Role of Quantum Coherence in Thermodynamics

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We find necessary and sufficient conditions to determine the interconvertibility of quantum systems under time-translation covariant evolution, and use it to solve several problems in quantum thermodynamics both in the single-shot and asymptotic regimes. It is well known that the resource theory of quantum athermality is not reversible, but in Brandão *et al.* [Phys. Rev. Lett. 111, 250404 (2013)] it was claimed that the theory becomes reversible "provided a sublinear amount of coherent superposition over energy levels is available." Here we show that if a sublinear amount of coherence among energy levels were considered free, then the resource theory of athermality would become trivial. Instead, we show that by considering a sublinear amount of energy to be free, the theory of athermality becomes reversible for the pure-state case. A proof of the same claim for the mixed-state case is still lacking.

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I. INTRODUCTION

Thermodynamics is one of the most prevailing theories in physics with vast applications spreading from its early-day focus on steam engines to modern applications in biochemistry, nanotechnology, and black=hole physics, to name just a few [1-3]. Despite the success of this field, the foundations of thermodynamics remain controversial even today. Not only is there persistent confusion over the relation between the macroscopic and microscopic laws, in particular, their reversibility and time symmetry, there is not even consensus on how best to formulate the second law. Indeed, as the Nobel Laureate Percy Bridgman remarked in 1941 "there are almost as many formulations of the second law as there have been discussions of it" and the situation has not improved much since then. In recent years, researchers have begun to adopt a new perspective on these foundational problems by reformulating thermodynamics as a resource theory [4-6]. In this approach to thermodynamics, a system that is not in equilibrium with its environment is considered as a resource called athermality. Athermality is the fuel that is consumed, in work extraction, computational erasure operation, and other thermodynamical tasks [6–31].

The resource-theoretic approach to thermodynamics focuses on how to quantify a state's deviation from equilibrium, how to use this for useful tasks in quantum thermodynamics, and what the necessary and sufficient conditions are for one state to be converted to another. In this approach one can consider various notions of state conversion: exact and approximate, single copy and multiple copy, with and without the help of a catalyst. Such quantum information techniques lead to many novel insights, particularly given the historical significance of the notion of information for foundational topics, such as Maxwell's demon [32], the thermodynamic reversibility of computation [33,34], Landauer's principle about the work cost of erasure [7,35], and Jaynes's use of maximum entropy principles in deriving statistical mechanics [36,37]. Moreover, the resource-theoretic approach to thermodynamics demonstrates that the standard formulation of the second law of thermodynamics, as the nondecrease of entropy, is inadequate as a criterion for deciding whether or not a given state conversion is possible. Nonetheless, one can identify a set of measures of the degree of nonequilibrium (including the entropy), such that the state conversion is possible if and only if all of these measures are not increasing [12,27].

The role of quantum coherence in the resource theory of athermality has several subtleties that were overlooked in some of these works, including the seminal paper [10] that introduced the resource theory of athermality [38]. Specifically, one of the main results of Ref. [10] asserts that the free energy "quantifies the rate at which resource states can be reversibly interconverted asymptotically, provided that a sublinear amount of coherent superposition over energy

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levels is available, a situation analogous to the sublinear amount of classical communication required for entanglement dilution." However, it is relatively simple to show [see Ref. [39] as well as Eq. (123) below] that the quantum coherence of a pure quantum state $|\psi\rangle^{\otimes n}$ grows at most logarithmically with *n*, so that if a sublinear amount of coherence among energy levels were considered free, then the resource theory of athermality would become trivial. What is meant in Ref. [10] is that coherence is provided among energy levels that grows sublinearly with *n*. In other words, they assume that the total energy (not coherence) grows sublinearly with *n*, but the proof given in Ref. [10] contains some gaps; see the Appendix for more specific details.

In this paper we refine this assumption, by considering "asymptotically negligible resources" to be sequences of quantum states $\{\omega_n\}_{n\in\mathbb{N}}$, with $O(\log(n))$ amount of coherence, but whose total energy grows sublinearly with *n*. Since the energy of *n* copies of any (non-zero-energy) state grows linearly with *n*, this assumption is reasonable as it allows for coherence only among energy eigenvectors with asymptotically negligible energy. Under this mild assumption we are able to recover the reversibility of the resource theory of quantum athermality in the pure-state regime.

The paper is organized as follows. After introducing notations and several pertaining preliminary results in Sec. II, we develop the resource theory of time-translation asymmetry in Sec. III, in which we find simple necessary and sufficient conditions for exact manipulation of quantum coherence. We then apply this result in Sec. IV for interconversions among athermality states in the single-shot regime. In Sec. V we develop the resource theory of quantum athermality in the asymptotic regime, and prove that it is reversible if we allow for a sublinear amount of quantum athermality. Finally, in Sec. VI we end with a discussion and outlook.

II. NOTATIONS AND PRELIMINARIES

In this section we introduce our notations and several results from earlier works. We also present some new results and observations. We denote both quantum systems as well as their corresponding Hilbert spaces by the letters A, B, A', B', and R. We consider only finite-dimensional systems and use vertical lines such as |A|, |B|, to denote the dimension of systems A, B, respectively. Replicas of a physical system will be denoted with the tilde symbol above them. For example, \tilde{A} and \tilde{B} are replicas of A and B, respectively, and in particular $|\tilde{A}| = |A|$ and $|B| = |\tilde{B}|$. The set of positive semidefinite matrices acting on system A will be denoted by $\mathcal{Pos}(A)$, and quantum states (also called density matrices) in $\mathcal{Pos}(A)$ will be denoted by $\mathcal{PURE}(A)$. The set of all completely positive trace-preserving (CPTP)

maps, i.e., quantum channels, from system A to B are denoted by $\text{CPTP}(A \rightarrow B)$.

We use superscripts to indicate actions on subsystems of a composite physical system. For example, let $\rho \in \text{Pos}(A)$, $\sigma \in \text{Pos}(AB)$, and $\mathcal{E} \in \text{CPTP}(B \to B')$. Then, the notation $\rho^A \sigma^{AB}$ is a short version corresponding to $(\rho^A \otimes I^B) \sigma^{AB}$, and similarly $\mathcal{E}^{B \to B'}(\sigma^{AB})$ is a short notation of $(\text{id}^A \otimes \mathcal{E}^{B \to B'})(\sigma^{AB})$, where id^A is the identity channel. With these notations, the Choi matrix of a channel $\mathcal{E} \in \text{CPTP}(A \to B)$ is defined as

$$J_{\mathcal{E}}^{AB} := \mathcal{E}^{\tilde{A} \to B} \left(\Phi^{A\tilde{A}} \right), \tag{1}$$

where $\Phi^{A\tilde{A}} := |\Phi^{A\tilde{A}}\rangle\langle\Phi^{A\tilde{A}}|$, and $|\Phi^{A\tilde{A}}\rangle := \sum_{x=1}^{m} |xx\rangle^{A\tilde{A}}$ (with m := |A|) is the unnormalized maximally entangled state.

In this paper we consider only physical systems whose Hamiltonians are well defined (i.e., no interactions with other systems). For example, the Hamiltonians of physical systems A and B is denoted, respectively, by H^A and H^B . Moreover, the Hamiltonian of system A (and similarly of system B, etc.) is expressed as

$$H^A = \sum_{x=1}^m a_x \Pi_x^A,$$
 (2)

where $\{a_x\}_{x=1}^m$ are distinct energy eigenvalues, and $\{\Pi_x\}_{x=1}^m$ are orthogonal projectors satisfying $\Pi_x^A \Pi_y^A = \delta_{xy} \Pi_x^A$ for all $x, y \in [m] := \{1, \dots, m\}$.

A. Notations of types

Let $x^n := (x_1, \ldots, x_n)$ be a sequence with *n* elements such that $x_i \in [m]$ for all $i = 1, \ldots, n$. For any $z \in [m]$ let $N(z|x^n)$ be the number of elements in the sequence $x^n := (x_1, \ldots, x_n)$ that are equal to *z*. The *type* of the sequence x^n is a probability vector in \mathbb{R}^m_+ given by

$$\mathbf{t}(x^{n}) := \left(t_{1}(x^{n}), \dots, t_{m}(x^{n})\right)^{T},$$
(3)

where

$$t_z(x^n) := \frac{1}{n} N(z|x^n) \quad \forall \ z \in [m].$$

$$\tag{4}$$

The significance of types to our work comes into play when we consider an independent identically distributed (IID)~ **p** source. In this case, the probability of a sequence x^n drawn from the source is given by (see e.g., Ref. [40])

$$p_{x^{n}} := p_{x_{1}} \cdots p_{x_{n}} = 2^{-n \left(H(\mathbf{t}(x^{n})) + D(\mathbf{t}(x^{n}) \| \mathbf{p}) \right)}, \qquad (5)$$

where $H(\mathbf{t}(x^n))$ is the Shannon entropy of the type of the sequence x^n , and $D(\mathbf{t}(x^n) || \mathbf{p})$ is the Kullback-Leibler divergence between $\mathbf{t}(x^n)$ and \mathbf{p} .

We denote by Type(n, m) the set of all types of sequences in $[m]^n$, and point out that its number of elements is bounded by [40]

$$|\mathsf{Type}(n,m)| \leqslant (n+1)^m. \tag{6}$$

The set of all sequences x^n of a given type $\mathbf{t} = (t_1, \ldots, t_m)$ is denoted as $x^n(\mathbf{t})$. We emphasize that $x^n(\mathbf{t})$ denotes a *set* of sequences whose type is \mathbf{t} , whereas $\mathbf{t}(x^n)$ denotes a *sin-gle* probability vector (i.e., the type of a specific sequence x^n). The number of sequences in the set $x^n(\mathbf{t})$ is given by the combinatorial formula of arranging nt_1, \ldots, nt_m objects in a sequence; in other words,

$$\left|x^{n}(\mathbf{t})\right| = \binom{n}{nt_{1}, \dots, nt_{m}} \coloneqq \frac{n!}{\prod_{x=1}^{m}(nt_{x})!}.$$
 (7)

The above formula is somewhat cumbersome, but by using Stirling's approximation it can be bounded by [40]

$$\frac{1}{(n+1)^m} 2^{nH(\mathbf{t})} \leqslant \left| x^n(\mathbf{t}) \right| \leqslant 2^{nH(\mathbf{t})}.$$
 (8)

B. Time-translation symmetry

In this subsection we state a few facts about timetranslation symmetry. We say that a quantum state ρ^A is time-translation invariant, or quasiclassical, if for all $t \in \mathbb{R}$ we have

$$e^{-iH^A t} \rho^A e^{iH^A t} = \rho^A. \tag{9}$$

Definition II.1: Let $\mathcal{E} \in \text{CPTP}(A \to B)$. We say that $\mathcal{E}^{A \to B}$ is time-translation covariant if for all $t \in \mathbb{R}$ and all $\rho \in \mathfrak{D}(A)$

$$\mathcal{E}^{A \to B}\left(e^{-iH^{A}t}\rho^{A}e^{iH^{A}t}\right) = e^{-iH^{B}t}\mathcal{E}^{A \to B}\left(\rho^{A}\right)e^{iH^{B}t}.$$
 (10)

The set of all the channels in $\text{CPTP}(A \rightarrow B)$ that are timetranslation covariant is denoted by $\text{COV}(A \rightarrow B)$.

The set of channels $COV(A \rightarrow A)$ contains a special quantum channel known as the *twirling* channel. Expressing the Hamiltonian of system A as in Eq. (2), the twirling channel on system A is defined by

$$\mathcal{P}^{A \to A}\left(\rho^{A}\right) := \sum_{x=1}^{m} \Pi_{x}^{A} \rho^{A} \Pi_{x}^{A}.$$
 (11)

This twirling channel, also known as the "pinching channel" (see, e.g., Ref. [41]), has the property that a state $\rho \in \mathfrak{D}(A)$ is quasiclassical if and only if $\mathcal{P}(\rho) = \rho$, and if a quantum channel $\mathcal{E} \in \text{COV}(A \to A)$ then $\mathcal{P} \circ \mathcal{E} = \mathcal{E} \circ \mathcal{P}$. Moreover, if the Hamiltonian H^A is nondegenerate then

$$\mathcal{P}^{A \to A} = \Delta^{A \to A},\tag{12}$$

where $\Delta^{A \to A}$ is the completely dephasing channel defined as

$$\Delta^{A \to A} \left(\rho^{A} \right) = \sum_{x=1}^{m} \langle x | \rho^{A} | x \rangle | x \rangle \langle x |^{A} \qquad \forall \ \rho \in \mathfrak{D}(A).$$
(13)

The twirling channel can also be used to quantify timetranslation asymmetry. For example, the relative entropy distance of a quantum state $\rho \in \mathfrak{D}(A)$ to its twirled state $\mathcal{P}(\rho)$ is a time-translation asymmetry (sometimes referred to as coherence) measure given by

$$C(\rho) := D\left(\rho \| \mathcal{P}(\rho)\right) = H(\mathcal{P}(\rho)) - H(\rho), \qquad (14)$$

where $D(\rho || \sigma) := \operatorname{Tr}[\rho \log \rho] - \operatorname{Tr}[\rho \log \sigma]$ is the Umegaki relative entropy and $H(\rho) := -\operatorname{Tr}[\rho \log \rho]$ is the von Neumann entropy. The above function is nonincreasing under time-translation covariant operations, and achieves its maximal value of $\log |A|$ on the maximally coherent state $|+\rangle := 1/\sqrt{|A|} \sum_{x=1}^{|A|} |x\rangle$. For *n* copies of system *A*, we denote by $\mathcal{P}_n \in$

For *n* copies of system *A*, we denote by $\mathcal{P}_n \in \text{COV}(A^n \to A^n)$ the pinching channel associated with the total Hamiltonian H^{A^n} given by

$$H^{A^{n}} := H^{A} \otimes I^{A} \otimes \cdots \otimes I^{A} + I^{A} \otimes H^{A} \otimes \cdots \otimes I^{A} + \cdots + I^{A} \otimes \cdots \otimes I^{A} \otimes H^{A}.$$
(15)

With these notations we have $\mathcal{P} = \mathcal{P}_1$. In Ref. [39] it was shown that $C(\rho^{\otimes n})$ grows logarithmicly with *n* [see also Eq. (123) below] and in particular,

$$\lim_{n \to \infty} \frac{1}{n} C\left(\rho^{\otimes n}\right) = 0.$$
(16)

C. The resource theory of athermality

In this subsection we review the resource theory of athermality. We put emphasis on some subtleties that are quite often overlooked in the existing literature. In particular, we distinguish between thermal operations and closed thermal operations. Moreover, we prove some new results. Specifically, to the author's knowledge, all the lemmas and theorems presented here are new.

1. Free states and athermality resource states

The free states in the resource theory of athermality correspond to physical systems that are in thermal equilibrium with their surrounding. For a heat bath that is held at a fixed inverse temperature $\beta := 1/k_B T$, the free thermodynamics state,

$$\gamma^{B} := \frac{e^{-\beta H^{B}}}{\operatorname{Tr}\left[e^{-\beta H^{B}}\right]},\tag{17}$$

is the thermal equilibrium state known as the Gibbs state (here H^B is the Hamiltonian associated with the heat bath). We always use the Greek letter γ to indicate a Gibbs state. For example, the notation γ^A , $\gamma^{A'}$, and γ^B , corresponds to the Gibbs states of systems A, A', and B, respectively. Moreover, the joint Gibbs state of two non-interacting systems A and B will be denoted simply by $\gamma^{AB} = \gamma^A \otimes \gamma^B$.

In the quantum resource theory (QRT) of athermality, every physical system that can be used as a resource has a well-defined Hamiltonian. Therefore, a physical system A cannot be characterized just by a density matrix $\rho \in \mathfrak{D}(A)$ since the resourcefulness of the state depends also on the Hamiltonian of the system, H^A . For this reason, every thermodynamic state in quantum thermodynamics comprises of a quantum state $\rho \in \mathfrak{D}(A)$ acting on the Hilbert space A, and a time-independent Hamiltonian $H^A \in \text{Pos}(A)$ that governs the dynamics of the quantum system A. That is, a *state* of athermality can be characterized by a pair (ρ^A, H^A). This is indeed the characterization used extensively in the literature.

From the resource-theoretic perspective, this characterization of an athermality state has several drawbacks. First, it is not invariant under an energy shift of the form $H^A \mapsto H^A + cI^A$, where $c \in \mathbb{R}$ is some constant. Indeed, the choice of setting the minimal energy of a system to be zero is somewhat arbitrary. Second, the resourcefulness of the state ρ^A is determined in relation to its deviation from the Gibbs state γ^A of system A. Therefore, it seems more natural to characterize athermality states (i.e., the "objects" of this theory) by pairs of the form (ρ^A, γ^A) . Note that all the relevant information about the Hamiltonian H^A is contained in the Gibbs state γ^A , which is invariant under energy shifts.

