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Quantum Thermodynamics of Correlated-Catalytic State Conversion at Small Scale

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The class of possible thermodynamic conversions can be extended by introducing an auxiliary system called *catalyst*, which assists in state conversion while its own state remains unchanged. We reveal a complete characterization of catalytic state conversion in quantum and single-shot thermodynamics by allowing an infinitesimal correlation between the system and the catalyst. Specifically, we prove that a single thermodynamic potential, which provides the necessary and sufficient condition for the correlated-catalytic state conversion, is given by the standard nonequilibrium free energy defined with the Kullback-Leibler divergence. This resolves the conjecture raised by Wilming, Gallego, and Eisert [Entropy **19**, 241 (2017)] and by Lostaglio and Müller [Phys. Rev. Lett. **123**, 020403 (2019)] in the positive. Moreover, we show that, with the aid of the work storage, any quantum state can be converted into another by paying the work cost equal to the nonequilibrium free energy difference. Our result would serve as a step towards establishing resource theories of catalytic state conversion in the fully quantum regime.

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Introduction.—The extension of thermodynamics to small-scale quantum systems has attracted attention in various research fields. A variety of the second laws employing the Rényi entropies and divergences [1–4] or majorization [5–8] naturally arise in the small scale, which is contrastive to conventional thermodynamics where only a single thermodynamic potential such as the equilibrium free energy characterizes state convertibility [9]. Recent studies pushing toward this direction have been developed in terms of resource theories [6,7,10]. The resource theory of athermality [4,11–13] paves the way for establishing the information-theoretic foundation of thermodynamics.

In resource theories, an auxiliary system called *catalyst* plays a key role [14], assisting the state conversion while the catalyst itself does not change. To formulate the catalytic state conversion, we suppose the composite system of the system and the catalyst, and consider a state conversion $\rho \otimes c \to \sigma \otimes c$, where ρ, σ are states of the system and c is a state of the catalyst. On one hand, if we require the exact return of the catalyst, an infinite family of Rényi entropies or divergences characterizes possible catalytic state conversion [1–4]. On the other hand, if we allow a small finite error in the final state of the catalyst, any state conversion is possible, which is called *embezzling* [4,15,16]. Here our focus lies in their intermediate regime, where another nontrivial characterization of state convertibility emerges.

Specifically, we consider the situation that the catalyst returns to its initial state exactly but with a negligibly small correlation between the system and the catalyst. As observed in Refs. [17,18], stochastic independence (absence of correlations) is a resource of thermodynamic state conversions. Along with this idea, Wilming, Gallego, and Eisert [19] conjectured that the nonequilibrium free energy defined by the quantum Kullback-Leibler (KL) divergence gives the unique criterion of correlated-catalytic state conversion via a Gibbs-preserving map with a negligibly small correlation. In the classical case, this conjecture has been solved in the positive by Müller [20] and generalized by Rethinasamy and Wilde [21]. However, these results cannot apply to the quantum case, because unlike the classical case known criteria for quantum relative majorization are highly complicated [22–24]. Therefore, the original conjecture raised in Ref. [19] (also raised in Ref. [25] in a rigorous manner) for the quantum cases has still been left as a highly nontrivial open problem.

In this Letter, we solve this problem for the quantum case [19,25] in the affirmative: We prove that the KL divergence indeed characterizes quantum correlated-catalytic state conversion in a necessary and sufficient manner. That is, the correlated-catalytic state conversion between two given quantum states by a Gibbs-preserving map is possible *if and only if* the nonequilibrium free energy defined by the KL divergence does not increase. We further prove that even if the final free energy is larger than the initial one, we can still convert the initial state to the final one by adding a two-level work storage and paying the work cost equal to or greater than the free energy difference. Our result implies that the conventional form of the second law given by the KL divergence is restored even in the quantum regime, if the catalyst is allowed to correlate with the system.

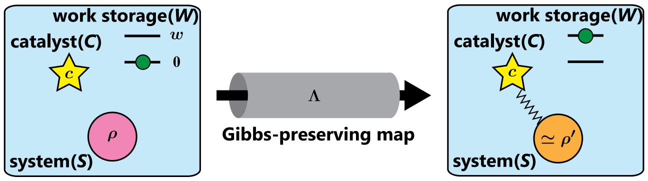

FIG. 1. Schematic of our setup. We convert the system S from ρ to ρ' with the aid of the catalyst C and the work storage W. The catalyst C returns to its original state while it can correlate with the system. The work storage W changes its state with energy difference $w \le F(\rho) - F(\rho')$ with probability arbitrarily close to unity.

