Tiago Debarba, Emanuel Schwarzhans, Fabien Clivaz, and Marcus Huber
(*,† These authors contributed equally)
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Book Chapters (in reverse chronological order)

(iii) Giuseppe Vitagliano, Claude Klöckl, Marcus Huber, and Nicolai Friis
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Perspective Articles (in reverse chronological order)

(iv) Nicolai Friis
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(I) Nicolai Friis
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