2. Free operations

The set of free operations relative to a background heat bath at temperature *T* comprises of three basic steps:

- 1. Thermal equilibrium. Any subsystem *B*, with Hamiltonian $H^B \in Pos(B)$, can be prepared in its thermal Gibbs state γ^B .
- 2. Conservation of energy. Unitary operation on a composite physical system that commutes with the total Hamiltonian can be implemented.
- 3. Discarding subsystems. It is possible to trace over any subsystem (with a well-defined Hamiltonian) of a composite system.

Any CPTP map comprising of the above three steps is called a *thermal operation*. Any thermal operation $\mathcal{E} \in \text{CPTP}(A \to A)$ can be expressed as

$$\mathcal{E}^{A \to A}(\rho^{A}) = \operatorname{Tr}_{B}\left[\mathcal{U}^{AB \to AB}\left(\rho^{A} \otimes \gamma^{B}\right)\right], \qquad (18)$$

where $\mathcal{U} \in \text{CPTP}(AB \to AB)$ is a unitary channel that is Gibbs preserving; in other words,

$$\mathcal{U}^{AB\to AB}\left(\gamma^{AB}\right) = \gamma^{AB},\tag{19}$$

where $\gamma^{AB} = \gamma^A \otimes \gamma^B$. In the lemma below we show that $\text{CPTP}(A \to A')$ with $|A| \neq |A'|$ also contains thermal operations.

Lemma II.1: Let AB, A'B' be two composite physical systems with |AB| = |A'B'|, and let $\mathcal{U} \in \text{CPTP}(AB \to A'B')$ be a Gibbs preserving unitary channel; that is,

$$\mathcal{U}^{AB\to A'B'}\left(\gamma^{AB}\right)=\gamma^{A'B'}.$$

Then, the map (defined on all $\omega \in \mathfrak{D}(A)$)

$$\mathcal{N}^{A \to A'}\left(\omega^{A}\right) := \operatorname{Tr}_{B'}\left[\mathcal{U}^{AB \to A'B'}\left(\omega^{A} \otimes \gamma^{B}\right)\right]$$
(20)

is a thermal operation.

Proof. Consider the joint Gibbs state $\gamma^{ABA'B'} := \gamma^{AB} \otimes \gamma^{A'B'}$ and let $\mathcal{V} \in \text{CPTP}(ABA'B' \to ABA'B')$ be the unitary channel given by

$$\mathcal{V}^{ABA'B'\to ABA'B'} := \mathcal{U}^{AB\to A'B'} \otimes \mathcal{U}^{*A'B'\to AB}.$$
 (21)

Observe that $\mathcal{V}^{ABA'B'} \rightarrow ABA'B'}$ preserves the joint Gibbs state $\gamma^{ABA'B'}$. Hence, the channel

$$\begin{aligned} \operatorname{Tr}_{ABB'} \left[\mathcal{V}^{ABA'B' \to ABA'B'} \left(\omega^{A} \otimes \gamma^{BA'B'} \right) \right] \\ &= \operatorname{Tr}_{ABB'} \left[\mathcal{U}^{AB \to A'B'} \left(\omega^{A} \otimes \gamma^{B} \right) \otimes \mathcal{U}^{*A'B' \to AB} (\gamma^{A'B'}) \right] \\ &= \operatorname{Tr}_{B'} \left[\mathcal{U}^{AB \to A'B'} \left(\omega^{A} \otimes \gamma^{B} \right) \right] \\ &= \mathcal{N}^{A \to A'} (\omega^{A}) , \end{aligned}$$

is a thermal operation. This completes the proof.

We denote by $TO(A \rightarrow A')$ the set of all thermal operations in $CPTP(A \rightarrow A')$. For fixed systems *A* and *A'* the set $TO(A \rightarrow A')$ is in general not closed and not convex. It stems from the fact that the dimensions of systems *B* and *B'* as appearing in Eq. (20) are unbounded. Therefore, it will be convenient to define the closure of $TO(A \rightarrow$ *A'*), denoted by $CTO(A \rightarrow A')$, as a set of channels in $CPTP(A \rightarrow A')$ with the property that $\mathcal{E} \in CTO(A \rightarrow A')$ if and only if there exists a sequence of thermal operations $\{\mathcal{E}_k\}_{k\in\mathbb{N}}$, where each $\mathcal{E}_k \in \mathrm{TO}(A \to A')$ and

$$\lim_{k \to \infty} \mathcal{E}_k = \mathcal{E}.$$
 (22)

By definition, the set $CTO(A \rightarrow A')$ is closed. We now prove that it is also convex.

Theorem II.1: *The set* $CTO(A \rightarrow A')$ *is convex.*

Proof. We start by showing that $TO(A \rightarrow A')$ is closed under convex combination with rational coefficients. Specifically, let

$$\mathcal{N}^{A \to A'} := \sum_{x=1}^{\ell} \frac{m_x}{m} \mathcal{N}_x^{A \to A'}, \qquad (23)$$

where each $m_x \in \mathbb{N}$, $m := \sum_{x=1}^{\ell} m_x$, and each $\mathcal{N}_x \in \mathrm{TO}(A \to A')$. Since each \mathcal{N}_x is a thermal operation it can be expressed as

$$\mathcal{N}_{x}^{A \to A'}\left(\omega^{A}\right) := \operatorname{Tr}_{B'_{x}}\left[\mathcal{U}_{x}^{AB_{x} \to A'B'_{x}}\left(\omega^{A} \otimes \gamma^{B_{x}}\right)\right], \quad (24)$$

for some systems B_x, B'_x and some unitary channel $\mathcal{U}_x \in$ $CPTP(AB_x \to A'B'_x)$. For each $y \in [m]$, let k_y be the integer in $[\ell]$ satisfying

$$\sum_{x=1}^{k_y-1} m_x \leqslant y < \sum_{x=1}^{k_y} m_x,$$
 (25)

and define

$$B := \bigoplus_{y=1}^{m} B_{k_y} , \quad B' := \bigoplus_{y=1}^{m} B'_{k_y}, \text{ and } \gamma^B := \frac{1}{m} \bigoplus_{y=1}^{m} \gamma^{B_{k_y}}.$$
(26)

Finally, for any $\eta^{AB} = \bigoplus_{\nu=1}^{m} \eta^{AB_{k_{\nu}}} \in \mathfrak{D}(AB)$ we define the action of the unitary channel $\mathcal{U} \in \text{CPTP}(AB \to A'B')$ as

$$\mathcal{U}^{AB \to A'B'}(\eta^{AB}) = \bigoplus_{y=1}^{m} \mathcal{U}^{AB_{k_y} \to A'B'_{k_y}}_{k_y}(\eta^{AB_{k_y}}).$$
(27)

With these definitions we get

$$\operatorname{Tr}_{B'}\left[\mathcal{U}^{AB\to A'B'}\left(\omega^{A}\otimes\gamma^{B}\right)\right]$$

$$=\frac{1}{m}\sum_{y=1}^{m}\operatorname{Tr}_{B'_{ky}}\left[\mathcal{U}^{AB_{ky}\to A'B'_{ky}}\left(\omega^{A}\otimes\gamma^{B_{ky}}\right)\right]$$

$$=\frac{1}{m}\sum_{x=1}^{\ell}m_{x}\operatorname{Tr}_{B'_{x}}\left[\mathcal{U}^{AB_{x}\to A'B'_{x}}_{x}\left(\omega^{A}\otimes\gamma^{B_{x}}\right)\right], \quad (28)$$

where in the last line we use the fact that for any $x \in$ $[\ell]$ there exist m_x values of $y \in [m]$ for which $k_y = x$. Finally, observe that the rhs of the equation above is precisely $\mathcal{N}^{A \to A'}(\omega^A)$. Therefore, $\mathcal{N}^{A \to A'}$ is a thermal operation. This completes the proof that any rational convex combination of thermal operations is a thermal operation.

To prove the convexity of $CTO(A \rightarrow A')$ let $\{\mathcal{M}_x\}_{x=1}^k$ be k channels in $CTO(A \rightarrow A')$ and let

$$\mathcal{M}^{A \to A'} := \sum_{x=1}^{k} p_x \mathcal{M}_x^{A \to A'}$$
(29)

be a convex combination of the k channels $\{\mathcal{M}_x\}$. For each $n \in \mathbb{N}$ let $\mathcal{M}_x^{(n)} \in \mathrm{TO}(A \to A')$ be such that $\lim_{n\to\infty} \mathcal{M}_x^{(n)} = \mathcal{M}_x$, and let $\{p_x^{(n)}\}_{x=1}^k$ be a rational probability distribution with the property that $\lim_{n\to\infty} p_x^{(n)} = p_x$. Now, from the previous argument we have that for all $n \in \mathbb{N}$ the rational convex combination

$$\sum_{x=1}^{k} p_x^{(n)} \mathcal{M}_x^{(n)}$$
(30)

is in $TO(A \rightarrow A')$. Therefore, by definition, the limit

$$\lim_{n \to \infty} \sum_{x=1}^{k} p_x^{(n)} \mathcal{M}_x^{(n)} = \mathcal{M}$$
(31)

is in $CTO(A \rightarrow A')$. This completes the proof.

Every thermal operation $\mathcal{E} \in \text{CPTP}(A \to A')$ has two key properties:

- 1. $\mathcal{E}^{A \to A'}$ is *Gibbs preserving operation* (GPO); that is, $\mathcal{E}(\gamma^A) = \gamma^{A'}.$ 2. $\mathcal{E}^{A \to A'}$ is time-translation covariant; in other words,
- $\mathcal{E} \in \mathrm{COV}(A \to A').$

The set of all Gibbs preserving operations in $\text{CPTP}(A \rightarrow$ A') will be denoted by $GPO(A \rightarrow A')$, and those that are Gibbs preserving covariant (GPC) quantum channels (i.e., channels that satisfy the above two properties) are denoted by $GPC(A \rightarrow A')$. In what follows, we also use the notations

$$(\rho^A, \gamma^A) \xrightarrow{\mathfrak{F}} (\sigma^B, \gamma^B),$$
 (32)

to indicate that (ρ^A, γ^A) can be converted to (σ^B, γ^B) by the free operations \mathfrak{F} . In our context, \mathfrak{F} can stand for thermal operations, closed thermal operations (CTO), GPC. and GPO. Since GPC form a closed set of operations we have for any two systems A and A'

$$TO(A \to A') \subset CTO(A \to A') \subset GPC(A \to A')$$
$$\subset GPO(A \to A'). \quad (33)$$

We now show that the pinching channel is a thermal operation.

Lemma II.2: Consider the pinching channel $\mathcal{P} \in CPTP(A \rightarrow A)$ associated with the Hamiltonian of system *A. Then,* $\mathcal{P} \in TO(A \rightarrow A)$.

Proof. Expressing the Hamiltonian of system A as in Eq. (2), the pinching channel $\mathcal{P} \in \text{CPTP}(A \to A)$ can be written as a mixture of unitaries of the form (see, for example, Ref. [41])

$$\mathcal{P}(\rho) = \frac{1}{m} \sum_{x=1}^{m} U_x \rho U_x^* \qquad \forall \ \rho \in \mathfrak{D}(A), \qquad (34)$$

where

$$U_x^A := \sum_{x' \in [m]} e^{\frac{2\pi i x x'}{m}} \Pi_{x'}^A.$$
 (35)

Clearly, each of the *m* unitaries $\{U_x^A\}$ commutes with the Hamiltonian H^A . Therefore, each unitary channel $\mathcal{U}_x \in CPTP(A \rightarrow A)$, defined via $\mathcal{U}_x(\cdot) := U_x(\cdot)U_x^*$, is a thermal operation. In the proof of Theorem II.1 we show that any rational convex combination of thermal operations is itself a thermal operation. Therefore, the mixture of unitaries in Eq. (34) is a thermal operation. This completes the proof.

3. Quasiclassical athermality

We say that an athermality state (ρ^A, γ^A) is quasiclassical if ρ^A and γ^A commute; that is, ρ is diagonal in the energy eigenbasis of system A. In this case, we denote the athermality state (ρ^A, γ^A) as $(\mathbf{p}^A, \mathbf{g}^A)$, where \mathbf{p}^A and \mathbf{g}^A are probability vectors consisting of the diagonals of ρ^A and γ^A , respectively. In this quasiclassical regime, for two athermality states $(\mathbf{p}^A, \mathbf{g}^A)$ and $(\mathbf{p}^B, \mathbf{g}^B)$ we have (see Theorem 5 in Ref. [7])

$$(\mathbf{p}^{A}, \mathbf{g}^{A}) \xrightarrow{\text{CTO}} (\mathbf{q}^{B}, \mathbf{g}^{B}) \iff (\mathbf{p}^{A}, \mathbf{g}^{A}) \succ (\mathbf{q}^{B}, \mathbf{g}^{B}),$$
(36)

where > denotes *relative majorization*. Relative majorization is a preorder defined between two pairs of probability vectors. Specifically, we say that $(\mathbf{p}^A, \mathbf{g}^A)$ relatively majorizes $(\mathbf{q}^B, \mathbf{g}^B)$ (and write it as in the equation above) if there exists a column stochastic matrix E such that $\mathbf{q}^B = E\mathbf{p}^A$ and $\mathbf{g}^B = E\mathbf{g}^A$. Relative majorization has several characterizations including a geometrical one given by Lorenz curves and testing regions (see, e.g., Ref. [42]).

If the Hamiltonian of system A is fully degenerate (we say in this case that the Hamiltonian is trivial) then $H^A = cI^A$ for some constant $c \ge 0$ and the corresponding Gibbs state,

$$\mathbf{g}^{A} = \mathbf{u}^{(m)} := \frac{1}{m} \begin{pmatrix} 1 \\ \vdots \\ 1 \end{pmatrix}, \qquad (37)$$

is the *m*-dimensional uniform probability vector. We say that two athermality states, $(\mathbf{p}^{A}, \mathbf{g}^{A})$ and $(\mathbf{p}^{B}, \mathbf{g}^{B})$, are equivalent, and write

$$(\mathbf{p}^A, \mathbf{g}^A) \sim (\mathbf{p}^B, \mathbf{g}^B)$$
 (38)

if both $(\mathbf{p}^A, \mathbf{g}^A) \succ (\mathbf{p}^B, \mathbf{g}^B)$ and $(\mathbf{p}^B, \mathbf{g}^B) \succ (\mathbf{p}^A, \mathbf{g}^A)$. One of the remarkable properties of quasiclassical thermodynamics is that a dense set of athermality states are equivalent to states with a trivial (i.e., zero) Hamiltonian [43]. Specifically, let $\mathbf{g} = (g_1, \dots, g_m)^T$ be the Gibbs state of system Aand suppose that its components $\{g_x\}$ are rational. Then, there exists $k_1, \dots, k_m \in \mathbb{N}$ such that for each $x \in [m]$ we have $g_x = k_x/k$, where $k := \sum_{x=1}^m k_x$ is the common denominator. With such a Gibbs state, for any probability vector $\mathbf{p} = (p_1, \dots, p_m)^T$ we have that [43]

$$(\mathbf{p}, \mathbf{g}) \sim (\mathbf{r}, \mathbf{u}^{(k)})$$
 where $\mathbf{r} := \bigoplus_{x=1}^{m} p_x \mathbf{u}^{(k_x)}$. (39)

The above equivalence indicates that athermality of the the quasiclassical system *A* can be fully characterized by the *nonuniformity* of the vector **r**, with $\mathbf{r} = 1/k(1,...,1)^T$ being the least resourceful and $\mathbf{r} = (1, 0, ..., 0)^T$ being the most resourceful. Therefore, in the quasiclassical regime the resource theory of athermality is essentially equivalent to the resource theory of nonuniformity, also known as the resource theory of informational nonequilibrium [16].

4. The golden unit of athermality

A "golden unit" of a resource theory is a constituent of a resource that can be used to measure the resource very much like ebits are used to measure entanglement. Due to the equivalence between athermality and nonuniformity in the quasiclassical regime, we can use units of nonuniformity to measure the athermality of a given state. Specifically, we can take the golden unit to have the form $(|0\rangle\langle 0|^4, \mathbf{u}^4)$. This golden unit is equivalent to [44]

$$(|0\rangle\langle 0|^{A}, \mathbf{u}^{A}) \sim \left(|0\rangle\langle 0|^{X}, \mathbf{u}_{m}^{X}\right), \qquad (40)$$

where X is a two-dimensional classical system, m := |A|, and

$$\mathbf{u}_m^X := \frac{1}{m} |0\rangle \langle 0|^X + \frac{m-1}{m} |1\rangle \langle 1|^X.$$
(41)

Therefore, we can always consider the golden unit to be a qubit. Moreover, note that \mathbf{u}_m^X is well defined even

if *m* is not an integer. This can help simplifying certain expressions, and we therefore consider also the states $(|0\rangle \langle 0|^X, \mathbf{u}_m^X)$ with $m \in \mathbb{R}_+$.