Setup and the main claim.—Consider a finite-dimensional quantum system with Hamiltonian H. We investigate state conversion through a particular class of the completely positive and trace-preserving (CPTP) maps, called Gibbs-preserving maps Λ, which keep the Gibbs state invariant: $\Lambda(\rho_{\mathrm{Gibbs}}) = \rho_{\mathrm{Gibbs}}$. Here, $\rho_{\mathrm{Gibbs}} \coloneqq e^{-\beta H}/Z$ is the Gibbs state with the inverse temperature β of the environment. We set the Boltzmann constant to unity. In terms of the resource theory of athermality, the Gibbs state is a free state (with zero athermality), and Gibbs-preserving maps do not generate any nonfree state (with nonzero athermality) from a free state.

We employ an external system called the catalyst denoted by C, which assists state conversion of the system S while the state of C itself does not change (see also Fig. 1). As in Refs. [4,13,17,18,20,21,26–28], we allow a negligibly small error on the final state of S, while the marginal state of C exactly goes back to the initial state. The most crucial assumption is to allow a negligibly small correlation between S and C in the final state. This assumption is motivated by the fact that negligibly small correlations are always allowed between the system and the environment in conventional thermodynamics. In terms of resource theories, a catalyst for a system can be reused as a catalyst for other systems even when a correlation with the first system remains.

We define the nonquilibrium free energy as $F(\rho) \coloneqq S_1(\rho||\rho_{\mathrm{Gibbs}})$, where $S_1(\rho||\rho_{\mathrm{Gibbs}}) \coloneqq \mathrm{Tr}[\rho \ln \rho] - \mathrm{Tr}[\rho \ln \rho_{\mathrm{Gibbs}}]$ is the KL divergence [29]. We now state our first main theorem:

Theorem 1.—Consider two quantum states of S; ρ and ρ'. Then, $F(\rho) \ge F(\rho')$ is satisfied if and only if there exist a catalyst C and its state c, and a Gibbs-preserving map Λ satisfying $\Lambda(\rho \otimes c) = \tau$ such that (i) $\mathrm{Tr}_S[\tau] = c$, (ii) $\mathrm{Tr}_C[\tau]$ is arbitrarily close to ρ', (iii) the correlation between S and C in the final state is arbitrarily small.

The fully rigorous statement of the above theorem and its proof are presented in the Supplemental Material [30]. Here, we only remark that the closeness between states is quantified by the trace distance $d_1(\rho', \rho'') \coloneqq \frac{1}{2}\mathrm{Tr}[|\rho' - \rho''|]$ and the amount of the correlation is quantified by the mutual information $I_{\mathrm{SC}}[\tau] \coloneqq S_1(\tau||\rho'' \otimes c)$, where $\rho'' = \mathrm{Tr}_C[\tau]$ is the reduced state of τ on S. An arbitrarily small error can be achieved by choosing an appropriate catalyst, which might be very large. This theorem manifests that the free energy $F(\rho)$ serves as the single monotone of quantum thermodynamics at the small scale if we allow a negligibly small correlation between the system and the catalyst.

In the case of $F(\rho) < F(\rho')$, Theorem 1 implies that we cannot convert ρ to ρ' through any Gibbs-preserving map. However, even in this case, we can convert ρ to ρ' with the aid of the work storage W (see Fig. 1). The work storage is a two-level system which compensates for the energy change in S by investing the work cost. The initial state of W is an energy eigenstate $|a\rangle$ with energy E_a, and the final state is arbitrarily close to another energy eigenstate $|b\rangle$ with energy E_b. Thus, the work value is almost deterministic, which is an approximate version of the single-shot scenario [8,20,31].

