Quantum Thermodynamics with Coherence: Covariant Gibbs-Preserving Operation Is Characterized by the Free Energy

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The resource theory with covariant Gibbs-preserving operations, also called enhanced thermal operations, is investigated. We prove that with the help of a correlated catalyst, the state convertibility for any coherent state is fully characterized by the free energy defined with the quantum relative entropy. We can extend this result to general resource theories in the form that imposing the covariant condition to a general resource theory does not change the state convertibility, as long as the initial state is coherent and distillable and this resource theory admits the phase estimation and the phase shift. This means that adding a constraint from the law of energy conservation is irrelevant in the correlated-catalytic framework.

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The central subject in quantum thermodynamics is the controllability of small quantum systems in a thermal environment. The fundamental problems are what constraints can be seen in a small thermal environment and how robust the second law of thermodynamics is [1–3]. One standard implementation of this setup is an operational class of thermal operations, where we employ an energy-conserving unitary operation by consuming an auxiliary system at the Gibbs state [4–6]. However, since thermal operations are formulated in a bottom-up fashion and are not easy to handle directly [7–11], it is convenient to focus on some key properties of thermal operations and examine them separately.

One key characterization of thermal operations is the Gibbs-preserving property that an initial Gibbs state results in the same Gibbs state [12–15]. Unlike conventional macroscopic thermodynamics, various novel constraints other than the conventional second law emerge in small systems [12,14,16–20]. However, interestingly, these novel constraints easily disappear under a small modification, and in various setups, the conventional second law of thermodynamics with a single free energy can be recovered in small quantum regimes [13,15,21–27]. One of the standard setups for this recovery is that with a correlated catalyst [28–30]: In this framework, there is an auxiliary system C whose reduced state does not change through the process while it helps the state conversion such that a state conversion $\rho \to \rho'$ is implemented by $\rho \otimes c \to \tau$ with $\mathrm{Tr}_S[\tau] = c$ and $\mathrm{Tr}_C[\tau] = \rho'$, where c is the state of the catalyst. In the resource theory with Gibbs-preserving operations, the state convertibility with a correlated catalyst is characterized by a single second law in both classical [24] and quantum systems [15].

Thermal operations have, however, another key characterization: the covariant condition, which reflects the restriction from the law of energy conservation. The covariant condition is a genuinely quantum property imposing a constraint on the transformation of coherence among energy eigenstates. We cannot convert, for example, an energy eigenstate into a superposition of two energy eigenstates without any additional help, which serves as a severe restriction on possible operations. The Gibbs-preserving map and thermal operation coincide in the classical regime [5,31], while these two have a gap in the quantum regime due to the restriction on coherence [32].

The restriction from the covariant condition has been studied in the resource theory of asymmetry, or unspeakable coherence [33–37]. Previous studies on the resource theory of asymmetry with a correlated catalyst have revealed two contrastive faces. If the initial state has no coherence, then the final state still has no coherence [38,39]. On the other hand, if the initial state has maybe small but finite coherence, then a covariant operation can convert this state into any state including a maximally coherent state [40,41]. In other words, in covariant operations, whether coherence is zero or nonzero matters while the amount of coherence does not matter.

The Gibbs-preserving condition and the covariant condition are considered to be two main restrictions on thermal operations [2]. This motivates us to introduce a class of operations that are both covariant and Gibbs-preserving, also called the "enhanced thermal operation" [7,8,42,43] (see Fig. 1). Although it is theoretically more tractable than thermal operations, the treatment of covariant Gibbs-preserving operations is still complicated since these two conditions restrict operations in a completely different manner. In fact, the state convertibility with enhanced thermal operations in the single-shot regime obtained in Ref. [8] is extremely complicated. A clear understanding of

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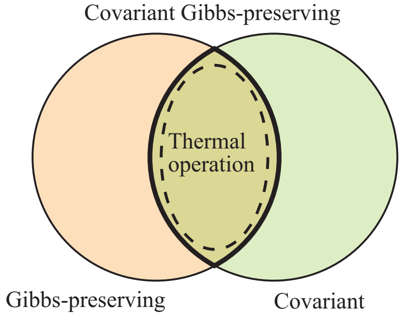

FIG. 1. The relation of covariant Gibbs-preserving operations and thermal operations. The covariant Gibbs-preserving operations (surrounded by a solid line) are the intersection of Gibbs-preserving operations and covariant operations, and thermal operations (surrounded by a dashed line) are its subset. Whether it is a strict subset in the framework with a correlated catalyst is an open problem.

the combination of covariance and Gibbs-preserving properties is still elusive.