5. Cost and distillation

We denote by \mathfrak{F} the free operations of the resource theory of athermality. We consider three cases in which $\mathfrak{F} =$ CTO, $\mathfrak{F} =$ GPO, and $\mathfrak{F} =$ GPC. In either of these cases, we define the conversion distance as

$$d_{\mathfrak{F}}((\rho^{A},\gamma^{A})\to(\sigma^{B},\gamma^{B})) := \min_{\mathcal{E}\in\mathfrak{F}(A\to B)}\frac{1}{2} \|\sigma^{B}-\mathcal{E}(\rho^{A})\|_{1}.$$
(42)

The conversion distance measures the closest distance (in trace norm) that ρ^A can reach σ^B by using only free operations. For any $\varepsilon > 0$ and $\rho, \gamma \in \mathfrak{D}(A)$, this conversion distance can be used to define the ε -single-shot distillable athermality as

Distill^{$$\varepsilon$$} _{\mathfrak{F}} $(\rho, \gamma) := \log \sup_{0 < m \in \mathbb{R}} \left\{ m : d_{\mathfrak{F}} \left(\left(\rho^{A}, \gamma^{A} \right) \to \left(|0\rangle \langle 0|^{X}, \mathbf{u}_{m}^{X} \right) \right) \leqslant \varepsilon \right\}.$ (43)

The asymptotic distillation of an athermality state (ρ, γ) is defined as

Distill_{$$\mathfrak{F}$$} $(\rho, \gamma) := \lim_{\varepsilon \to 0^+} \sup_{\ell, n \in \mathbb{N}} \left\{ \frac{\ell}{n} : d_{\mathfrak{F}} \left(\left(\rho^{\otimes n}, \gamma^{\otimes n} \right) \to \left(|0\rangle \langle 0|^{\otimes \ell}, \mathbf{u}_2^{\otimes \ell} \right) \right) \leqslant \varepsilon \right\},$ (44)

where \mathbf{u}_2 is the two-dimensional maximally mixed state. The single-shot and asymptotic distillation rates are related by

$$\text{Distill}_{\mathfrak{F}}(\rho,\gamma) = \lim_{\varepsilon \to 0^+} \limsup_{n \to \infty} \frac{1}{n} \text{Distill}_{\mathfrak{F}}^{\varepsilon} \left(\rho^{\otimes n}, \gamma^{\otimes n}\right).$$
(45)

We point out that $\text{Distill}_{\mathfrak{F}}(\rho, \gamma)$ has the property that for any $k \in \mathbb{N}$

$$\frac{1}{k} \text{Distill}_{\mathfrak{F}} \left(\rho^{\otimes k}, \gamma^{\otimes k} \right) \\
= \lim_{\varepsilon \to 0^+} \limsup_{n \to \infty} \frac{1}{nk} \text{Distill}_{\mathfrak{F}}^{\varepsilon} \left(\rho^{\otimes nk}, \gamma^{\otimes nk} \right) \\
\leqslant \lim_{\varepsilon \to 0^+} \limsup_{n' \to \infty} \frac{1}{n'} \text{Distill}_{\mathfrak{F}}^{\varepsilon} \left(\rho^{\otimes n'}, \gamma^{\otimes n'} \right) \\
= \text{Distill}_{\mathfrak{F}}(\rho, \gamma).$$
(46)

Similarly, the conversion distance can be used to define the ε -single-shot athermality cost as

$$\operatorname{Cost}_{\mathfrak{F}}^{\varepsilon}(\rho,\gamma) := \log \inf_{0 < m \in \mathbb{R}} \left\{ m : d_{\mathfrak{F}}\left(\left(|0\rangle\langle 0|^{X}, \mathbf{u}_{m}^{X} \right) \to \left(\rho^{A}, \gamma^{A} \right) \right) \leqslant \varepsilon \right\}.$$
(47)

The asymptotic athermality cost of the state (ρ, γ) is defined as

$$\operatorname{Cost}_{\mathfrak{F}}(\rho,\gamma) := \lim_{\varepsilon \to 0^{+}} \inf_{m,n \in \mathbb{N}} \left\{ \frac{m}{n} : d_{\mathfrak{F}}\left(\left(|0\rangle\langle 0|^{\otimes m}, \mathbf{u}_{2}^{\otimes m} \right) \to \left(\rho^{\otimes n}, \gamma^{\otimes n}\right) \right) \leqslant \varepsilon \right\}.$$
(48)

The single-shot and asymptotic athermality costs are related by

$$\operatorname{Cost}_{\mathfrak{F}}(\rho,\gamma) = \lim_{\varepsilon \to 0^+} \liminf_{n \to \infty} \frac{1}{n} \operatorname{Cost}_{\mathfrak{F}}^{\varepsilon} \left(\rho^{\otimes n}, \gamma^{\otimes n} \right).$$
(49)

For the case that $\mathfrak{F} = \text{GPO}$ all the quantities above have relatively simple closed formulas. In the single-shot regime we have [44]

$$Distill_{GPO}^{\varepsilon}(\rho,\gamma) = D_{\min}^{\varepsilon}(\rho \| \gamma),$$

$$Cost_{GPO}^{\varepsilon}(\rho,\gamma) = D_{\max}^{\varepsilon}(\rho \| \gamma),$$
(50)

where D_{\min}^{ε} is the hypothesis testing divergence defined as

$$D_{\min}^{\varepsilon}(\rho \| \gamma) := \min_{0 \leq \Lambda \leq I^{A}} \left\{ \operatorname{Tr}[\gamma \Lambda] : \operatorname{Tr}[\Lambda \rho] \geq 1 - \varepsilon \right\}$$
(51)

and D_{\max}^{ε} is the smoothed max relative entropy defined as

$$D_{\max}^{\varepsilon}(\rho \| \gamma) := \min \left\{ D_{\max}(\rho' \| \gamma) : \frac{1}{2} \| \rho - \rho' \|_1 \leqslant \varepsilon \right\}$$
(52)

and $D_{\max}(\rho \| \gamma) := \log \min\{t \ge 0 : t\gamma \ge \rho\}$. In the asymptotic regime, under GPO, the resource theory of athermality is reversible as reflected by the equality

$$\text{Distill}_{\mathfrak{F}}(\rho,\gamma) = \text{Cost}_{\mathfrak{F}}(\rho,\gamma) = D(\rho \| \gamma), \qquad (53)$$

where $D(\rho \| \gamma) := \text{Tr}[\rho \log \rho] - \text{Tr}[\rho \log \gamma]$ is the Umegaki relative entropy.

The hypothesis testing divergence that appears above in the formula for the single-shot distillable athermality is neither additive nor subadditive under tensor products. Instead it satisfies a weaker type of subadditivity given in the lemma below. **Lemma II.3:** Let $\varepsilon > 0$, $\rho, \gamma \in \mathfrak{D}(A)$, and $\rho', \gamma' \in \mathfrak{D}(A')$. *Then,*

$$D_{\min}^{\varepsilon}\left(\rho \otimes \rho' \| \gamma \otimes \gamma'\right) \leqslant D_{\min}^{\varepsilon}(\rho \| \gamma) + D_{\max}\left(\rho' \| \gamma'\right).$$
(54)

Proof. By definition,

$$2^{-D_{\min}^{\varepsilon}(\rho \otimes \rho' \| \gamma \otimes \gamma')} = \min_{\operatorname{Tr}\left[(\rho \otimes \rho')\Lambda\right] \ge 1-\varepsilon} \operatorname{Tr}\left[\left(\gamma \otimes \gamma'\right)\Lambda\right],$$
(55)

where the minimum is over all effects $\Lambda \in Pos(AA')$ that satisfies $\Lambda \leq I^{AA'}$. The key idea is to use the inequality

$$\gamma' \geqslant 2^{-D_{\max}(\rho' \| \gamma')} \rho'. \tag{56}$$

This inequality follows directly from the definition of $D_{\max}(\rho' \| \gamma')$. Therefore, from the above two equations we get

$$2^{-D_{\min}^{\varepsilon}(\rho \otimes \rho' \| \gamma \otimes \gamma')} \geq 2^{-D_{\max}(\rho' \| \gamma')} \min_{\operatorname{Tr}[(\rho \otimes \rho')\Lambda] \ge 1-\varepsilon} \operatorname{Tr}\left[(\gamma \otimes \rho')\Lambda\right] = 2^{-D_{\max}(\rho' \| \gamma')} \min_{\operatorname{Tr}[\rho\Gamma] \ge 1-\varepsilon} \operatorname{Tr}\left[\gamma\Gamma\right],$$
(57)

where the second minimum is over all effects $\Gamma \in Pos(A)$ of the form

$$\Gamma := \operatorname{Tr}_{A'} \left[\left(I^A \otimes \rho' \right) \Lambda^{AA'} \right].$$
(58)

By removing the constraint (58) on Γ and taking instead the minimum over all operators $0 \leq \Gamma \leq I^A$ we get that the minimization $\min_{\mathrm{Tr}[\rho\Gamma] \geq 1-\varepsilon} \mathrm{Tr}[\gamma\Gamma]$ is equal by definition to $2^{-D_{\min}(\rho \parallel \gamma)}$. Therefore, since the removal of the constraint (58) can only decrease the second minimization in Eq. (57) we conclude that

$$2^{-D_{\min}^{\varepsilon}(\rho \otimes \rho' \| \gamma \otimes \gamma')} \geqslant 2^{-D_{\max}^{\varepsilon}(\rho' \| \gamma')} 2^{-D_{\min}^{\varepsilon}(\rho \| \gamma)}.$$
(59)

This completes the proof.

6. The relative entropy of athermality

The relative entropy of athermality of a state (ρ^{A}, γ^{A}) is defined in terms of the Umegaki relative entropy as $D(\rho^{A} || \gamma^{A})$. This function quantifies the athermality of the state (ρ^{A}, γ^{A}) , and is related to the free energy via $D(\rho || \gamma) = \beta (F(\rho^{A}) - F(\gamma^{A}))$, where $F(\rho^{A})$ is the free

energy of ρ^A (see Ref. [10]). The relative entropy distance can also be expressed as

$$D(\rho \| \gamma) = -H(\rho) - \operatorname{Tr} [\rho \log \gamma]$$

= $-H(\rho) - \operatorname{Tr} [\mathcal{P}(\rho) \log \gamma]$
= $D(\mathcal{P}(\rho) \| \gamma) + H(\mathcal{P}(\rho)) - H(\rho)$
= $D(\mathcal{P}(\rho) \| \gamma) + C(\rho).$ (60)

That is, the athermality of the state (ρ, γ) can be decomposed into two components:

- 1. Its nonuniformity that is quantified by $D(\mathcal{P}(\rho) \| \gamma)$.
- 2. Its asymmetry (or coherence between energy eigenspaces) that is quantified by the time-translation asymmetry measure $C(\rho)$.

Moreover, since the regularization of the coherence vanishes [see Eq. (16)] we conclude that

$$\lim_{n \to \infty} \frac{1}{n} D\left(\mathcal{P}_n\left(\rho^{\otimes n}\right) \| \gamma^{\otimes n} \right) = D\left(\rho \| \gamma\right).$$
(61)

III. TIME-TRANSLATION SYMMETRY

We start by developing the resource theory of timetranslation asymmetry. Specifically, we provide necessary and sufficient conditions for state conversions in this model. As we mention in the preliminary section, we are considering only in this paper physical systems whose Hamiltonians are well defined. It turns out that the degeneracy of these Hamiltonians play an important role in the manipulation of asymmetry.

A. Degenerate versus nondegenerate Hamiltonians

Let H^A and H^B be the Hamiltonians of two systems A and B, of dimensions m := |A| and n := |B|. The Hamiltonians can be expressed in their spectral decomposition as

$$H^{A} = \sum_{x=1}^{m} a_{x} |x\rangle \langle x|^{A} \quad \text{and} \quad H^{B} = \sum_{y=1}^{n} b_{x} |y\rangle \langle y|^{B}, \quad (62)$$

where $\{a_x\}$ and $\{b_y\}$ are the energy eigenvalues of H^A and H^B , respectively.

Definition III.1: We say that the Hamiltonians H^A and H^B , as defined in Eq. (62), are *relatively nondegenerate* if for all $x, x' \in [m]$ and $y, y' \in [n]$ we have

$$a_x - a_{x'} = b_y - b_{y'} \quad \Rightarrow \quad x = x' \text{ and } y = y'.$$
 (63)

If the condition above does not hold we say that the Hamiltonians are relatively degenerate.

Note that if H^A and H^B are relatively nondegenerate, then each of them is also nondegenerate. For example,

suppose H^A is degenerate with $a_x = a_{x'}$ for some $x \neq x' \in [m]$. Then, for y = y' we get $a_x - a_{x'} = 0 = b_y - b_{y'}$ even though $x \neq x'$. Therefore, relative nondegeneracy is a stronger notion than nondegeneracy. In fact, relative nondegeneracy of H^A and H^B is equivalent to the nondegeneracy of the joint Hamiltonian $H^{AB} = H^A \otimes I^B + I^A \otimes H^B$. Moreover, in the generic case in which H^A and H^B are arbitrary (chosen at random) the Hamiltonians are relatively nondegenerate. For this case, time-translation covariant channels have a very simple characterization.

Theorem III.1: Let A and B be two physical systems with relatively nondegenerate Hamiltonians. Then, $\mathcal{N} \in$ CPTP($A \rightarrow B$) is a time-translation covariant channel if and only if

$$\mathcal{N}^{A \to B} = \Delta^{B \to B} \circ \mathcal{N}^{A \to B} \circ \Delta^{A \to A}, \tag{64}$$

where $\Delta^{A \to A}$ and $\Delta^{B \to B}$ are the completely dephasing channels of systems A and B, respectively. In other words, for physical systems with relatively nondegenerate Hamiltonians only classical channels are time-translation covariant.

Proof. We start by expressing Eq. (10) in the Choi representation. Specifically, by replacing ρ in Eq. (10) with the unnormalized maximally entangled state $\Phi^{A\tilde{A}}$, the rhs becomes

$$e^{-iH^{B}t}\mathcal{E}^{\tilde{A}\to B}\left(\Phi^{A\tilde{A}}\right)e^{iH^{B}t} = e^{-iH^{B}t}J_{\mathcal{E}}^{AB}e^{iH^{B}t},\qquad(65)$$

and the lhs of Eq. (10) can be expressed as

$$\mathcal{E}^{\tilde{A} \to B} \left(e^{-iH^{\tilde{A}}t} \Phi^{A\tilde{A}} e^{iH^{\tilde{A}}t} \right) = \mathcal{E}^{\tilde{A} \to B} \left(e^{-iH^{A}t} \Phi^{A\tilde{A}} e^{iH^{A}t} \right)$$
$$= e^{-iH^{A}t} J_{\mathcal{E}}^{AB} e^{iH^{A}t}, \tag{66}$$

where in the first equality we use the fact that $|\Phi^{A\tilde{A}}\rangle = 1/\sqrt{|A|} \sum_{x=1}^{|A|} |x\rangle^A |x\rangle^{\tilde{A}}$ (here $\{|x\rangle^A\}$ and $\{|x\rangle^{\tilde{A}}\}$ are eigenbases of H_A and $H_{\tilde{A}}$, respectively) has the property that

$$e^{-iH^{\tilde{A}}t} |\Phi^{A\tilde{A}}\rangle = e^{-iH^{A}t} |\Phi^{A\tilde{A}}\rangle.$$
(67)

Hence, by equating Eq. (65) with Eq. (66) we get that in the Choi representation the condition on \mathcal{E} in Eq. (10) is equivalent to

$$e^{-iH^{A}t} \otimes e^{iH^{B}t} J_{\mathcal{E}}^{AB} e^{iH^{A}t} \otimes e^{-iH^{B}t} = J_{\mathcal{E}}^{AB}.$$
(68)

Now, substituting into the above equation $J_{\mathcal{E}}^{AB} = \sum_{x,x',y,y'} c_{xyx'y'} |x\rangle \langle x'| \otimes |y\rangle \langle y'|$ (where $c_{xyx'y'}$ are some

coefficients) gives

$$c_{xyx'y'}e^{i(-a_x+a_{x'}+b_y-b_{y'})t} = c_{xyx'y'} \quad \forall \ t \in \mathbb{R}.$$
 (69)

Hence, $c_{xyx'y'} = 0$ unless

$$a_x - b_y = a_{x'} - b_{y'}.$$
 (70)

Combining this with the fact that H^A and H^B are relatively nondegenerate we get that $c_{xyx'y'} = 0$ unless x = x' and y = y'. Hence, the Choi matrix $J_{\mathcal{E}}^{AB}$ can be expressed as

$$J_{\mathcal{E}}^{AB} = \sum_{x,y} c_{xyxy} |x\rangle \langle x|^{A} \otimes |y\rangle \langle y|^{B}$$
(71)

so that $\mathcal{E}^{A \to B}$ is a classical channel. This completes the proof.