By applying Theorem 1 to the composite system SW, we find that $\rho \otimes |a\rangle\langle a|$ can be converted to a state close to $\rho' \otimes |b\rangle\langle b|$ with a catalyst if we allow a correlation between SW and C. Further to that, we can prove a much stronger statement: the desired state conversion is possible even when there is no correlation between W and the remaining part SC:

Theorem 2.—Consider two quantum states ρ and ρ' of the system S with $F(\rho) - F(\rho') < 0$. Then, $F(\rho) - F(\rho') \ge w$ is satisfied if and only if there exist a catalyst C and its state c, a work storage W with $E_b - E_a \ge w$, and a Gibbs-preserving map Λ satisfying $\Lambda(\rho \otimes c \otimes |a\rangle\langle a|) = \tau \otimes \omega$ with τ and ω being states of SC and W, such that (i) $\mathrm{Tr}_S[\tau] = c$, (ii) $\mathrm{Tr}_C[\tau]$ is arbitrarily close to ρ', (iii) ω is arbitrarily close to $|b\rangle\langle b|$, (iv) the correlation between S and C in τ is arbitrarily small.

This theorem reveals the minimum work cost when C correlates only with S as depicted in Fig. 1, and represents the principle of maximum work [32–34]. The foregoing two theorems together provide the second law of quantum thermodynamics in the small scale, yet in the apparently same form as conventional macroscopic thermodynamics.

We note that Theorem 2 only applies the case of the work investment ($w < 0$), and does not cover the case of the work extraction ($w > 0$). We will, however, discuss a sufficient condition for the case of work extraction in the Supplemental Material (Lemma 3) [30].

Outline of the proof.—The *if* part is easy to prove by applying the superadditivity of the KL divergence. Therefore, we here summarize the outline of the proof of the *only if* part. The detailed idea is demonstrated along with a simple example soon later, and the full proofs are presented in the Supplemental Material [30]. We mainly treat Theorem 1 and briefly comment on Theorem 2.

Our proof consists of three steps. In Step 1, we provide a sufficient condition to convert a quantum state σ to another state σ' via a Gibbs-preserving map by explicitly constructing the desired map. We first perform a binary quantum measurement to determine whether the state is σ or the Gibbs state σ_{Gibbs}, and then prepare two quantum states depending on the measurement outcome. In the case of Theorem 2 (with work storage), we consider not binary but ternary measurements, which require more careful treatment. The derived sufficient condition employs two kinds of divergences; the quantum hypothesis testing divergence [35] and the quantum Rényi divergence. This sufficient condition has been obtained by some of the literature explicitly [36] and implicitly [8,27,28,37,38].

In Step 2, we apply the quantum Stein's lemma, which claims the convergence of the quantum hypothesis testing divergence rate to the KL divergence rate in the limit of infinitely many copies of given quantum systems [39,40]. The reason why quantum hypothesis testing appears in quantum thermodynamics is that the ε-smoothed Rényi-∞ divergence (introduced later) is bounded from both above and below by two quantum hypothesis testing divergences, and hence it also converges to the KL divergence [41,42]. In generic systems, the size of the catalyst diverges with the vanishing error and correlation (ε, $\delta \to 0$). The necessary size of a catalyst can be evaluated by examining the aforementioned convergence speed [39–42].

In Step 3, we reduce the result on the asymptotic state conversion (with multiple copies of states) of Step 1 and 2 to the catalytic state conversion. Although this type of reduction has been discussed in some of the literature [1,4,43], we need some modification on the existing technique to keep the catalyst at the same state, because our asymptotic state conversion accompanies errors. Combining these three steps, we arrive at the desired result.

Toy example of Theorem 1.—We demonstrate the proof of the *only if* part of Theorem 1 in a toy example, a two-level system spanned by $\{|0\rangle, |1\rangle\}$. We will construct two states that are not convertible from one to the other without catalyst, but are convertible with a correlated catalyst. For this purpose, we set $\rho = (3/200)|0\rangle\langle 0| + (197/200)|1\rangle\langle 1|$, $\rho' = |+\rangle\langle +|$ with $|+\rangle \coloneqq 1/\sqrt{2}(|0\rangle + |1\rangle)$, $\beta = 1$, $E_0 = 0$ and $E_1 = \ln 3$. The Gibbs state is given by $\rho_{\mathrm{Gibbs}} = (3/4)|0\rangle\langle 0| + (1/4)|1\rangle\langle 1|$. We set the upper bound of the error and the correlation strength as $\varepsilon = 0.01$ and $\delta = 0.06$, respectively. This state conversion is fully quantum because ρ' is not diagonal in the energy eigenbasis. We remark that any Gibbs-preserving map without catalyst cannot convert ρ to ρ'. To see this, we introduce the Rényi-∞ divergence $S_\infty(\sigma||\kappa) \coloneqq \ln(\min[\lambda : \sigma \leq \lambda\kappa])$ and its ε-smoothing $S_\infty^\varepsilon(\sigma||\kappa) \coloneqq \min_{d(\sigma',\sigma)\leq\varepsilon} S_\infty(\sigma||\kappa)$ with $\varepsilon > 0$. The Rényi divergence satisfies the monotonicity under CPTP maps, and hence $S_\infty^\varepsilon(\rho'||\rho_{\mathrm{Gibbs}}) \leq S_\infty(\rho||\rho_{\mathrm{Gibbs}})$ is a necessary (but not sufficient) condition for state conversion without catalyst. However, we can show $S_\infty^\varepsilon(\rho'||\rho_{\mathrm{Gibbs}}) > S_\infty(\rho||\rho_{\mathrm{Gibbs}})$ in the above parameter setting [see Fig. 2(a)]. We note that the above catalytic state conversion requires only 11 qubits, which is accessible by recent or near-term experimental techniques of, e.g., superconducting qubits [44,45].