In this Letter, we address this problem and establish the necessary and sufficient condition of state conversions for coherent states. We prove that the state convertibility of the covariant Gibbs-preserving operation (enhanced thermal operation) with a correlated catalyst is fully characterized by a single free energy defined by the relative entropy as long as the initial state has nonzero coherence. In other words, in the correlated-catalytic framework, the mere Gibbs-preserving operations and the covariant Gibbs-preserving operations have the same state conversion power, and the only exception is the incoherent case. This serves as strong support for the conjecture on thermal operations raised in Ref. [41]. We emphasize that although the obtained condition is the composition of the conditions for covariant operations and Gibbs-preserving operations, this result including its proof is not trivial at all. In fact, in the single-shot regime, the condition of state conversions for the covariant Gibbs-preserving operations [8] is not the composition of the conditions for covariant operations [35] and Gibbs-preserving operations [15].

The obtained result can be extended to a wide class of other resource theories. We demonstrate that in the correlated-catalytic framework, the addition of the covariant condition does not change the state convertibility as long as the initial state is coherent and distillable and this resource theory admits the phase estimation and the phase shift. The above result applies to, e.g., the resource theory of entanglement. This result justifies our ignorance of the constraint from the law of energy conservation in these resource theories.

Setup and main result—Consider a system whose energy level spacings are integer multiples of some value Δ (i.e., all level spacings are relatively rational). This assumption is employed in various previous studies in the resource theory of (unspeakable) coherence [34,44–46], and thus we follow this convention. Any actual system in experiments has ambiguity in the evaluation of the value of eigenenergy, and thanks to the denseness of rational numbers, within this ambiguity there exists a system with relatively rational level spacings. (For more discussion related to this point, see Supplemental Material [47]). Under this assumption, any state ρ has period $\tau = 2\pi/\mathbf{\Delta}$, i.e., $e^{-iH\tau}\rho e^{iH\tau} = \rho$. Here, we normalize the Planck constant to unity.

A map $\mathcal{E}\colon S \to S$ is called a covariant Gibbs-preserving operation (CGPO) if (i) $\mathcal{E}(\rho_{\mathrm{Gibbs}}) = \rho_{\mathrm{Gibbs}}$ and (ii) $\mathcal{E}(e^{-iHt}\rho e^{iHt}) = e^{-iHt}\mathcal{E}(\rho)e^{iHt}$ for any t are satisfied. Here, $\rho_{\mathrm{Gibbs}} \coloneqq e^{-\beta H}/Z$ is a Gibbs state with a given inverse temperature β. The CGPO is also called "enhanced thermal operation" in Ref. [7].

We investigate the correlated-catalytic conversion with CGPO in this Letter. We say that ρ is convertible to ρ' through CGPO with a correlated catalyst if for any $\varepsilon > 0$ there exist an auxiliary system C called a catalyst, its state c, and a CGPO $\mathcal{E}\colon S \otimes C \to S \otimes C$ such that $\tau = \mathcal{E}(\rho \otimes c)$ with $\mathrm{Tr}_S[\tau] = c$ and $|\mathrm{Tr}_C[\tau] - \rho'|_1 < \varepsilon$. If the last condition is replaced by $\mathrm{Tr}_C[\tau] = \rho'$, we say this conversion is exact.

Now we state our first main result, which serves as an important milestone toward a fully quantum thermodynamics with quantum coherence. Here, we define the free energy $F(\rho) \coloneqq S(\rho||\rho_{\mathrm{Gibbs}})$ with the relative entropy S [65].

Theorem 1—Consider two states ρ and ρ' in S. We assume that the shortest period of ρ is $2\pi/\mathbf{\Delta}$ (i.e., all modes are coherent). Then, ρ is convertible to ρ' through CGPO with a correlated catalyst if and only if $F(\rho) \geq F(\rho')$. In addition, if $F(\rho) > F(\rho')$ and ρ' is full-rank, then this conversion is exact.

This theorem clearly shows the recovery of the second law of thermodynamics in CGPO as long as the initial state has coherence. This is the first result establishing the universality of the second law of thermodynamics under the covariant operation without any consumption of coherence in external systems. As clearly demonstrated by this theorem, the presence of quantum coherence does not disturb thermodynamic state conversions, if small but finite coherence exists at the initial stage. This shows a clear contrast to previous suggestions that the quantum coherence serves as a severe constraint in quantum thermodynamics [7–10].