We consider now the interesting case in which A = B. In this case, we have in particular $H^A = H^B$ so we cannot apply the result above to this case.

Definition III.2: Let H^A be the Hamiltonian of a system A with energy eigenvalues $\{a_x\}_{x=1}^m$. We say that H^A has a nondegenerate Bohr spectrum if it has the property that for any $x, y, x', y' \in [m]$

$$a_x - a_y = a_{x'} - a_{y'} \iff x = x' \text{ and } y = y'$$

or $x = y \text{ and } x' = y';$

that is, there are no degeneracies in the nonzero differences of the energy levels of H^A .

Observe that almost all Hamiltonians have a nondegenerate Bohr spectrum (i.e., the set of all Hamiltonians that do not have a nondegenerate Bohr spectrum is of measure zero). Therefore, the results below that involves the Hamiltonian with a nondegenerate Bohr spectrum will apply to almost all systems. Such time-translation covariant channels with respect to nondegenerate Bohr spectrums have the following characterization.

Lemma III.1: Let H^A be a Hamiltonian with a nondegenerate Bohr spectrum, $\mathcal{E} \in \text{CPTP}(A \to A)$, and m := |A|. Then, $\mathcal{E} \in \text{COV}(A \to A)$ if and only if there exists a conditional probability distribution $\{p_{y|x}\}_{x,y\in[m]}$, and an $m \times m$ positive semidefinite matrix Q (with components denoted as $\{q_{xy}\}_{x,y\in[m]}$) whose diagonal components are $q_{xx} = p_{x|x}$ (for all $x \in [m]$) such that the Choi matrix of $\mathcal{E}^{A \to A}$ is given by

$$J_{\mathcal{E}}^{A\tilde{A}} = \sum_{\substack{x,y \in [m] \\ y \neq y \\ mathbf{x},y \in [m]}} p_{y|x} |x\rangle \langle x|^{A} \otimes |y\rangle \langle y|^{\tilde{A}} + \sum_{\substack{x \neq y \\ mathbf{x},y \in [m]}} q_{xy} |x\rangle \langle y|^{A} \otimes |x\rangle \langle y|^{\tilde{A}}.$$
(72)

Proof. Following the same lines as in Theorem III.1, by replacing H^B with H^A everywhere, we get that a quantum channel $\mathcal{E} \in \text{CPTP}(A \to A)$ is time-translation covariant if and only if its Choi matrix $J_{\mathcal{E}}^{A\tilde{A}} = \sum_{x,x',y,y'} c_{xyx'y'} |x\rangle \langle x'| \otimes |y\rangle \langle y'|$ satisfies $c_{xyx'y'} = 0$ unless

$$a_x - a_y = a_{x'} - a_{y'}.$$
 (73)

Since H^A is generic (i.e., has a nondegenerate Bohr spectrum) we get that the Choi matrix $J_{\mathcal{E}}^{A\tilde{A}}$ corresponds to a time-translation covariant channel if and only if $c_{xyx'y'} = 0$ unless x = x' and y = y', or x = y and x' = y'. Denoting by $p_{y|x} := c_{xyxy}$ and $q_{xy} := c_{xxyy}$ we conclude that $\mathcal{E} \in CPTP(A \to A)$ is a time-translation covariant channel if and only if $J_{\mathcal{E}}^{A\tilde{A}'}$ has the form Eq. (72). Since $J_{\mathcal{E}}^{A\tilde{A}} \ge 0$ we have, in particular, that each $p_{y|x} \ge 0$, and the condition that the marginal $J^A = I^A$ implies that $\sum_y p_{y|x} = 1$ for all $x = 1, \ldots, m$. Note that the two terms on the rhs of Eq. (72) have orthogonal support. Therefore, $J^{AB} \ge 0$ if and only if both $p_{y|x} \ge 0$ for all x and y, and $Q \ge 0$. This completes the proof.

Remark: Observe that even if the spectrum of the Hamiltonian H^A has degeneracies, any quantum channel $\mathcal{E} \in \text{CPTP}(A \to A)$ whose Choi matrix has the form Eq. (72) is necessarily time-translation covariant. Therefore, several of the results below will also be useful for Hamiltonians with degenerate spectrum.

B. Exact interconversions

In this subsection we consider the exact conversion of one state to another under time-translation covariant channels. Specifically, let $\{|x\rangle^A\}_{x\in[m]}$ be the energy eigenbasis of a Hamiltonian H^A , and let

$$\rho^{A} = \sum_{x,x' \in [m]} r_{xx'} |x\rangle \langle x'|^{A} \text{ and } \sigma^{A} = \sum_{x,x' \in [m]} s_{xx'} |x\rangle \langle x'|^{A}$$
(74)

be two density matrices in $\mathfrak{D}(A)$ with components $\{r_{xx'}\}$ and $\{s_{xx'}\}$, respectively.

Theorem III.2: Let $\rho, \sigma \in \mathfrak{D}(A)$ be as in Eq. (74) and suppose that $r_{xx'} \neq 0$ for all $x, x' \in [m]$, and the Hamiltonian H^A has a nondegenerate Bohr spectrum. Then, the following statements are equivalent:

- 1. There exists $\mathcal{E} \in \text{COV}(A \to A)$ such that $\sigma = \mathcal{E}(\rho)$.
- 2. The $m \times m$ matrix Q, with components

$$q_{xy} := \begin{cases} \min\left\{1, \frac{s_{xx}}{r_{xx}}\right\} & if x = y\\ \frac{s_{xy}}{r_{xy}} & otherwise, \end{cases}$$
(75)

is positive semidefinite.

Moreover, the second statement implies the first statement even if the Hamiltonian H^A has a degenerate Bohr spectrum.

Remark: In the proof below we see that if $r_{xy} = 0$ for some off-diagonal terms (i.e., $x \neq y$) then s_{xy} must also be zero. However, in this case, we see that for any $x \neq y \in [m]$ with $r_{xy} = 0$, the components of q_{xy} can be arbitrary. This means that in this case the condition becomes cumbersome, as we need Q to exist as defined above but with no restriction on the components q_{xy} for which $r_{xy} = 0$.

Proof. From Lemma III.1 it follows that there exists $\mathcal{E} \in \text{COV}(A \to A)$ such that $\sigma = \mathcal{E}(\rho)$ if and only if there exists a conditional probability distribution $\{p_{y|x}\}$, and an $m \times m$ positive semidefinite matrix Q, such that

$$\sigma = \mathcal{E}(\rho) = \operatorname{Tr}_{A} \left[J_{\mathcal{E}}^{A\tilde{A}}(\rho^{T} \otimes I^{\tilde{A}}) \right]$$
$$= \sum_{x,y} p_{y|x} r_{xx} |y\rangle \langle y| + \sum_{x \neq y} q_{xy} r_{xy} |x\rangle \langle y|. \quad (76)$$

That is, $\sigma = \mathcal{E}(\rho)$ if and only if

$$s_{yy} = \sum_{x=1}^{m} p_{y|x} r_{xx} \quad \forall y \in [m] \text{ and}$$

$$s_{xy} = q_{xy} r_{xy} \quad \forall x \neq y \in [m].$$
(77)

Hence, for the off-diagonal terms, $s_{xy} = 0$ whenever $r_{xy} = 0$. Since we assume that all the off-diagonal terms of ρ are nonzero, i.e., $r_{xy} \neq 0$ for $x \neq y$, there is no freedom left in the choice of the off-diagonal terms of Q and we must have $q_{xy} = s_{xy}/r_{xy}$. Since Q must be positive semidefinite we maximize its diagonal terms $\{p_{x|x}\}_{x=1}^{m}$ given the constraint that $s_{yy} = \sum_{x=1}^{m} p_{y|x}r_{xx}$. This constraint immediately gives $s_{yy} \geq p_{y|y}r_{yy}$ so that we must have $p_{y|y} \leq s_{yy}/r_{yy}$. Clearly, we also have $p_{y|y} \leq 1$ so we conclude that

$$p_{y|y} \leqslant \min\left\{1, \frac{s_{yy}}{r_{yy}}\right\}.$$
(78)

Remarkably, this condition is sufficient since there exists conditional probabilities $\{p_{y|x}\}$, with both $p_{y|y} = \min\{1, s_{yy}/r_{yy}\}$ and $s_{yy} = \sum_{x=1}^{m} p_{y|x}r_{xx}$. Indeed, for simplicity set $r_x := r_{xx}$ and $s_x := s_{xx}$, and define

$$p_{y|x} := \begin{cases} \min\left\{1, \frac{s_x}{r_x}\right\} & \text{if } x = y\\ \frac{1}{\mu r_x}(s_y - r_y)_+(r_x - s_x)_+ & \text{otherwise} \end{cases}$$
(79)

where

$$\mu := \sum_{y \in [m]} (s_y - r_y)_+ = \frac{1}{2} \|\mathbf{s} - \mathbf{r}\|_1,$$
(80)

and we use the notation $(s_y - r_y)_+ := s_y - r_y$ if $s_y \ge r_y$ and $(s_y - r_y)_+ := 0$ if $s_y < r_y$. Clearly, $p_{y|x} \ge 0$, and it Finally, if H^A has a degenerate Bohr spectrum and $Q \ge 0$ then we still get that the Choi matrix of the form Eq. (72) [with $p_{y|x}$ as in Eq. (79) and q_{xy} as in Eq. (75)] corresponds to a quantum channel $\mathcal{E} \in \text{CPTP}(A \to A)$ with the property that $\sigma = \mathcal{E}(\rho)$. As discussed below the proof of Lemma III.1, all channels with a Choi matrix of the form Eq. (72) are time-translation covariant. Hence, $\mathcal{E} \in \text{COV}(A \to A)$. This completes the proof.

Remark: In the proof above we saw that if $r_{xy} = 0$ for some $x \neq y$ then $\sigma = \mathcal{E}(\rho)$ for some $\mathcal{E} \in \text{COV}(A \to A)$ only if $s_{xy} = 0$. This in particular implies that if ρ has a block diagonal form $\rho = (\tilde{\rho} \ 0 \ 0)$, and if it can be converted by a time-translation covariant channel to σ , then σ must have the form $\sigma = (\tilde{\rho} \ 0 \ D)$ where *D* is some diagonal matrix.

As an example for the theorem above, consider the qubit case in which both

$$\rho = \begin{pmatrix} a & z \\ \bar{z} & 1-a \end{pmatrix} \quad \text{and} \quad \sigma = \begin{pmatrix} b & w \\ \bar{w} & 1-b \end{pmatrix}$$

are qubit states. Without loss of generality suppose that $a \ge b$ [we can always rearrange the order of the diagonals of ρ and σ by a permutation in COV $(A \rightarrow A)$]. In this case the matrix Q can be expressed as

$$Q = \begin{pmatrix} \frac{b}{a} & \frac{w}{z} \\ \frac{\bar{w}}{\bar{z}} & 1 \end{pmatrix}.$$
 (81)

Therefore, $Q \ge 0$ if and only if

$$\frac{b}{a} \ge \left|\frac{w}{z}\right|^2. \tag{82}$$

Observe that if ρ is a pure state, so that $|z| = \sqrt{a(1-a)}$, then the above equation holds if and only if $|w|^2 \leq b(1-a)$. Now, since $\sigma \geq 0$ we have $|w|^2 \leq b(1-b)$ so that $1 - \frac{|w|^2}{b} \geq b$. Therefore, for any *a* in the range

$$a \in \left[b, 1 - \frac{|w|^2}{b}\right] \tag{83}$$

we get both $|w|^2 \leq b(1-a)$ and $a \geq b$. That is, for any mixed state σ there exists a pure state ψ that can be converted to σ . On the other hand, if σ is pure [i.e., $|w|^2 = b(1-b)$] and ρ arbitrary qubit, then the condition

in Eq. (82) becomes

$$|z|^2 \ge a(1-b). \tag{84}$$

Since $\rho \ge 0$ we also have $|z|^2 \le b(1-b)$. Combining both inequalities we find that the only way ρ can be converted to a pure qubit state σ is if b = a (since $a \ge b$ is the initial assumption) and $|z|^2 = a(1-a)$. That is, ρ is a pure state itself, and up to a diagonal unitary is equal to σ . Hence, pure coherence cannot be obtained from mixed coherence, and deterministic interconversion among inequivalent pure resources is not possible.

The example above shows that there is no unique "golden unit" that can be used as the ultimate resource in two-dimensional systems. Instead, any pure resource (i.e., pure state that is not an energy eigenstate) is maximal in the sense that there is no other resource that can be converted into it (up to the equivalence class of diagonal unitaries). However, the set of all pure qubit resources is maximal (i.e., any mixed state can be reached from some pure state by time-translation covariant operations). We now show that this latter property holds in general.

Corollary III.1: Let $\sigma \in \mathfrak{D}(A)$ be an arbitrary state, and denote by $p_x := \langle x | \sigma | x \rangle$ the diagonal elements of σ in the energy eigenbasis $\{|x\rangle\}_{x=1}^m$ of system A. Then, the pure quantum state

$$|\psi\rangle := \sum_{x=1}^{m} \sqrt{p_x} |x\rangle \tag{85}$$

can be converted to σ by a time-translation covariant channel.

Proof. Observe that the diagonal elements Q are all 1, and the off-diagonal terms are given by

$$q_{xy} = \frac{s_{xy}}{\sqrt{p_x p_y}} \qquad \forall x, y \in [m], \ x \neq y.$$
(86)

Therefore, we can express $Q = D_{\mathbf{p}}^{-1} \sigma D_{\mathbf{p}}^{-1}$, where $D_{\mathbf{p}}$ is the diagonal matrix whose diagonal is $(\sqrt{p_1}, \dots, \sqrt{p_m})$. Since $D_{\mathbf{p}} > 0$ and $\sigma \ge 0$ it follows that $Q \ge 0$.

IV. QUANTUM ATHERMALITY IN THE SINGLE-SHOT REGIME

In Sec. III A we saw that if A and B are two physical systems with relatively nondegenerate Hamiltonians, then a quantum channel $\mathcal{N} \in \text{CPTP}(A \rightarrow B)$ is time-translation covariant if and only if it is classical. Since thermal operations are time-translation covariant, it follows that for relatively nondegenerate Hamiltonians thermal operations must be classical. This observation has the following consequence.

Corollary IV.1: Let \mathfrak{F} be either CTO or GPC, and let *A* and *B* be two physical systems with relatively nondegenerate Hamiltonians. Let (ρ^A, γ^A) and (σ^B, γ^B) be two athermality states on system *A* and *B*, and \mathbf{r}^A , \mathbf{s}^B , \mathbf{g}^A , and \mathbf{g}^B , be the probability vectors whose components are the elements on the diagonals of ρ^A , σ^B , γ^A , and γ^B , respectively. Then, the following are equivalent:

1.
$$(\rho^{A}, \gamma^{A}) \xrightarrow{\$} (\sigma^{B}, \gamma^{B}).$$

2. σ^{B} is diagonal in the energy eigenbasis and $(\mathbf{r}^{A}, \mathbf{g}^{A}) \succ (\mathbf{s}^{B}, \mathbf{g}^{B}).$

Remark: Note that in the generic case of relatively nondegenerate Hamiltonians, GPC can destroy only the coherence between the energy levels of the input state ρ^A . In this case, coherence cannot be manipulated, but only destroyed.