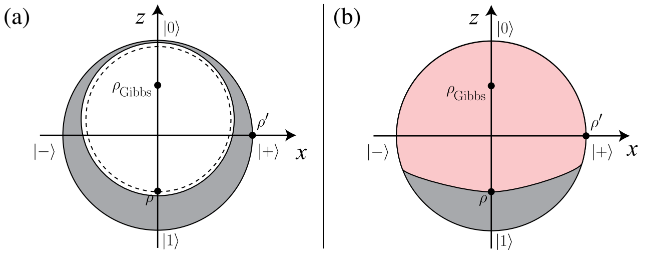

FIG. 2. (a) Schematic of the criterion $S_\infty(\rho'||\rho_{\mathrm{Gibbs}}) < S_\infty(\rho||\rho_{\mathrm{Gibbs}})$ represented by the dashed line in the x-z plane of the Bloch sphere. We draw the states inconvertible from ρ within error ε in gray. (b) Schematic of the criterion $S_1(\rho'||\rho_{\mathrm{Gibbs}}) < S_1(\rho||\rho_{\mathrm{Gibbs}})$ in the x-z plane of the Bloch sphere, where we draw the convertible and inconvertible states with a correlated catalyst from ρ in red and gray, respectively. In particular, there exists a Gibbs-preserving map with the correlated catalyst converting $\rho \to \rho'$.

We treat Step 1 and Step 2 in parallel. Consider a composite system of eight copies of the two-level system: $\{|0\rangle, |1\rangle\}^{\otimes 8}$. We construct a CPTP map which converts $\rho^{\otimes 8}$ to a state Ξ satisfying $d_1(\Xi, \rho'^{\otimes 8}) < \varepsilon$ while keeping ${\rho_{\mathrm{Gibbs}}}^{\otimes 8}$ unchanged. We introduce the projection operator Q onto the subspace of $\{|0\rangle, |1\rangle\}^{\otimes 8}$ spanned by a subset of the computational basis that contains at most one $|0\rangle$. We perform the binary measurement with $\{Q, 1-Q\}$ in order to distinguish $\rho^{\otimes 8}$ and $\rho_{\mathrm{Gibbs}}^{\otimes 8}$. By this measurement, $\rho^{\otimes 8}$ outputs Q with probability $0.994\cdots > 1-\varepsilon$, and $\rho_{\mathrm{Gibbs}}^{\otimes 8}$ outputs $1-Q$ with probability $1-(25/4^8)$. Their differences from 1 (i.e., $0.005\cdots$ and $25/4^8$) correspond to the error of the first and the second kinds [30], respectively. We then prepare quantum states depending on the measurement outcome. If the outcome is Q, we prepare the state $|+\rangle\langle +|$, and if the outcome is $1-Q$, we prepare the state ζ expressed as

$$\zeta = \frac{1}{1-\frac{25}{4^8}}\left(\rho_{\mathrm{Gibbs}}^{\otimes 8} - \frac{25}{4^8}|+\rangle\langle +|^{\otimes 8}\right), \tag{1}$$

which is positive semidefinite because $\rho_{\mathrm{Gibbs}}^{\otimes 8} - \lambda|+\rangle\langle +|^{\otimes 8} \geq 0$ for $\lambda \leq (3/8)^8$ [30] and $25/4^8 < (3/8)^8$. This measurement-and-preparation procedure indeed converts ρ_{Gibbs} to ρ_{Gibbs} by construction and converts ρ to $\Xi \coloneqq (0.994\cdots)|+\rangle\langle +|^{\otimes 8} + (0.005\cdots)\zeta$, which satisfies $d_1(\Xi, |+\rangle\langle +|^{\otimes 8}) < \varepsilon$. We denote this CPTP map by Λ.