To prove this theorem, a special type of asymptotic conversion, called the "marginal-asymptotic conversion" [66,67], plays a pivotal role. We say that ρ is convertible to ρ' in a marginal asymptotic conversion through CGPO with conversion rate r with arbitrarily small error if for any $\varepsilon > 0$ there exists a sufficiently large integer N and a CGPO $\Lambda\colon S^{\otimes N} \to S'^{\otimes \lfloor rN \rfloor}$ such that $\Xi = \Lambda(\rho^{\otimes N})$ with $|\mathrm{Tr}_{\setminus i}[\Xi] - \rho'|_1 < \varepsilon$ for all $i = 1, 2 \ldots, \lfloor rN \rfloor$. The difference between the conventional asymptotic conversion and the marginal-asymptotic conversion lies in the fact that we measure the error by the final state of the entire copies in conventional asymptotic conversions, while by reduced states of each copy in marginal-asymptotic conversions. This apparently small difference in definitions is in fact so important that the following lemma, which is a key step of the proof of Theorem 1, is valid only for marginal-asymptotic conversions.

Lemma 1—Assume the same assumptions as Theorem 1 and $F(\rho) \geq F(\rho')$. Then, for any $\delta > 0$ there exists a marginal-asymptotic conversion from ρ to ρ' through CGPO with conversion rate $1 - \delta$ with arbitrarily small error.

It is known that an approximate asymptotic conversion can be transformed into a correlated-catalytic conversion [15,41,68] (see also [69–76] for its applications). Using this connection, Lemma 1 directly implies the existence of a correlated-catalytic conversion from ρ to ρ' with arbitrarily small error (Theorem 1).

Extension to general resource theories—As will be seen soon later, the proof of Theorem 1 does not utilize detailed properties of the Gibbs-preserving property. What we use is the fact that (a) distillation is possible by a Gibbs-preserving operation (GPO), and (b) the phase estimation and the phase shift can be performed by a GPO. Using these facts, the problem is reduced to whether a marginal-asymptotic conversion from ρ to ρ' with rate 1 exists in GPO.

Thus, if the above two conditions are satisfied by a resource theory $\mathbb{O}$, our proof technique also works for this resource theory, and state convertibility in this resource theory with adding the covariant condition $\mathbb{O} \cap \mathrm{Cov}$ is fully determined whether a marginal-asymptotic conversion from ρ to ρ' with rate 1 exists in $\mathbb{O}$. In this situation, whether the desired marginal-asymptotic conversion exists is the central problem.

Fortunately, a recent result by Ganardi *et al.* [67] reveals the equivalence of a marginal-asymptotic conversion with rate 1 and a correlated-catalytic conversion for distillable states. Applying this result, we conclude that if the initial state is distillable and coherent, and this resource theory admits the phase estimation and the phase shift, then the state convertibility does not change by further imposing the covariant condition. This is the second main result of this Letter.

Theorem 2—Consider a resource theory whose free operations are $\mathbb{O}$. Suppose that ρ is convertible to ρ' with a correlated catalyst, ρ is distillable, and ρ has period $2\pi/\Delta$. In addition, the phase estimation and the phase shift can be performed in $\mathbb{O}$. Then, in a resource theory with free operations, $\mathbb{O} \cap \mathrm{Cov}$, ρ is convertible to ρ' with a correlated catalyst.

Surprisingly, the covariant condition, which forces us to pay attention to quantum coherence, puts no restriction on state convertibility in a wide class of resource theories. In the investigation of resource theories, we usually ignore the constraint from energy conservation, though the law of energy conservation is universal, and thus any resource theory should take this restriction into account. This striking theorem justifies this ignorance by showing that a resource theory with and without the covariant condition has essentially the same state conversion power in the framework with a correlated catalyst. Let us take as an example the resource theory of entanglement, where the set of free operations $\mathbb{O}$ is LOCC (local operations and classical communications) [65]. In this case, the true set of accessible operations in our world is LOCC $\cap$ Cov (local operations and classical communications under energy conservation). However, by recalling that a state tomography is possible by LOCC, and thus the phase estimation is tractable in the framework with LOCC, it suffices to treat simply the class of LOCC, since LOCC and LOCC $\cap$ Cov provide the same state convertibility for all distillable states with finite coherence.

Conversion protocol for CGPO—Below, we construct a CGPO protocol realizing the desired state conversion claimed in Lemma 1. To construct our protocol, we employ the following two established facts. One is on a GPO, stating that state convertibility by a GPO in the conventional asymptotic conversion is fully characterized by the free energy.