Consider the conversion of one athermality state (ρ^A, γ^A) to another athermality state (σ^B, γ^B) under any of the free operations, \mathfrak{F} , discussed above. Such a conversion is equivalent to a conversion with the same input and output Gibbs states, since appending a Gibbs state is a reversible thermal operation. To see this explicitly, observe first that

$$\begin{pmatrix} \rho^{A}, \gamma^{A} \end{pmatrix} \stackrel{\mathfrak{F}}{\leftrightarrow} \begin{pmatrix} \rho^{A} \otimes \gamma^{B}, \gamma^{AB} \end{pmatrix},$$

$$(\sigma^{B}, \gamma^{B}) \stackrel{\mathfrak{F}}{\leftrightarrow} \begin{pmatrix} \gamma^{A} \otimes \sigma^{B}, \gamma^{AB} \end{pmatrix},$$

$$(87)$$

where \mathfrak{F} is one of the four sets TO, CTO, GPC, and GPO, and the symbol $\stackrel{\mathfrak{F}}{\Leftrightarrow}$ indicates conversion under \mathfrak{F} in both directions. Therefore, the conversion of (ρ^A, γ^A) to (σ^B, γ^B) is equivalent to the conversion of $(\rho^A \otimes \gamma^B, \gamma^{AB})$ to the state $(\gamma^A \otimes \sigma^B, \gamma^{AB})$. Note that the latter conversion has the same input and output Gibbs state γ^{AB} . Therefore, interconversions among states with the same Gibbs state (and, in particular, with |A| = |B|) is general enough to capture also interconversions with $|B| \neq |A|$ (as long as we do not impose some additional nondegeneracy constraints); see Fig. 1.

We now focus on interconversions among states that are all in $\mathfrak{D}(A)$, and unless necessary, will drop the system superscript A from the states. However, we assume that the Hamiltonian H^A has a nondegenerate Bohr spectrum. This will reduce a bit from the generality of the results, however, as discussed above, this is the generic case and almost all Hamiltonians having such a spectrum.

A. Exact conversions

Consider a conversion of the form $(\rho, \gamma) \xrightarrow{\text{GPC}} (\sigma, \gamma)$, where $\rho, \sigma, \gamma \in \mathfrak{D}(A)$, and *all* the off-diagonal terms of ρ are nonzero. In this case, Theorem III.2 states that ρ can be converted to σ by a time-translation covariant channel if and only if the matrix Q as defined in Eq. (75) is positive



FIG. 1. Equivalence of conversions. The top conversion with two different Gibbs states γ^A and γ^B is equivalent to the bottom conversion with the same Gibbs state γ^{AB} .

semidefinite. Since CGP channels are, in particular, covariant under the time-translation group, the condition $Q \ge 0$ is a necessary (but not sufficient) condition for $(\rho, \gamma) \xrightarrow{\text{GPC}} (\sigma, \gamma)$. To get the full necessary and sufficient conditions, let J^{AB} be the Choi matrix of a time-translation covariant channel $\mathcal{E} \in \text{COV}(A \to A)$ that satisfies $\mathcal{E}(\rho) = \sigma$ and $\mathcal{E}(\gamma) = \gamma$. Recall that the Choi matrix of such a channel has the form [cf. Eq. (72)]

$$J^{A\tilde{A}} = \sum_{x,y} p_{y|x} |x\rangle \langle x|^{A} \otimes |y\rangle \langle y|^{\tilde{A}} + \sum_{x \neq y} \frac{s_{xy}}{r_{xy}} |x\rangle \langle y|^{A} \otimes |x\rangle \langle y|^{\tilde{A}},$$
(88)

where $P = (p_{y|x})$ is some column stochastic matrix, and we assume that the off-diagonal terms of ρ are nonzero. Let **r** and **s** be the probability vectors consisting of the diagonals of ρ and σ , respectively, and identify the diagonal matrix γ with the Gibbs vector **g** consisting of its diagonal. Then, the Choi matrix above facilitates such a channel \mathcal{E} if and only if it is positive semidefinite *and*

$$P\mathbf{r} = \mathbf{s} \quad \text{and} \quad P\mathbf{g} = \mathbf{g}.$$
 (89)

Note that the above condition implies that $(\mathbf{r}, \mathbf{g}) \succ (\mathbf{s}, \mathbf{g})$, however, it is not sufficient since we also require that $J^{A\bar{A}} \ge 0$. This latter condition is equivalent to the requirement that the matrix obtained by replacing the diagonal elements of Q [as defined in Eq. (75)] with $\{p_{x|x}\}_{x \in [m]}$ is positive semidefinite. We summarize these considerations in the following lemma.

Lemma IV.1: Let (ρ^A, γ^A) and (σ^A, γ^A) be two athermality states of a system A, whose Hamiltonian H^A has a nondegenerate Bohr spectrum. Using the same notations as in Eq. (74), suppose that $r_{xy} \neq 0$ for all $x \neq y$. Then, the following statements are equivalent:

1. $(\rho^A, \gamma^A) \xrightarrow{\text{GPC}} (\sigma^A, \gamma^A).$

2. There exists a column stochastic matrix P that satisfies both Eq. (89) and the matrix

$$\sum_{x=1}^{m} p_{x|x} |x\rangle \langle x| + \sum_{x \neq y \in [m]} \frac{s_{xy}}{r_{xy}} |x\rangle \langle y| \ge 0.$$
(90)

Moreover, the second statement implies the first statement even if the Hamiltonian H^A has a degenerate Bohr spectrum.

The lemma above does not provide much computational simplification over the results in Ref. [27], since determining the existence of such a column stochastic matrix P is itself a semidefinite programming (SDP) problem. However, the significance of this lemma is that it makes the role of quantum coherence in such conversions of athermality much more apparent, as demonstrated by the following theorem. Moreover, we see below that in the qubit case the lemma above provides a simple criterion for exact interconversions under GPC.

Theorem IV.1: Let (ρ^A, γ^A) and (σ^A, γ^A) be two quantum athermality states of dimension m := |A|, whose Hamiltonian H^A has a nondegenerate Bohr spectrum. For any $x, y \in [m]$ let $r_{xy} := \langle x | \rho | y \rangle$ and $s_{xy} := \langle x | \sigma | y \rangle$ be, respectively, the xy component of ρ and σ in the energy eigenbasis. Suppose that $r_{xy} \neq 0$ for all $x, y \in [m]$ and that $r_{xx} = s_{xx}$ for all $x \in [m]$. Then, the following statements are equivalent:

1.
$$(\rho^{A}, \gamma^{A}) \xrightarrow{\text{GPC}} (\sigma^{A}, \gamma^{A}).$$

2. $Q^{A} := I^{A} + \sum_{x \neq y \in [m]} \frac{s_{xy}}{r_{yy}} |x\rangle \langle y|^{A} \ge 0.$

Moreover, the second statement implies the first statement even if the Hamiltonian H^A has a degenerate Bohr spectrum.

Remark: The condition in the theorem above that ρ and σ have the same diagonals means that ρ and σ have the same nonuniformity and they differ only by their coherence (asymmetry) properties. In fact, observe that the condition $Q^A \ge 0$ is identical to the condition given in Theorem III.2 for the case that the diagonals of ρ and σ are the same. Therefore, in this case we have $(\rho^A, \gamma^A) \xrightarrow{\text{CGP}} (\sigma^A, \gamma^A)$ if and only if ρ^A can be converted to σ^A by time-translation covariant operations. In particular, the Gibbs state, γ^A , does not play a role in such conversions since ρ and σ have the same nonuniformity (i.e., same diagonals).

Proof. Since the diagonals of ρ and σ are the same, we get that if $Q^A \ge 0$ then by taking the stochastic matrix P to be the identity matrix, all the conditions of Lemma IV.1 are satisfied so that $(\rho^A, \gamma^A) \xrightarrow{\text{CGP}} (\sigma^A, \gamma^A)$. Conversely, if

 $(\rho^A, \gamma^A) \xrightarrow{\text{CGP}} (\sigma^A, \gamma^A)$ then by Lemma IV.1 there exists a stochastic matrix P with a diagonal $\{p_{x|x}\}$ that satisfies Eq. (90). By adding the positive semidefinite matrix $\sum_{x=1}^{m} (1 - p_{x|x}) |x\rangle \langle x|$ to the matrix in Eq. (90) we get that also $Q^A \ge 0$. This completes the proof.

The theorem above has the following consequence.

Corollary IV.2: Let $\rho \in \mathfrak{D}(A)$ be an arbitrary state, and denote by $p_x := \langle x | \rho | x \rangle$ the diagonal elements of ρ in the energy eigenbasis $\{|x\rangle\}_{x=1}^m$ of system A. Then,

$$\left(\psi^{A},\gamma^{A}\right) \xrightarrow{\text{GPC}} \left(\rho^{A},\gamma^{A}\right)$$
 (91)

where $|\psi^A\rangle := \sum_{x=1}^m \sqrt{p_x} |x\rangle$.

Proof. Since ψ^A and ρ^A have the same diagonals, it follows from Theorem IV.1 and the discussion above that $(\psi^A, \gamma^A) \xrightarrow{\text{GPC}} (\rho^A, \gamma^A)$ if and only if ψ^A can be converted to ρ^A by time-translation covariant operations. The latter conversion is possible due to Corollary III.1. This completes the proof.

Lemma IV.1 can also be used to give the precise conditions for interconversions under GPC of qubit athermality states. For this purpose, let $\rho, \sigma, \gamma \in \mathfrak{D}(A)$ with |A| = 2. Denote

$$\rho = \begin{pmatrix} r & a \\ \bar{a} & 1-r \end{pmatrix}, \ \sigma = \begin{pmatrix} s & b \\ \bar{b} & 1-s \end{pmatrix}, \ \gamma = \begin{pmatrix} g & 0 \\ 0 & 1-g \end{pmatrix}.$$
(92)

We also denote the diagonals of the matrices above by $\mathbf{r} := (r, 1 - r)^T$, $\mathbf{s} := (s, 1 - s)^T$, and $\mathbf{g} = (g, 1 - g)^T$. We would like to find the conditions under which $(\rho, \gamma) \xrightarrow{\text{GPC}} (\sigma, \gamma)$. Recall that if a = 0 then we must have b = 0 since GPC cannot generate coherence between energy levels. Therefore, the case a = 0 has already been covered by the quasiclassical regime. Note also that the case g = 1/2 also corresponds to the quasiclassical case (since in this case ρ and σ can be diagonalized). We therefore assume in the rest of this subsection that $a \neq 0$ and $g \neq 1/2$.

Theorem IV.2: [cf. Ref. [45]] Let $\rho, \sigma, \gamma \in \mathfrak{D}(A)$ be three qubit states as above and suppose $a \neq 0$ and $g \neq 1/2$. Then, for $\mathbf{r} \neq \mathbf{g}$, $(\rho, \gamma) \xrightarrow{\text{GPC}} (\sigma, \gamma)$ if and only if $(\mathbf{r}, \mathbf{g}) \succ (\mathbf{s}, \mathbf{g})$ and

$$\frac{|b|^2}{|a|^2} \leqslant \frac{\det \begin{pmatrix} s & 1-r \\ g & 1-g \end{pmatrix} \det \begin{pmatrix} r & 1-s \\ g & 1-g \end{pmatrix}}{(r-g)^2}.$$
 (93)

For $\mathbf{r} = \mathbf{g}$, $(\rho, \gamma) \xrightarrow{\text{GPC}} (\sigma, \gamma)$ if and only if $\mathbf{s} = \mathbf{g}$ and $|a| \ge |b|$.

Remark: If s = g (but $r \neq g$) then the condition in Eq. (93) can be simplified. Specifically, in this case we get that (ρ, γ) can be converted to (σ, γ) by GPC if and only if

$$\frac{|b|^2}{|a|^2} \leqslant \det(\gamma). \tag{94}$$

Proof. From Lemma IV.1 it follows that (ρ, γ) can be converted to (σ, γ) by GPC if and only if there exists a 2 × 2 column stochastic matrix $P = \{p_{y|x}\}_{x,y \in \{0,1\}}$ that satisfies $P\mathbf{r} = \mathbf{s}, P\mathbf{g} = \mathbf{g}$, and

$$\begin{pmatrix} p_{0|0} & b/a \\ \bar{b}/\bar{a} & p_{1|1} \end{pmatrix} \geqslant 0.$$
(95)

Note that this last condition is equivalent to

$$\frac{|b|^2}{|a|^2} \leqslant p_{0|0} p_{1|1}. \tag{96}$$

The conditions $P\mathbf{r} = \mathbf{s}$ and $P\mathbf{g} = \mathbf{g}$ can be expressed as the following linear systems of equations:

$$\begin{bmatrix} r & 1-r \\ g & 1-g \end{bmatrix} \begin{bmatrix} p_{0|0} \\ p_{0|1} \end{bmatrix} = \begin{bmatrix} s \\ g \end{bmatrix}$$
(97)

and

$$\begin{bmatrix} r & 1-r \\ g & 1-g \end{bmatrix} \begin{bmatrix} p_{1|0} \\ p_{1|1} \end{bmatrix} = \begin{bmatrix} 1-s \\ 1-g \end{bmatrix}.$$
 (98)

Note that the equations involving $p_{1|0}$ and $p_{1|1}$ follow trivially from the ones involving $p_{0|0}$ and $p_{0|1}$ since *P* is column stochastic. From Cramer's rule it then follows that for the case that $r \neq g$

$$p_{0|0} = \frac{\det \begin{pmatrix} s & 1-r \\ g & 1-g \end{pmatrix}}{\det \begin{pmatrix} r & 1-r \\ g & 1-g \end{pmatrix}} \quad \text{and} \quad p_{1|1} = \frac{\det \begin{pmatrix} r & 1-s \\ g & 1-g \end{pmatrix}}{\det \begin{pmatrix} r & 1-r \\ g & 1-g \end{pmatrix}}.$$
(99)

Finally, substituting the above expression in Eq. (96) gives Eq. (93).

For the case that r = g we also have s = g [otherwise, $(\mathbf{r}, \mathbf{g}) \neq (\mathbf{s}, \mathbf{g})$] and the linear system of equations in Eq. (97) has a unique solution given by $p_{0|0} = p_{1|1} =$ 1. Therefore, in this case, Eq. (96) gives $|b| \leq |a|$. This completes the proof.

From the remark below Theorem IV.2 it follows that already in the qubit case, conversions under GPC have a certain type of discontinuity. To see this, consider the case s = g, and observe that the condition $|a|^2 \det(\gamma) \ge |b|^2$ is stronger than the condition $|a| \ge |b|$ that one obtains if also r = g. In particular, since $det(\gamma) \leq 1/4$, there exists an $\varepsilon > 0$ and $\rho, \sigma, \gamma \in \mathfrak{D}(A)$ such that for any $\rho \in \mathfrak{B}^{\varepsilon}(\sigma)$ [here $\mathfrak{B}^{\varepsilon}(\sigma)$ is the ball of all density matrices that are ε close to σ] we have that (ρ, γ) cannot be converted by GPC to (σ, γ) unless $\rho = \sigma$ (up to free diagonal unitary). As an explicit example, let

$$\sigma = \frac{1}{6} \begin{pmatrix} 2 & \sqrt{2} \\ \sqrt{2} & 4 \end{pmatrix} \quad \text{and} \quad \gamma = \frac{1}{3} \begin{pmatrix} 1 & 0 \\ 0 & 2 \end{pmatrix}. \quad (100)$$

According to the theorem above, in this example, $(\rho, \gamma) \xrightarrow{\text{GPC}} (\sigma, \gamma)$ if and only if either $\rho = \sigma$ or

$$a \geqslant \frac{b}{\sqrt{\det(\gamma)}} = \frac{1}{2}.$$
 (101)

However, note that for sufficiently small $\varepsilon > 0$ the condition $\rho \in \mathfrak{B}^{\varepsilon}(\sigma)$ would imply that *a* cannot be too far away from $b = \sqrt{2}/6 < 1/2$. Therefore, for sufficiently small $\varepsilon > 0$ the condition $\rho \in \mathfrak{B}^{\varepsilon}(\sigma)$ implies that $(\rho, \gamma) \xrightarrow{\text{GPC}} (\sigma, \gamma)$ if and only if $\rho = \sigma$ (up to a free diagonal unitary).

B. Approximate single-shot conversions

For the case that $\mathfrak{F} = GPC$, the conversion distance given in Eq. (42) can be expressed as

$$d_{\mathfrak{F}}((\rho^{A},\gamma^{A})\to(\sigma^{A'},\gamma^{A'}))$$

$$:=\min_{\mathcal{E}\in\mathrm{COV}(A\to A')}\left\{\frac{1}{2}\|\sigma-\mathcal{E}(\rho)\|_{1} : \gamma^{A'}=\mathcal{E}(\gamma^{A})\right\}.$$
(102)

Since the trace distance between two density matrices can be expressed as

$$\frac{1}{2} \|\sigma - \mathcal{E}(\rho)\|_1 = \min_{\substack{\Lambda \in \operatorname{Pos}(\mathcal{A}')\\\Lambda \geqslant \sigma - \mathcal{E}(\rho)}} \operatorname{Tr}[\Lambda], \qquad (103)$$

the conversion distance can be expressed as the following minimization problem:

$$d_{\mathfrak{F}}((\rho,\gamma) \to (\sigma,\gamma)) = \min \operatorname{Tr}[\Lambda] \qquad (104)$$

subject to

1.
$$\Lambda^{A'} \ge \sigma^{A'} - \operatorname{Tr}_A \left[J^{AA'} \left(\rho^T \otimes I^{A'} \right) \right].$$

2. $J^A = I^A.$
3. $\gamma^{A'} = \operatorname{Tr}_A \left[J^{AA'} \left(\gamma^A \otimes I^{A'} \right) \right].$
4. $\left[J^{AA'}, e^{-iH^A t} \otimes e^{iH^{A'} t} \right] = 0$ for all $t \in \mathbb{R}.$
5. $\Lambda \in \operatorname{Pos}(A')$ and $J \in \operatorname{Pos}(AA').$

The optimization problem above can be solved efficiently with semidefinite programs, however, when the output state is quasiclassical the conversion distance takes a much simpler form.