We next move to Step 3. We identify the system S to S_1 and the catalyst C to $S_2 \otimes \cdots \otimes S_8 \otimes A$, where A is an auxiliary system spanned by a basis $\{|1\rangle, |2\rangle, \ldots, |8\rangle\}$. The Hamiltonian of A is set to be trivial (i.e., all the states in A take the same energy). Using Ξ on $S_1 \otimes \cdots \otimes S_8$ introduced above, we define Ξ_i ($i = 1, \ldots, 8$) as the reduced state of Ξ on $S_1 \otimes \cdots \otimes S_i$. We set $\Xi_0 \coloneqq 1 \in \mathbb{R}$ (i.e., the

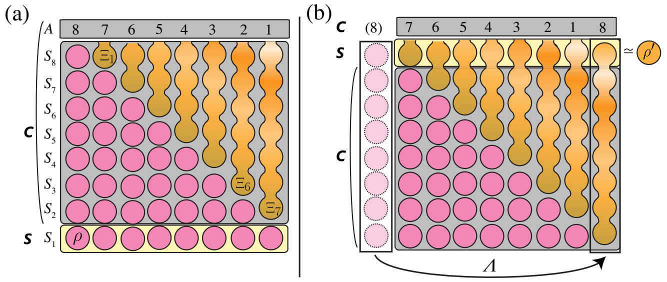

FIG. 3. Schematic of Step 3 of the proof. (a) The initial state of the composite SC. The vertical direction represents different systems $S_1, \ldots, S_8$, and the horizontal direction means their classical mixture. (b) Schematic of how the CPTP map Λ gives the desired catalytic state conversion.

trivial state) for convenience. Using these states, we set the state of C as

$$c \coloneqq \frac{1}{8}\sum_{k=1}^{8} \rho^{\otimes k-1} \otimes \Xi_{8-k} \otimes |k\rangle\langle k|, \tag{2}$$

where $\rho^{\otimes k-1}$ is the state of $S_2 \otimes \cdots \otimes S_k$, and Ξ_{8-k} is now the state of $S_{k+1} \otimes \cdots \otimes S_8$. The initial state of the composite system is $\rho \otimes c = (1/8)\sum_{k=1}^{8} \rho^{\otimes k} \otimes \Xi_{8-k} \otimes |k\rangle\langle k|$ [see Fig. 3(a)]. We now construct the desired CPTP map as follows: If the auxiliary system A is $|8\rangle\langle 8|$, then we apply Λ to $S_1 \otimes \cdots \otimes S_8$, and leave it unchanged otherwise. Then, we shift the auxiliary system A as $|8\rangle \to |1\rangle$ and $|n\rangle \to |n+1\rangle$. Through this process, $1/8\sum_{k=1}^{8} \rho^{\otimes k} \otimes \Xi_{8-k} \otimes |k\rangle\langle k|$ is converted into $\tau' = (1/8)\sum_{k=1}^{8} \rho^{\otimes k-1} \otimes \Xi_{9-k} \otimes |k\rangle\langle k|$ [see Fig. 3(b)]. Remarkably, the partial trace of τ' with respect to S_8 recovers the initial state of the catalyst c. In addition, by defining ξ_l as the reduced state of Ξ on S_l, the reduced state of τ' on S_8 is expressed as $1/8\sum_{l=1}^{8} \xi_l$, which is ε close to the desired state $\rho' = |+\rangle\langle +|$ because $d_1(\Xi, |+\rangle\langle +|^{\otimes 8}) < \varepsilon$. Moreover, since Λ is a Gibbs-preserving map, the constructed CPTP map is also Gibbs preserving. Thus, swapping the two-level systems as $S_n \to S_{n+1}$ and $S_8 \to S_1$ after the above CPTP map, we arrive at the desired Gibbs-preserving map: $\rho \otimes c$ is converted into τ with $\mathrm{Tr}_S[\tau] = c$, $d_1(\mathrm{Tr}_C[\tau], \rho') < \varepsilon$, and $I_{SC}(\tau) < \delta$. Here, since S is a two-level system, $d_1(\tau, \rho' \otimes c) < \varepsilon$ implies $I_{SC}(\tau) < -\varepsilon \ln \varepsilon - (1-\varepsilon)\ln(1-\varepsilon) = 0.056\cdots < 0.06$.