Proposition 1 (Gibbs-preserving map, used in [15,25])—Consider two states ρ and ρ' in S with $F(\rho) \geq F(\rho')$. Then, for any $\delta_1 > 0$ there exists a large M and a GPO $\Lambda\colon S^{\otimes M} \to S^{\otimes M}$ such that $|\Lambda(\rho^{\otimes M}) - \rho'^{\otimes M}|_1 < \delta_1$.

The other is on a covariant operation, stating that we can efficiently estimate the phase of a state by a covariant operation. Here, the phase (time) estimation is a task to evaluate τ from many copies of $e^{-iH\tau}\rho e^{iH\tau}$.

Proposition 2 (Covariant time estimation, used in [44])—There exists a time estimation protocol $S^{\otimes m} \to \mathbb{R}$ with probability distribution $P(t_{\mathrm{est}}|\kappa)$ for κ on $S^{\otimes m}$ such that (i) covariant $P(t_{\mathrm{est}}|e^{-iH^{\otimes m}\tau}\kappa e^{iH^{\otimes m}\tau}) = P(t_{\mathrm{est}} + \tau|\kappa)$ for any κ and τ, and (ii) the variance of $P(t_{\mathrm{est}}|\rho^{\otimes m})$ decays as $O(1/m)$ for any ρ in S whose shortest period is $2\pi/\Delta$.

Without loss of generality, we suppose that the average of t_{est} for ρ is zero (i.e., we set the origin of the phase as supposition).

Now we construct a protocol converting N copies of ρ into $(1-\delta)N$ copies of ρ' for sufficiently large N. We decompose N copies of ρ into $\nu \coloneqq (1-\delta)\sqrt{N}$ sets of $\sqrt{N}$ copies and two $\delta N/2$ copies, which we call A part, B_1 part, and B_2 part respectively. The sets in A part are labeled as $A_1, \ldots, A_\nu$. In addition, for state R on $S^{\otimes n}$, we introduce a time propagator by t defined as

$$\mathcal{T}_t(R) \coloneqq e^{-iH^{\otimes n}t} R e^{iH^{\otimes n}t}. \tag{1}$$

In the following, we choose n properly depending on state R.

Now we construct the marginal-asymptotic conversion protocol which consists of two steps: (i) We apply the time estimation protocol of Proposition 2 on B_1 part and B_2 part

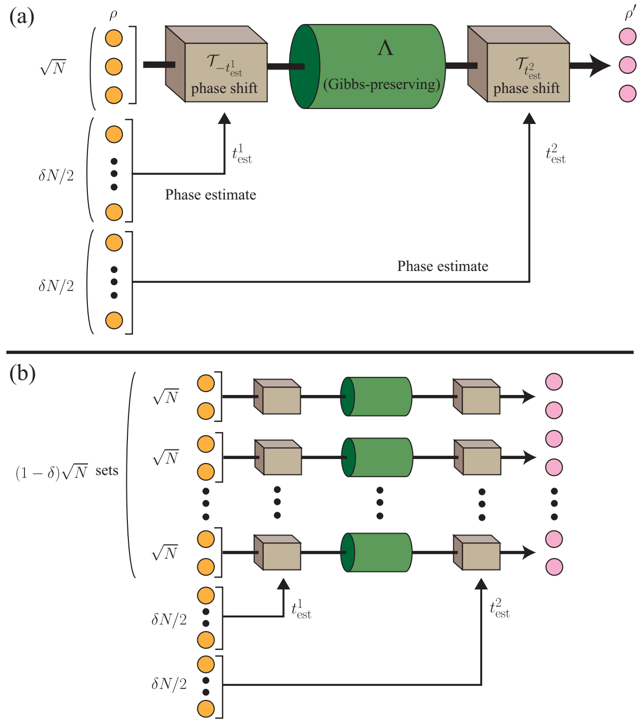

FIG. 2. (a) A protocol for a single set A_i converting $\rho^{\otimes\sqrt{N}}$ to $\rho'^{\otimes\sqrt{N}}$. We estimate the times t^1_{est} and t^2_{est} by using $\delta N/2$ copies of ρ, respectively, and apply the phase shift before and after the Gibbs-preserving operation. (b) The whole protocol of the marginal-asymptotic conversion from ρ to ρ' with transformation rate $1-\delta$. Here, all the phase shift protocols employ the same estimators t^1_{est} and t^2_{est}.