Consider the conversion distance from an arbitrary state (ρ^A, γ^A) to a quasiclassical state $(\sigma^{A'}, \gamma^{A'})$. In this case, a channel $\mathcal{E} \in \text{CPTP}(A \to A')$ is time-translation covariant if and only if $\mathcal{E} \circ \mathcal{P} = \mathcal{E}$ since the output of the channel is classical (and therefore time-translation invariant). Therefore, in this case we get

$$d_{\text{GPC}}\left((\rho^{A}, \gamma^{A}) \to (\sigma^{A'}, \gamma^{A'})\right)$$

$$\coloneqq \min_{\mathcal{E} \in \text{COV}(A \to A')} \left\{ \frac{1}{2} \| \sigma - \mathcal{E}(\rho) \|_{1} : \gamma^{A'} = \mathcal{E}(\gamma^{A}) \right\}$$

$$\geqslant \min_{\mathcal{E} \in \text{COV}(A \to A')} \left\{ \frac{1}{2} \| \mathcal{P}(\sigma - \mathcal{E}(\rho)) \|_{1} : \gamma^{A'} = \mathcal{E}(\gamma^{A}) \right\}$$

$$= \min_{\mathcal{E} \in \text{COV}(A \to A')} \left\{ \frac{1}{2} \| \sigma - \mathcal{E} \circ \mathcal{P}(\rho) \|_{1} : \gamma^{A'} = \mathcal{E}(\gamma^{A}) \right\}$$

$$\geqslant d_{\text{GPO}}\left((\mathcal{P}(\rho^{A}), \gamma^{A}) \to (\sigma^{A'}, \gamma^{A'})\right).$$
(105)

On the other hand, we have

$$d_{\text{GPO}}\left(\left(\mathcal{P}(\rho^{A}), \gamma^{A}\right) \rightarrow \left(\sigma^{A'}, \gamma^{A'}\right)\right)$$

$$= \min_{\mathcal{E} \in \text{CPTP}(A \rightarrow A')} \left\{ \frac{1}{2} \|\sigma - \mathcal{E} \circ \mathcal{P}(\rho)\|_{1} : \gamma^{A'} = \mathcal{E}\left(\gamma^{A}\right) \right\}$$

$$\geqslant \min_{\mathcal{E} \in \text{CPTP}(A \rightarrow A')} \left\{ \frac{1}{2} \|\sigma - \mathcal{P} \circ \mathcal{E} \circ \mathcal{P}(\rho)\|_{1} : \gamma^{A'} = \mathcal{E}\left(\gamma^{A}\right) \right\}$$

$$\geqslant \min_{\mathcal{E} \in \text{COV}(A \rightarrow A')} \left\{ \frac{1}{2} \|\sigma - \mathcal{E}(\rho)\|_{1} : \gamma^{A'} = \mathcal{E}\left(\gamma^{A}\right) \right\}$$

$$= d_{\text{GPC}}\left(\left(\rho^{A}, \gamma^{A}\right) \rightarrow \left(\sigma^{A'}, \gamma^{A'}\right)\right). \quad (106)$$

Therefore, combining the two expressions above we get that

$$d_{\text{GPC}}\left((\rho^{A}, \gamma^{A}) \to (\sigma^{A'}, \gamma^{A'})\right)$$
$$= d_{\text{GPO}}\left((\mathcal{P}(\rho^{A}), \gamma^{A}) \to (\sigma^{A'}, \gamma^{A'})\right).$$
(107)

One can then use the expression given in Ref. [42] for the conversion distance between two quasiclassical states.

The above observation can be used to get an exact closed formula for the ε -single-shot distillable athermality defined on any quantum athermality state (ρ, γ) as in Eq. (43) with $\mathfrak{F} = \text{GPC}$. Note that since the golden unit $(|0\rangle\langle 0|^X, \mathbf{u}_n^X)$ that appears in Eq. (43) is quasiclassical, it cannot be used to define the single-shot cost of an arbitrary quantum athermality state since quasiclassical states cannot be converted by GPC to states with coherence between

energy levels. Therefore, in this subsection we consider only single-shot distillation of athermality.

Theorem IV.3: Let $\rho, \gamma \in \mathfrak{D}(A)$ and $\varepsilon \in [0, 1]$. Then, the ε -single-shot distillable athermality of the state (ρ, γ) is given by

$$\text{Distill}_{\text{GPC}}^{\varepsilon}\left(\rho,\gamma\right) = D_{\min}^{\varepsilon}\left(\mathcal{P}(\rho) \| \gamma\right). \tag{108}$$

Proof. The relation in Eq. (107) immediately implies that

Distill^{\varepsilon}_{GPC} (\(\rho\), \(\cap\)) = log sup

$$\left\{m : d_{GPO}\left((\mathcal{P}(\rho), \(\cap\)) \to (|0\rangle\langle 0|, \mathbf{u}_m)\right) \leqslant \varepsilon\right\}$$

$$= \text{Distill}_{GPO}^\varepsilon (\mathcal{P}(\rho), \(\cap\)). (109)$$

Therefore, combining this with the first equality of Eq. (50) yields Eq. (108). This completes the proof.

V. QUANTUM ATHERMALITY IN THE ASYMPTOTIC REGIME

Unlike Gibbs preserving operations, both thermal operations and GPC operations cannot generate coherence between energy levels. This means that any number of copies of the golden unit ($|0\rangle\langle 0|$, **u**) cannot be converted even to a single copy of an athermality state (ρ^A, γ^A) that exhibits coherence between energy levels. However, it turns out that this type of irreversibility between the (infinite) cost to prepare the state (ρ^A, γ^A) versus the finite rate at which it can be used to distill golden units of athermality, can be removed if we allow for a relatively small amount of coherence to be added to the system.

This section is organized as follows. We start by showing that the distillable athermality of (ρ^A, γ^A) is equal to the Umegaki relative entropy (historically, a version of this result was first proved in Ref. [10], however, our proof has a pedagogical value as it is relatively short and is based on the single-shot regime). We then introduce a few new concepts, such as asymptotic scaling, energy spread, and sublinear athermality resources, in order to show how reversibility can be restored by appending the free operations with resources that are asymptotically negligible.

A. Distillation of quantum athermality

The formula given in Eq. (108) for the ε -single-shot distillable athermality can be used to derive the asymptotic distillable athermality. Specifically, we have

$$\text{Distill}_{\text{GPC}}\left(\rho,\gamma\right) = \lim_{\varepsilon \to 0^{+}} \limsup_{n \to \infty} \frac{1}{n} D_{\min}^{\varepsilon} \left(\mathcal{P}_{n}(\rho^{\otimes n}) \left\|\gamma^{\otimes n}\right).$$
(110)

We now use this observation, and obtain a closed formula for the rhs of the equation above.

Theorem V.1: (cf. Ref. [10]) Let (ρ, γ) be an athermality state of a quantum system A. Then,

$$\text{Distill}_{\mathfrak{F}}(\rho, \gamma) = D(\rho \| \gamma). \tag{111}$$

where *D* is the Umegaki relative entropy, and \mathfrak{F} stands for either TO, CTO, or GPC.

Proof. Since $\mathcal{P}(\gamma) = \gamma$ we get from Eq. (108) and the data processing inequality (DPI) that

$$Distill_{GPC}^{\varepsilon}(\rho,\gamma) = D_{\min}^{\varepsilon} \left(\mathcal{P}(\rho) \| \mathcal{P}(\gamma) \right)$$
$$DPI \to \leq D_{\min}^{\varepsilon} \left(\rho \| \gamma \right),$$
(112)

and consequently

$$Distill_{GPC}(\rho, \gamma) \leq \lim_{\varepsilon \to 0^+} \limsup_{n \to \infty} \frac{1}{n} D_{\min}^{\varepsilon} \left(\rho^{\otimes n} \| \gamma^{\otimes n} \right)$$
$$= D(\rho \| \gamma), \qquad (113)$$

where the equality follows from the quantum Stein's lemma. Since TO and CTO are subsets of GPC the above inequality also holds if we replace GPC with TO or CTO.

To get the opposite inequality, fix $k \in \mathbb{N}$ and apply the pinching channel $\mathcal{P}_k \in \text{CTO}(A^k \to A^k)$ on k copies of ρ . The resulting state, $\sigma_k := \mathcal{P}_k(\rho^{\otimes k})$, is quasiclassical. Now, from Eq. (46) we get

$$\begin{aligned} \text{Distill}_{\text{TO}}\left(\rho,\gamma\right) &\geq \frac{1}{k} \text{Distill}_{\text{TO}}\left(\rho^{\otimes k},\gamma^{\otimes k}\right) \\ &\geq \frac{1}{k} \text{Distill}_{\text{TO}}\left(\sigma_{k},\gamma^{\otimes k}\right), \end{aligned} \tag{114}$$

where in the second inequality we use the fact that the pinching channel is a thermal operation (see Lemma II.2) so that by definition, σ_k cannot have a higher distillable rate than $\rho^{\otimes k}$. Since σ_k is a quasiclassical state we have that $\text{Distill}_{\text{TO}}(\sigma_k, \gamma^{\otimes k})$ equals $D(\sigma_k || \gamma^{\otimes k})$. Therefore, the equation above gives

$$\text{Distill}_{\text{TO}}\left(\rho,\gamma\right) \geqslant \frac{1}{k} D\left(\mathcal{P}_{k}\left(\rho^{\otimes k}\right) \|\gamma^{\otimes k}\right).$$
(115)

Now, since the above inequality holds for all $k \in \mathbb{N}$ we conclude that

Distill_{TO}
$$(\rho, \gamma) \ge \limsup_{k \to \infty} \frac{1}{k} D\left(\mathcal{P}_k\left(\rho^{\otimes k}\right) \| \gamma^{\otimes k}\right)$$
 (116)
(61) $\to = D(\rho \| \gamma).$

This completes the proof.

B. Scaling of time-translation asymmetry

In this subsection we show that the coherence of *n* copies of a states grows logarithmically with *n*. Let *A* be a physical system with Hamiltonian $H \in Pos(A)$ and a state $\psi \in PURE(A)$ given by

$$H^{A} = \sum_{x=1}^{m} a_{x} |x\rangle \langle x|$$
 and $|\psi\rangle = \sum_{x=1}^{m} \sqrt{p_{x}} |x\rangle$,

where m = |A|. For any $n \in \mathbb{N}$, the state $\psi^{\otimes n}$ has the form

$$|\psi\rangle^{\otimes n} = \sum_{x^n \in [m]^n} \sqrt{p_{x^n}} |x^n\rangle$$
$$= \sum_{x^n \in [m]^n} 2^{-\frac{n}{2} \left(H(\mathbf{t}(x^n)) + D(\mathbf{t}(x^n) \| \mathbf{p}) \right)} |x^n\rangle$$
(117)

where we use (5). For any type $\mathbf{t} \in \text{Type}(n, m)$ define

$$|\mathbf{t}\rangle^{A^n} := \frac{1}{\binom{n}{n_{t_1,\dots,nt_m}}^{1/2}} \sum_{x^n \in x^n(\mathbf{t})} |x^n\rangle, \qquad (118)$$

where the sum runs over all sequences x^n of the same type **t**. With the above notations

$$|\psi\rangle^{\otimes n} = \sum_{\mathbf{t}\in \mathrm{Type}(n,m)} \sqrt{r_{\mathbf{t},n}} |\mathbf{t}\rangle^{A^n}, \qquad (119)$$

where

$$r_{\mathbf{t},n} := |x^n(\mathbf{t})| 2^{-n\left(H(\mathbf{t}) + D(\mathbf{t} \parallel \mathbf{p})\right)}.$$
 (120)

Note that the vectors $|\mathbf{t}\rangle^{A^n}$ are eigenvectors of the Hamiltonian of system A^n . Specifically,

$$H^{\otimes n}|\mathbf{t}\rangle^{A^n} = n \sum_{x=1}^m t_x a_x |\mathbf{t}\rangle^{A^n}, \qquad (121)$$

so that the energy in the state $|\mathbf{t}\rangle^{A^n}$ is *n* times the average energy with respect to the type **t**. In the generic case, the energy eigenvalues $\{a_1, \ldots, a_m\}$ are rationally independent; in other words, for any set of *m* integers $\ell_1, \ldots, \ell_m \in \mathbb{Z}$ we have

$$\ell_1 a_1 + \dots + \ell_m a_m = 0 \iff \ell_1 = \dots = \ell_m = 0.$$
(122)

Under this mild assumption (which we do *not* assume, but is still worth mentioning), each type in Type(n,m) determines uniquely the energy of the system.

Since each $|\mathbf{t}\rangle^{A^n}$ is an energy eigenstate, it follows from Eq. (119) that we can always write $|\psi\rangle^{\otimes n}$ as a linear combination of $|\text{Type}(n,m)| \leq (n+1)^m$ energy eigenstates. In

other words, the coherence of $|\psi\rangle^{\otimes n}$ can be compressed into an $(n+1)^m$ dimensional vector (which is polynomial in *n*).

The observation above has the following consequence. In Corollary IV.2 we saw that for any mixed state in $\mathfrak{D}(A)$ there exists a pure state in PURE(*A*) that can be converted to it by GPC. Combining this with the above observation implies that the pure state coherence cost of preparing a state $\rho^{\otimes n} \in \mathfrak{D}(A^n)$ cannot exceed $m \log(n + 1)$, and specifically we have

$$C\left(\rho^{\otimes n}\right) \leqslant m\log(n+1),\tag{123}$$

where *C* is the coherence measure defined in Eq. (14). Therefore, the asymmetry cost rate, i.e., coherence cost per copy of ρ , cannot exceed $m(\log(n + 1))/n$, which goes to zero as $n \to \infty$. On the other hand, the nonuniformity cost (i.e., quasiclassical athermality cost) does *not* go to zero in the asymptotic limit since the energy of $\rho^{\otimes n}$ grows linearly with *n*.

To summarize, athermality comprises of two types of resources, namely nonuniformity (also known as informational nonequilibrium) and time-translation asymmetry (or coherence in short). Therefore, the athermality asymptotic cost of an athermality state also comprises of two types, nonuniformity cost and coherence cost. The latter however goes to zero in the asymptotic limit, and therefore needs to be rescaled differently. This makes the QRT of athermality very subtle, and consequently several important questions in the theory are still open (see Sec. VI).

C. The energy spread

The energy spread of a given pure state $\psi \in \text{PURE}(A)$ is defined as the difference between the maximal and minimal energies that appear when writing ψ as a superposition of energy eigenvectors. In the discussion above we saw that *n* copies of a state $\psi \in \text{PURE}(A)$ can be expressed as a linear combination of no more that $(n + 1)^m$ energy eigenvectors. Among these energy eigenvectors are the zero-energy eigenvector [corresponding to the type $\mathbf{t} = (1, 0, \dots, 0)^T$] and the maximal energy eigenvector [corresponding to the type $\mathbf{t} = (0, \dots, 0, 1)^T$]. Therefore, since the energy in the decomposition (119) spreads from zero to na_m (where a_m is the maximal energy of a single copy of system *A*), we conclude that the energy spread of $\psi^{\otimes n}$ is na_m .