Discussion.—The obtained results solve in the positive the conjecture raised in Refs. [19,25]. Note that Müller [20] proved this conjecture for classical systems by showing an elaborate way to explicitly construct a catalyst, which is completely different from our approach. Thus, our proof restricted to the classical regime serves as an alternative proof of Müller's.

In this work, we have considered Gibbs-preserving maps as thermodynamic processes instead of thermal operations, while thermal operations are often regarded as proper operations in terms of the resource theory of thermodynamics. These two classes of operations are equivalent in the classical regime [12,46], while some Gibbs-preserving maps cannot be implemented by thermal operations in the quantum regime [47]. The original conjecture of Ref. [19] is about Gibbs-preserving maps, and a stronger conjecture with quantum thermal operations was raised in Ref. [20]. However, Refs. [25,48] solved the latter stronger conjecture in the negative by proving that coherence cannot be broadcast. In other words, no thermal operation converts an incoherent initial state to any coherent state even with the aid of a correlated catalyst. This implies that one should consider a broader class of operations than thermal operations in order to enable characterization of state convertibility by the KL divergence. In the present work, we focus on Gibbs-preserving maps that can still give a positive answer to the original conjectures [19,25]. However, we expect that there may be a pathway to reduce Gibbs-preserving maps to thermal operations in the context of a correlated catalyst. For example, as considered in Ref. [27], the assistance of a small amount of coherence carried by an auxiliary system may enable such reduction in the asymptotic limit (i.e., the large catalyst limit in our setup). Investigation of such a direction is an important future problem.

We also note that Müller [20] performs trivialization of the catalyst Hamiltonian, but we did not. Here, we say a catalyst trivialized when the Hamiltonian of the catalyst is trivial. The hardness of trivialization in the fully quantum regime comes from the fact that merging and splitting states are irreversible due to decoherence in quantum systems. Owing to this difficulty, we did not trivialize the catalyst in the present work.

Besides the correlated classical cases [17,18,20,49], in some setups of quantum thermodynamics and resource theories, a single thermodynamic potential with the KL divergence also appears [4,13,26–28,50–53]. However, those previous results are different from our result in some important aspects: Some of them allow small changes in the catalyst [4] or other external systems [50–53] (instead they consider more restricted classes of operations compared to Gibbs-preserving maps), and some others consider asymptotic (macroscopic) conversion [13,26–28]. It is yet interesting to see that the same thermodynamic potential appears in these various setups.

Meanwhile, we can further extend Theorem 1 to general CPTP maps by employing a similar proof idea. This is about the quantum counterpart of catalytic d-majorization (also called relative majorization) [6,8]:

Theorem 3.—Consider four quantum states ρ, ρ', η, and η' of the system S. Then, $S(\rho||\eta) \geq S(\rho'||\eta')$ holds if and only if there exists a catalyst C and its two states c, d, and a CPTP map which converts $\eta \otimes d$ to $\eta' \otimes d$ and $\rho \otimes c$ to τ satisfying (i) $\mathrm{Tr}_S[\tau] = c$, (ii) $\mathrm{Tr}_C[\tau]$ is arbitrarily close to ρ', (iii) the correlation between S and C in τ is arbitrarily small.

We present the proof of this theorem in the Supplemental Material [30]. This theorem almost solves the conjecture of Rethinasamy and Wilde [21], who proved the classical case. The only difference between our result and the conjecture is that we did not trivialize catalyst c.

Meanwhile, Brandao, and Gour [54] have established that various resource theories concerning asymptotic state conversion with a small error are characterized by the KL divergence. Their result applies to the resource theories of entanglement, coherence, contextuality, and stabilizer computation. By employing our technique (in particular, Step 3 of the proof), we see that the KL divergence also serves as a single monotone in these single-shot resource theories with a correlated catalyst. More generally, our approach developed in the present Letter sheds new light on single-shot resource theories with a catalyst, as recently demonstrated in Ref. [55].

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