(two $\delta N/2$ copies of ρ) and obtain t^1_{est} and t^2_{est}. The variance of t_{est} decays as $O(1/\delta N)$. (ii) We apply the map

$$\mathcal{M}(\rho^{\otimes\nu\sqrt{N}}) \coloneqq \mathcal{T}_{t^2_{est}} \circ \Lambda^{\otimes\nu} \circ \mathcal{T}_{-t^1_{est}}(\rho^{\otimes\nu\sqrt{N}}) \tag{2}$$

on each set A_i (see Fig. 2). Here, Λ is the Gibbs-preserving map converting $\rho^{\sqrt{N}}$ to $\rho'^{\sqrt{N}}$, whose existence is shown in Proposition 1.

It is not hard to demonstrate that this protocol is indeed covariant, Gibbs-preserving, and realizes the desired conversion (see Supplemental Material [47]). The covariant condition is demonstrated as follows. Consider an input state in the form of $e^{-iHt}\rho e^{iHt}$. The first phase estimator t^1_{est} cancels the phase shift in the initial state, which guarantees that the input state for Λ is always the same regardless of the initial phase t. Then, the second phase estimator t^2_{est} recovers the phase shift, which fulfills the covariant condition. Note that we need to perform time estimation twice since the reuse of estimator t_{est} accompanies unwanted correlation between two phase-shift operations and the covariant condition becomes unsatisfied.

Some complicated construction, e.g., decomposition into sets with $\sqrt{N}$ copies, is necessary to realize the desired transition with small errors. Proposition 2 implies that $\mathcal{T}_{t_{est}}$ on a copies of ρ with t_{est} estimated from b copies of ρ has phase variance of order (a/b). Since we need to vanish the phase variance, the number of copies in each set ($\sqrt{N}$ in our case) should be sublinear in the number of copies used for phase estimation ($\delta N/2$ in our case). In addition, to realize the conversion rate to be close to 1, the number of copies should satisfy the following order relation: (copies in a single set) $\ll$ (copies for phase estimation) $\ll$ (total converted copies), which is chosen so that negligibly few copies are consumed for phase estimations compared to the converted copies to ρ'.

Comments and perspectives—We established the necessary and sufficient condition of state conversions with covariant Gibbs-preserving operations (enhanced thermal operations) in the correlated-catalytic framework. Our result confirms the recovery of the second law of thermodynamics in a fully quantum regime under the law of energy conservation, and the only exception is the incoherent case, whose measure in the state space is zero. The constraint from the energy conservation only separates the coherent case and the incoherent case, and no further restriction is imposed on coherent states. This structure is not special to quantum thermodynamics but widely seen in general resource theories. Namely, we demonstrated that the addition of the covariant condition does not change the state convertibility, as long as the state is coherent and distillable and this resource theory admits the phase estimation and the phase shift. This fact supports why various resource theories work well without taking into account the universal and evidently present constraint from the law of energy conservation.

We note that the present result applies only to systems whose energy level spacings are relatively rational, which is necessary for our present phase estimation processes. This limitation has the same route as the sublinear asymptotic conversion protocol under the covariant condition proposed by Marvian [44], which can be diverted into a marginal-asymptotic conversion protocol [30]. To overcome this limitation, it appears fruitful to employ a recent result on the triviality of the resource theory of (unspeakable) coherence with a correlated catalyst [40,41], which applies to general systems with relatively irrational energy level spacings. We strongly expect that the same result holds for general systems, though combining the above result and the results presented in this Letter seems not straightforward.

An interesting question is the connection to thermal operations, which is the most common and physically supported class of free operations in quantum thermodynamics. The Gibbs-preserving condition and the covariant condition are the two central conditions of thermal operations, and whether these two are the only relevant conditions is an open problem [2]. If the answer to the above question is positive, then the covariant Gibbs-preserving operations fully capture the structure of thermal operations, and the conjecture raised in Ref. [41] that the quantum thermodynamics with thermal operations is characterized by a single free energy for coherent states is resolved in the affirmative. At present, our result provides a consistent view to this conjecture. We note that the covariant Gibbs-preserving operations and thermal operations are known to have a gap in the single-shot regime [7,42]. However, since many inequivalent resource theories collapse in the correlated-catalytic framework [77], it is still plausible to expect that the gap between the covariant Gibbs-preserving operations and thermal operations is unstable and these two have the same conversion law in the correlated-catalytic framework. Whether these two are indeed equivalent or not is left as an open problem to be solved.

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