The energy spread can be reduced drastically if one allows for a small deviation from the state $\psi^{\otimes n}$. Explicitly, for any $\varepsilon > 0$ we can split $|\psi^{\otimes n}\rangle$ into two parts

$$|\psi\rangle^{\otimes n} = \sum_{\mathbf{t}\in\mathfrak{S}_{n,\varepsilon}} \sqrt{r_{\mathbf{t},n}} |\mathbf{t}\rangle^{A^n} + \sum_{\mathbf{t}\in\mathfrak{S}_{n,\varepsilon}^c} \sqrt{r_{\mathbf{t},n}} |\mathbf{t}\rangle^{A^n}, \quad (124)$$

where

$$\mathfrak{S}_{n,\varepsilon} := \left\{ \mathbf{t} \in \operatorname{Type}(n,m) : \frac{1}{2} \|\mathbf{t} - \mathbf{p}\|_1 \leqslant \varepsilon \right\}, \quad (125)$$

and $\mathfrak{S}_{n,\varepsilon}^c$ is the complement of $\mathfrak{S}_{n,\varepsilon}$ in Type(n, m). By substituting the bounds in Eq. (8) into the definition of the coefficients $r_{t,n}$ we get that

$$\frac{1}{(n+1)^m} 2^{-nD(\mathbf{t}\|\mathbf{p})} \leqslant r_{\mathbf{t},n} \leqslant 2^{-nD(\mathbf{t}\|\mathbf{p})}.$$
 (126)

Therefore, the fidelity of $|\psi^{\otimes n}\rangle$ with the second term on the rhs of Eq. (124) is given by

$$\sum_{\mathbf{t}\in\mathfrak{S}_{n,\varepsilon}^{c}} r_{\mathbf{t},n} \leqslant \sum_{\mathbf{t}\in\mathfrak{S}_{n,\varepsilon}^{c}} 2^{-nD(\mathbf{t}\|\mathbf{p})}$$
Pinsker's inequality $\rightarrow \leqslant \sum_{\mathbf{t}\in\mathfrak{S}_{n,\varepsilon}^{c}} 2^{-2n\varepsilon^{2}}$

$$\leqslant 2^{-2n\varepsilon^{2}} |\operatorname{Type}(n,m)|$$
(6) $\rightarrow \leqslant 2^{-2n\varepsilon^{2}}(n+1)^{m}$

$$\xrightarrow{n\to\infty} 0.$$
(127)

That is, for any $\varepsilon > 0$ and sufficiently large *n*, the state $|\psi\rangle^{\otimes n}$ can be made arbitrarily close to the state

$$|\psi_{\varepsilon}^{n}\rangle := \frac{1}{\sqrt{\nu_{\varepsilon}}} \sum_{\mathbf{t}\in\mathfrak{S}_{n,\varepsilon}} \sqrt{r_{\mathbf{t},n}} |\mathbf{t}\rangle^{A^{n}}, \qquad (128)$$

where $\nu_{\varepsilon} := \sum_{\mathbf{t} \in \mathfrak{S}_{n,\varepsilon}} r_{\mathbf{t},n}$ is the normalization factor. Now, from Eq. (121) the energy of any state $|\mathbf{t}\rangle^{A^n}$ with type $\mathbf{t} \in \mathfrak{S}_{n,\varepsilon}$ is $\mu_{\mathbf{t}} := n \sum_{x=1}^{m} t_x a_x$. Expressing $\mathbf{t} = \mathbf{p} + \mathbf{r}$ we get that $1/2 \|\mathbf{r}\|_1 \leq \varepsilon$ and

$$|\mu_{\mathbf{t}} - \mu_{\mathbf{p}}| \leq n \sum_{x=1}^{m} a_{x} |t_{x} - p_{x}| = n \sum_{x=1}^{m} a_{x} |r_{x}| \leq 2n\varepsilon \sum_{x=1}^{m} a_{x}.$$
(129)

Therefore, for any two types $\mathbf{t}, \mathbf{t}' \in \text{Type}(n, m)$ that are ε close to **p** we have

$$|\mu_{\mathbf{t}} - \mu_{\mathbf{t}'}| \leqslant 4n\varepsilon \sum_{x=1}^{m} a_x.$$
(130)

In other words, the energy spread of the state $|\psi_{\varepsilon}^{n}\rangle$ is no greater than $4n\varepsilon \sum_{x=1}^{m} a_{x}$.

Note that by taking $\varepsilon > 0$ sufficiently small we can make the energy spread $4n\varepsilon \sum_{x=1}^{m} a_x$ much smaller than na_m . However, we still get that the energy spread of ψ_{ε}^n is linear in *n*. We show now that there exist states in PURE(A^n) that are very close to $\psi^{\otimes n}$ but with energy spread that is sublinear in *n*.

Lemma V.1: Let $\psi \in \text{PURE}(A)$ and $\alpha \in (1/2, 1)$. Then, there exists a sequence of pure state $\{\chi_n\}_{n \in \mathbb{N}}$ in $\text{PURE}(A^n)$ with the following properties:

1. The limit

$$\lim_{n \to \infty} \left\| \psi^{\otimes n} - \chi_n \right\|_1 = 0.$$
 (131)

2. The energy spread of χ_n is no more than $4n^{\alpha} \sum_{x=1}^{m} a_x$.

Remark: Note that due to the inequality $\text{Type}(n, m) \leq (n+1)^m$ it follows that any pure state in A^n , including $|\chi_n\rangle$, can be expressed as a linear combination of no more than $(n+1)^m$ energy eigenstates.

Proof. Set $\varepsilon_n := n^{\alpha-1}$. Since $\alpha \in (1/2, 1)$ we have $\lim_{n\to\infty} \varepsilon_n = 0$ and $\lim_{n\to\infty} n\varepsilon_n^2 = \infty$. The latter implies that if we replace ε in (127) with ε_n we still get the zero limit of (127). Hence, the pure state $\chi_n := \psi_{\varepsilon_n}^n$ satisfies Eq. (131). Finally, from Eq. (130) we get that the energy spread of χ_n cannot exceed

$$4n\varepsilon_n \sum_{x=1}^m a_x = 4n^\alpha \sum_{x=1}^m a_x.$$
 (132)

This completes the proof.

D. Sublinear athermality resources

The lemma above asserts that the state $\psi^{\otimes n}$ is very close to a state χ_n , whose energy spread is sublinear in *n*. However, the average energy $\langle \chi_n | H^{\otimes n} | \chi_n \rangle$ grows linearly in *n*. This is consistent with our next assumption that systems whose energy grows sublinearly in *n* can be viewed as asymptotically negligible resources.

Definition V.1: A sublinear athermality resource (SLAR) is a sequence of quantum athermality systems $\{R_n\}_{n\in\mathbb{N}}$, such that $|R_n|$ grows polynomially with n, and there exists two constants independent of n, $0 \le \alpha < 1$ and c > 0, such that

$$\left\|H^{R_n}\right\|_{\infty} \leqslant cn^{\alpha} \quad \forall \ n \in \mathbb{N}.$$
(133)

The key assumption in the definition above is that the energy of systems R_n grows sublinearly with n. Therefore, in the asymptotic limit in which $n \to \infty$ the resourcefulness of any sequence of athermality states $\{(\omega^{R_n}, \gamma^{R_n})\}_{n\in\mathbb{N}}$ becomes negligible relative to the resourcefulness of n copies of the golden unit $(|0\rangle\langle 0|^A, \mathbf{u}^A)$. Specifically, in Appendix B we show that the distillation rate of athermality as given in Theorem V.1 does not change if we replace CTO (or GPC) by CTO+SLAR (or GPC+SLAR). While this small amount of an athermality resource does not change the distillation rate, we see now that it does change the cost rate and thereby sufficient to restore reversibility.

E. Cost of pure states

For any athermality system *R* (i.e., system *R* has a welldefined Hamiltonian H^R and a Gibbs state γ^R) we define the *R*-assisted conversion distance of one athermality state (ρ^A, γ^A) to another athermality state (σ^B, γ^B) as

$$d_{\text{CTO}}^{R} \left(\left(\rho^{A}, \gamma^{A} \right) \to \left(\sigma^{B}, \gamma^{B} \right) \right)$$

$$:= \inf_{\omega \in \mathfrak{D}(R)} d_{\text{CTO}} \left(\left(\rho^{A} \otimes \omega^{R}, \gamma^{AR} \right) \to \left(\sigma^{B}, \gamma^{B} \right) \right). \quad (134)$$

That is, d_{CTO}^R is the smallest distance that (ρ^A, γ^A) can be reached by CTO to (σ^B, γ^B) with the help of a system R, whose Hamiltonian H^R (or equivalently its Gibbs state γ^R) is fixed. With this at hand, we define the *R*-assisted ε cost of (ρ^A, γ^A) to be

$$\operatorname{Cost}_{\operatorname{CTO}}^{\varepsilon,R}\left(\rho^{A},\gamma^{A}\right) := \\ \log \inf_{0 < m \in \mathbb{R}} \left\{ m : d_{\operatorname{CTO}}^{R}\left((|0\rangle\langle 0|^{X}, \mathbf{u}_{m}^{X}) \to (\rho^{A}, \gamma^{A}) \right) \leqslant \varepsilon \right\}.$$

The type of free operations that we consider here are CTO assisted with SLAR. We therefore set in this subsection \mathfrak{F} to be CTO+SLAR. Using the definitions above, we define the asymptotic cost of a state (ρ^A , γ^A) under \mathfrak{F} as

$$\operatorname{Cost}_{\mathfrak{F}}\left(\rho^{A},\gamma^{A}\right) := \\ \inf_{\{R_{n}\}} \lim_{\varepsilon \to 0^{+}} \liminf_{n \to \infty} \frac{1}{n} \operatorname{Cost}_{\operatorname{CTO}}^{\varepsilon,R_{n}}\left(\rho^{\otimes n},\gamma^{\otimes n}\right),$$
(135)

where the infimum is over all SLARs $\{R_n\}_{n \in \mathbb{N}}$.

Theorem V.2: Let (ψ^A, γ^A) be an athermality state with $\psi \in \text{PURE}(A)$. Then,

$$\operatorname{Cost}_{\mathfrak{F}}\left(\psi^{A},\gamma^{A}\right) = D\left(\psi^{A} \| \gamma^{A}\right), \quad (136)$$

where D is the Umegaki relative entropy.

Proof. Let $\mathfrak{S}_{n,\varepsilon}$ be the set of types given in Eq. (125) and set $\mathfrak{S}_n := \mathfrak{S}_{n,\varepsilon_n}$ with $\varepsilon_n := n^{\alpha-1}$. Let also $\{\chi_n\}_{n\in\mathbb{N}}$ be the sequence of pure states that satisfies all the properties outlined in Lemma V.1. In particular, each χ_n is very close to $\psi^{\otimes n}$ (for *n* sufficiently large) and the energy spread of χ_n is given by $4n^{\alpha} \sum_{x=1}^m a_x$ for some $\alpha \in (1/2, 1)$. Recall that each χ_n has the form [cf. Eq. (128)]

$$|\chi_n\rangle = \sum_{\mathbf{t}\in\mathfrak{S}_n} \sqrt{q_{\mathbf{t}}} |\mathbf{t}\rangle^{A^n}, \qquad (137)$$

where $\{q_t\}$ are some coefficients in \mathbb{R}_+ (that form a probability distribution over the set of types in \mathfrak{S}_n). Let $k_n := |\mathfrak{S}_n|$ be the number of terms in the superposition above [hence $k_n \leq (n+1)^m$], and let $\{\mu_i\}_{i=1}^{k_n}$ be the set of all

energy eigenvalues of the Hamiltonian $H^{\otimes n}$ that corresponds to all the energy eigenvectors $\{|\mathbf{t}\rangle^{A^n}\}_{\mathbf{t}\in\mathfrak{S}_n}$. That is, each $j \in [k_n]$ corresponds exactly to one type \mathbf{t} that appears in the superposition (137). Although the energies eigenvalues $\{\mu_j\}$ depend also on n, we did not add a subscript n to ease on the notations. Without loss of generality we also assume that $\mu_1 \leq \cdots \leq \mu_{k_n}$, so that its energy spread $\mu_{k_n} - \mu_1 \leq 4n^{\alpha} \sum_{x=1}^m a_x$ (see Lemma V.1). We also denote by $\mathbf{t}^{\min,n}$ the type in \mathfrak{S}_n that corresponds to the smallest energy μ_1 .

Let R_n be a k_n -dimensional quantum (reference) system whose Hamiltonian has a nondegenerate spectrum given by

$$H^{R_n} = \sum_{j=1}^{k_n} (\mu_j - \mu_1) |j\rangle \langle j|^{R_n}.$$
 (138)

Note that the Hamiltonian H^{R_n} has the same eigenvalues as the energies that appear in χ_n shifted by μ_1 . Set $\lambda_j := \mu_j - \mu_1$ to be the *j* th eigenvalue of H^{R_n} , and observe that

$$0 = \lambda_1 \leqslant \lambda_2 \leqslant \cdots \leqslant \lambda_k \leqslant 2n^{\alpha} \sum_{x=1}^m a_x.$$
(139)

Let $z^n \in [m]^n$ be a sequence of type $\mathbf{t}^{\min,n}$ so that $H^{\otimes n}|z^n\rangle^{A^n} = \mu_1|z^n\rangle^{A^n}$. Let also

$$|\phi^{R_n}\rangle := \sum_{j=1}^k \sqrt{q_j} |j\rangle^{R_n}, \qquad (140)$$

where $q_j := q_t$ with **t** being the type that corresponds to *j*; in other words, **t** is the type satisfying $H^{\otimes n} |\mathbf{t}\rangle^{A^n} = \mu_j |\mathbf{t}\rangle^{A^n}$. By construction, $\{R_n\}_{n \in \mathbb{N}}$ is a SLAR, and the pure state

$$\phi^{R_n} \otimes |z^n\rangle \langle z^n|^{A^n} \tag{141}$$

has the exact same energy distribution as the pure state

$$|1\rangle\langle 1|^{R_n} \otimes \chi_n^{A^n} \tag{142}$$

(recall that $|1\rangle^{R_n}$ corresponds to the zero energy of system R_n). Hence, the above two states are equivalent resources and can be converted from one to the other by reversible thermal operations (i.e., an energy-preserving unitary). We now use this resource equivalency to compute the cost of $\psi^{\otimes n}$ in terms of the cost of the quasiclassical state $|z^n\rangle \langle z^n|$.

Let $\varepsilon \in (0, 1/2)$ and let $n \in \mathbb{N}$ be sufficiently large such that $\psi^{\otimes n}$ is ε close to χ_n . Therefore, any positive real

number $0 < m \in \mathbb{R}$ that satisfies

$$d_{\text{CTO}}^{R_n}\left((|0\rangle\langle 0|^X, \mathbf{u}_m^X) \to (\chi_n^{A^n}, \gamma^{A^n})\right) \leqslant \varepsilon$$
(143)

also satisfies

$$d_{\text{CTO}}^{R_n}\left((|0\rangle\langle 0|^X, \mathbf{u}_m^X) \to (\psi^{\otimes n}, \gamma^{A^n})\right) \leqslant 2\varepsilon.$$
(144)

In particular, this means that

$$\operatorname{Cost}_{\operatorname{CTO}}^{2\varepsilon,R_n}\left(\psi^{\otimes n},\gamma^{A^n}\right) \leqslant \operatorname{Cost}_{\operatorname{CTO}}^{\varepsilon,R_n}\left(\chi_n^{A^n},\gamma^{A^n}\right).$$
(145)

We therefore focus now on bounding the expression on the rhs above.

By adding the resource $(|1\rangle\langle 1|^{R_n}, \gamma^{R_n})$ we can only increase the cost. Therefore,

$$\operatorname{Cost}_{\operatorname{CTO}}^{\varepsilon,R_{n}}\left(\chi_{n}^{\mathcal{A}^{n}},\gamma^{\mathcal{A}^{n}}\right)$$

$$\leqslant \operatorname{Cost}_{\operatorname{CTO}}^{\varepsilon,R_{n}}\left(|1\rangle\langle 1|^{R_{n}}\otimes\chi_{n}^{\mathcal{A}^{n}},\gamma^{R_{n}\mathcal{A}^{n}}\right)$$

$$= \operatorname{Cost}_{\operatorname{CTO}}^{\varepsilon,R_{n}}\left(\phi^{R_{n}}\otimes|z^{n}\rangle\langle z^{n}|^{\mathcal{A}^{n}},\gamma^{R_{n}\mathcal{A}^{n}}\right)$$

$$\leqslant \operatorname{Cost}_{\operatorname{CTO}}^{\varepsilon}\left(|z^{n}\rangle\langle z^{n}|^{\mathcal{A}^{n}},\gamma^{\mathcal{A}^{n}}\right)$$

$$= D_{\max}^{\varepsilon}\left(|z^{n}\rangle\langle z^{n}|^{\mathcal{A}^{n}} \|\gamma^{\mathcal{A}^{n}}\right)$$

$$\leqslant D_{\max}\left(|z^{n}\rangle\langle z^{n}|^{\mathcal{A}^{n}} \|\gamma^{\mathcal{A}^{n}}\right), \quad (146)$$

where in the first equality we use the resource equivalency between the athermality states in Eqs. (141) and (142). In the second inequality we use the fact that the cost of $|z^n\rangle\langle z^n|$ without the assistance of R_n cannot be smaller than the cost of $\phi^{R_n} \otimes |z^n\rangle\langle z^n|$ with the assistance of R_n , since the latter is defined in terms of an infimum over all states $\omega \in \mathfrak{D}(R_n)$ [cf. Eq. (134)]. In the second equality we use the second relation of Eq. (50) combined with the fact that in the quasiclassical regime GPO has the same conversion power as CTO [see Eq. (36)]. Finally, in the last inequality we use the fact that D_{\max} is always no smaller than its smoothed version. Combining this with the previous equation and with the definition of $\text{Cost}_{\mathfrak{F}}(\psi^A, \gamma^A)$, we conclude that

$$\operatorname{Cost}_{\mathfrak{F}}\left(\psi^{A},\gamma^{A}\right) \leqslant \liminf_{n \to \infty} \frac{1}{n} D_{\max}\left(|z^{n}\rangle\langle z^{n}|^{A^{n}} \| \gamma^{A^{n}}\right).$$
(147)

Now, observe that

$$D_{\max}\left(|z^{n}\rangle\langle z^{n}|^{A_{n}}\|\gamma^{A^{n}}\right) = -\log\left\langle z^{n}|\gamma^{A^{n}}|z^{n}\right\rangle$$
$$\gamma^{A^{n}} = \left(\gamma^{A}\right)^{\otimes n} \to = -\sum_{x=1}^{m} nt_{x}^{n,\min}\log\langle x|\gamma^{A}|x\rangle,$$
(148)

where in the last equality we use the fact that the sequence z^n has a type $\mathbf{t}^{\min,n}$. Combining this with the previous equation we conclude that

$$\operatorname{Cost}_{\mathfrak{F}}\left(\psi^{A},\gamma^{A}\right) \leqslant -\lim_{n \to \infty} \sum_{x=1}^{m} t_{x}^{n,\min} \log\langle x | \gamma^{A} | x \rangle$$
$$= -\sum_{x=1}^{m} p_{x} \log\langle x | \gamma^{A} | x \rangle$$
$$= D(\psi \| \gamma), \tag{149}$$

where we use the fact that $\mathbf{t}^{\min,n} \in \mathfrak{S}_n$ so that $\mathbf{t}^{\min,n}$ is ε_n close to **p**. Therefore, since $\lim_{n\to\infty} \varepsilon_n = 0$ we have $\lim_{n\to\infty} \mathbf{t}^{\min,n} = \mathbf{p}$. This completes the proof.

VI. DISCUSSION AND OUTLOOK

Quantum athermality can be viewed as a composite resource consisting of nonuniformity and quantum coherence. While the study of nonuniformity is well understood, the role coherence plays in quantum thermodynamics is far less understood. In this paper, we first developed the resource theory of time-translation asymmetry, which is the type of quantum coherence appearing in thermodynamics. Remarkably, we were able to find (Theorem III.2) a relatively simple criterion, determining if there exists a time-translation covariant channel between two given quantum states. We restricted our attention to Hamiltonians with a nondegenerate Bohr spectrum as almost all Hamiltonians have such a spectrum. However, it is worth noting that some important Hamiltonians, such as the Hamiltonian of the harmonic oscillator, do not have such a spectrum. For such Hamiltonians, some of our results do not apply, although significant progress has been made recently in this direction [46-48].

We use the resource theory of time-translation asymmetry to develop the theory of quantum athermality in the single-shot regime. We consider three types of free operations: TO, CTO, and GPC. In particular, Theorem IV.1 demonstrated in a rigorous way that two athermality states $\rho, \sigma \in \mathfrak{D}(A)$ with the same diagonal elements have the same nonuniformity content, and can only be different in their coherence content.

In this respect, it would be interesting to know if the same result holds also for CTO. One of the long-standing open problems in the resource-theoretic approach to quantum thermodynamics is whether under CTO, quantum athermality comprises of just nonuniformity and coherence. That is, since GPC is a larger set of operations than CTO it could well be that some interconversions between two athermality resources is possible under GPC operations but not under CTO. If this is the case, it would mean that quantum athermality contains another type of resource that is not captured solely by coherence and nonuniformity. In the asymptotic regime, however, GPC does not provide any advantage over CTO. Both sets of operations lead to the same distillable rate given in terms of the Umegaki relative entropy (see Theorem V.1). Since coherence is needed to create athermality states that are not quasiclassical, the cost rate of a nonquasiclassical state diverges. To get a meaningful result, we followed the idea of Ref. [10] to borrow a small amount of coherence, and showed that, for pure states, with the assistance of an asymptotically negligible quantum athermality, we can restore into the fully quantum domain, the reversibility that exists in the quasiclassical regime.

We defined asymptotically negligible resources as sequences $\{R_n\}_{n\in\mathbb{N}}$ whose maximal energy grows sublinearly with *n*. The intuition behind this definition is that the energy of *n* copies of a system *A* grows linearly with *n* so that for sufficiently large *n*, the energy of A^n is much larger than that of R_n . Indeed, such a sublinear athermality resource cannot increase the distillable athermality (see Appendix B).

In Theorem V.2 we showed that the cost rate of a *pure* athermality resource, under $\mathfrak{F} := \text{CTO} + \text{SLAR}$, is given by the Umegaki relative entropy. Moreover, in Theorem V.1 and Appendix B we showed that the distillable rate under \mathfrak{F} of *any* athermality resource is given by the Umegaki relative entropy. When combining these two results together we conclude that the rate of converting (by \mathfrak{F}) many copies of a mixed state (ρ^A , γ^A) to many copies of a pure state (ψ^B , γ^B) is given by

$$\operatorname{Rate}_{\mathfrak{F}}\left(\left(\rho^{A},\gamma^{A}\right)\to\left(\psi^{B},\gamma^{B}\right)\right)=\frac{D(\rho^{A}\|\gamma^{A})}{D(\psi^{B}\|\gamma^{B})}.$$
 (150)

For the specific case that also ρ^A is pure, the above formula indicates that the resource theory of pure athermality is reversible under \mathfrak{F} . For the mixed-state case the problem is still open.

As discussed above, under GPC and CTO, coherence among energy level is a resource that cannot be measured by the golden unit $(|0\rangle\langle 0|^X, \mathbf{u}_m^X)$ introduced in Ref. [44] [see Eq. (41)] for athermality under GPO. The reason is that this golden unit is quasiclassical, and it cannot be converted by GPC (or CTO) to any athermality state that is not quasiclassical (even if we take $m = \infty$). For this reason, one has to specify another golden unit that quantifies the coherence content of quantum athermality. We discuss now a candidate of such a golden unit.

For a given athermality state (ρ, γ) we can interpret the state $(\mathcal{P}(\rho), \gamma)$ as the nonuniformity contained in (ρ, γ) . If fact, we saw in Theorem IV.3 that for any $\varepsilon > 0$, Distill^{ε}_{GPC} $(\rho, \gamma) = \text{Distill}^{\varepsilon}_{\text{GPO}} (\mathcal{P}(\rho), \gamma)$, which supports this assertion. It is somewhat less clear how to characterize or isolate the time-translation asymmetry contained in (ρ, γ) . Consider an athermality state (σ, γ) with the property that $\mathcal{P}(\sigma) = \gamma$. Such an athermality state has zero nonuniformity, and consequently it contains only time-translation asymmetry. We can therefore call such states purely coherent athermality states. In Corollary IV.2 we saw that the purely coherent athermality state (ψ_{γ}, γ) , given by

$$|\psi_{\gamma}\rangle := \sum_{x=1}^{m} \sqrt{g_x} |x\rangle$$
 and $\gamma = \sum_{x=1}^{m} g_x |x\rangle \langle x|$, (151)

can be converted to any other purely athermality state of the form (ρ, γ) , where $\rho \in \mathfrak{D}(A)$ has the same diagonal as γ . Therefore, the athermality state (ψ_{γ}, γ) can be taken to be the golden unit for the coherence content of quantum athermality. Note however that unlike the golden unit $(|0\rangle\langle 0|, \mathbf{u}_m)$ used for the nonuniformity content of athermality, (ψ_{γ}, γ) depends explicitly on the Hamiltonian.

With this golden unit, we can now ask what is the coherence cost of an athermality state (ρ, γ) . To compute the asymptotic cost of preparing many copies, say *n*, of a given athermality state (ρ, γ) we can minimize the integers *m*, *k* for which the conversion

$$(\psi_{\gamma},\gamma)^{\otimes k} \otimes (|0\rangle\langle 0|,\mathbf{u})^{\otimes m} \xrightarrow{\text{GPC}} (\rho,\gamma)^{\otimes n}$$
 (152)

is possible with a small error that vanishes in the limit $n \rightarrow \infty$. We leave the investigation along these lines for future work.

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APPENDIX A: POSSIBLE GAPS IN THE ORIGINAL PROOF OF [10]

The proof given in Ref. [10] seems to have several gaps. Here we point out one such gap, and discuss an implicit assumption made in Ref. [10].

In Eqs. (35) and (37) of their Supplemental Material, the authors of Ref. [10] consider two states

$$\rho^{\otimes n} = \sum_{k,g} p_k |\Psi_{k,g}\rangle \langle \Psi_{k,g}| , \quad \rho_n = \sum p_k |t_k, s_g\rangle \langle t_k, s_g|,$$
(A1)

where for simplicity, the authors consider rank-2 state

$$\rho = p |\phi_1\rangle \langle \phi_1| + (1-p) |\phi_2\rangle \langle \phi_2|. \tag{A2}$$

In the first step of their protocol, one first creates the diagonal state ρ_n which has the same spectrum as $\rho^{\otimes n}$. Since ρ_n is diagonal, its eigenvectors $\{|t_k, s_g\rangle\}$ depend only on the Gibbs state $\gamma^{\otimes n}$. The authors of Ref. [10] do not specify in Eq. (37) (of their Supplemental Material) the range of k, but from Eq. (45) in the Supplemental Material it becomes clear that $k \in \text{Typ}_{\rho} := [np - \sqrt{n}, np + \sqrt{n}]$ [see the sentence above Eq. (38) in the Supplemental Material of Ref. [10] for the definition $\text{Typ}_{\rho} := [np - \sqrt{n}, np + \sqrt{n}]$].

The author then moves to claim that "From the result of the previous section it is not hard to see that this (i.e., the cost of preparing ρ_n) can be done at a rate given by the relative entropy distance of ρ to the Gibbs state, since in the limit of many copies, the regularized relative entropy distance is the same." However, there exists a simple argument why, in general, the cost of preparing ρ_n is not equal to $D(\rho || \gamma)$, where γ is the Gibbs state.

The argument goes as follows. Consider the two states ρ and $\sigma := V\rho V^{\dagger}$, where V is some unitary matrix. Since the eigenvalues of both ρ and σ are p and 1 - p, it follows that the construction of ρ_n would be exactly the same whether our initial state is ρ or whether it is σ . This is because ρ_n does not depend explicitly on the eigenvectors of ρ (only the eigenvalues). However, clearly, there exists a unitary V such that

$$D(V\rho V^* \| \gamma) \neq D(\rho \| \gamma). \tag{A3}$$

Since ρ_n as defined above would be the same for both ρ and $\sigma := V \rho V^*$ the cost rate of preparing ρ_n cannot be equal to $D(\rho || \gamma)$.

Another issue with the proof in Ref. [10] is that the matrix in Eq. (31) of the Supplemental Material of Ref. [10] is not a unitary matrix as claimed. Indeed, by direct calculation

$$U^{\text{inv}}(U^{\text{inv}})^{\dagger} = \sum_{i,i',j} u_{ij} \bar{u}_{i'j} |E_i\rangle \langle E_{i'}|$$

$$\times \otimes |h - E_i + E_j\rangle \langle h - E_{i'} + E_j |$$

$$\neq I. \qquad (A4)$$

Perhaps the intention of the authors of Ref. [10] is to include a sum over h in the definition of U^{inv} , and allowing

1

h to go from $-\infty$ to $+\infty$ so that

$$U^{\text{inv}}(U^{\text{inv}})^{\dagger}$$

$$= \sum_{i,i',j} u_{ij} \bar{u}_{i'j} |E_i\rangle \langle E_{i'}| \otimes \sum_{h=-\infty}^{\infty} |h - E_i + E_j\rangle$$

$$\times \langle h - E_{i'} + E_j|$$

$$= \sum_{i,i',j} u_{ij} \bar{u}_{i'j} |E_i\rangle \langle E_{i'}| \otimes \sum_{h=-\infty}^{\infty} |h - E_i\rangle \langle h - E_{i'}|$$

$$= \sum_{i,i'} \delta_{i,i'} |E_i\rangle \langle E_{i'}| \otimes \sum_{h=-\infty}^{\infty} |h - E_i\rangle \langle h - E_{i'}|$$

$$= I.$$
(A5)

Note that one has to allow for the Hamiltonian of the reference system to have an unbounded negative spectrum. Such Hamiltonians are known to lead to instabilities of the physical system, and occur, for example, in relation to spin-statistics theorem. However, we point out, that in the present paper the author assumes only ancillary systems of finite dimensions and with Hamiltonians whose spectrum is non-negative (i.e., bounded from below).

APPENDIX B: DISTILLATION UNDER GPC+SLAR

Lemma B.1: Let (ρ, γ) be an athermality state, and let $\mathfrak{F} := \text{GPC} + \text{SLAR}$. Then,

$$\text{Distill}_{\mathfrak{F}}(\rho, \gamma) = D(\rho \| \gamma). \tag{B1}$$

Proof. Let $\{(\omega^{R_n}, \gamma^{R_n})\}_{n \in \mathbb{N}}$ an an SLAR and observe that from (112) it follows that for any $n \in \mathbb{N}$ and any $\varepsilon \in (0, 1)$

$$\begin{aligned} \text{Distill}_{\text{GPC}}^{\varepsilon} \left(\rho^{\otimes n} \otimes \omega^{R_n}, \gamma^{A^n} \otimes \gamma^{R_n} \right) \\ &\leq D_{\min}^{\varepsilon} \left(\rho^{\otimes n} \otimes \omega^{R_n} \| \gamma^{A^n} \otimes \gamma^{R_n} \right) \\ (54) \rightarrow &\leq D_{\min}^{\varepsilon} \left(\rho^{\otimes n} \| \gamma^{A^n} \right) + D_{\max} \left(\omega^{R_n} \| \gamma^{R_n} \right), \end{aligned}$$

Now, it is well known (see, e.g., Ref. [43]) that all quantum relative entropies, in particular, D_{max} , satisfy

$$D_{\max}(\omega^{R_n} \| \gamma^{R_n}) \leq \log \left\| (\gamma^{R_n})^{-1} \right\|_{\infty}$$

= $\log \left(\operatorname{Tr} \left[e^{-\beta H^{R_n}} \right] \exp \left(\beta \| H^{R_n} \|_{\infty} \right) \right)$
 $\leq \beta \| H^{R_n} \|_{\infty} + \log \operatorname{Tr} \left[e^{-\beta H^{R_n}} \right]$
 $\leq \beta c n^{\alpha} + \log |R_n|,$

where the last line follows from the fact that $(\omega^{R_n}, \gamma^{R_n})$ is SLAR so there exists c > 0 independent of n, and $\alpha \in$

[0, 1) such that the maximal energy of system R_n does not exceed cn^{α} . Moreover, since $|R_n|$ is polynomial in n we get that for sufficiently large n, $\log |R_n| \leq \beta cn^{\alpha}$. Taking the supremum over all possible SLAR systems R_n we get that for any $\varepsilon > 0$

$$\overline{\lim_{n \to \infty}} \frac{1}{n} \operatorname{Distill}_{\mathfrak{F}}^{\varepsilon} \left(\rho^{\otimes n}, \gamma^{A^{n}} \right)$$

$$= \sup_{\{R_{n}\}_{n \in \mathbb{N}}} \overline{\lim_{n \to \infty}} \frac{1}{n} \operatorname{Distill}_{\operatorname{GPC}}^{\varepsilon} \left(\rho^{\otimes n} \otimes \omega^{R_{n}}, \gamma^{A^{n}R_{n}} \right)$$

$$\leq \sup_{\alpha \in [0,1], c \in \mathbb{R}_{+}} \overline{\lim_{n \to \infty}} \frac{1}{n} \left(D_{\min}^{\varepsilon} \left(\rho^{\otimes n} \| \gamma^{A^{n}} \right) + 2\beta c n^{\alpha} \right)$$

$$= D(\rho^{A} \| \gamma^{A}).$$
(B3)

This completes the proof.

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