Correlation in Catalysts Enables Arbitrary Manipulation of Quantum Coherence

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Quantum resource manipulation may include an ancillary state called a catalyst, which aids the transformation while restoring its original form at the end, and characterizing the enhancement enabled by catalysts is essential to reveal the ultimate manipulability of the precious resource quantity of interest. Here, we show that allowing correlation among multiple catalysts can offer arbitrary power in the manipulation of quantum coherence. We prove that *any* state transformation can be accomplished with an arbitrarily small error by covariant operations with catalysts that may create a correlation within them while keeping their marginal states intact. This presents a new type of embezzlement-like phenomenon, in which the resource embezzlement is attributed to the correlation generated among multiple catalysts. We extend our analysis to general resource theories and provide conditions for feasible transformations assisted by catalysts that involve correlation, putting a severe restriction on other quantum resources for showing this anomalous enhancement, as well as characterizing achievable transformations in relation to their asymptotic state transformations. Our results provide not only a general overview of the power of correlation in catalysts but also a step toward the complete characterization of the resource transformability in quantum thermodynamics with correlated catalysts.

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Introduction.—Quantum superposition, also known as *quantum coherence*, is one of the most striking quantum features and also a useful operational resource in quantum metrology [1], quantum clock [2], and work extraction [3]. In quantum thermodynamics, the presence of coherence is considered as the main source of difference between semiclassical and quantum setups [4]. Under the presence of a conserved quantity such as Hamiltonian, one is restricted to the operations that cannot create coherence. These operations, known as *covariant* operations, are subject to many restrictions originating from the superselection rule [5–11], while preshared coherent states can lift their operational capability [12–14]. This motivates us to obtain a precise understanding of how one could quantify and efficiently manipulate coherence, for which a resource-theoretic approach has been proven useful [15–17].

Characterizing the possible state transformations under given accessible operations is a central problem in any operational setting with physical restrictions. To understand the fundamental resource transformability, one needs to consider an ancillary system serving as a *catalyst*, which keeps its form at the end of the transformation. Several possible scenarios for catalytic transformations have been proposed. The first scenario considers an *uncorrelated catalyst* τ that enables the transformation from $\rho \otimes \tau$ to $\rho' \otimes \tau$ [18–27]. Although uncorrelated catalysts can enhance state transformation in some settings such as entanglement theory [18,19] and quantum thermodynamics [22,23], any pure uncorrelated catalyst fails to change the power of coherence transformation by covariant operations [28,29].

The second scenario extends the uncorrelated catalysts by allowing correlation between the system and the catalytic system at the end of the protocol, where we consider a transformation from $\rho \otimes \tau$ to $\tilde{\rho}_{SC}$ such that $\mathrm{Tr}_C\, \tilde{\rho}_{SC} = \rho'$ and $\mathrm{Tr}_S\, \tilde{\rho}_{SC} = \tau$, in which we call τ a *correlated catalyst* [30–39]. The power of correlated catalysts in covariant operations was discussed in terms of coherence broadcasting, where it was shown that correlated catalysts do not allow covariant operations to create finite coherence from zero coherence [40,41].

These observations on the limitations of catalysts in coherence transformation motivate us to investigate other forms of catalysts that could enhance covariant operations. An interesting setting was offered in quantum thermodynamics. Lostaglio *et al.* [42] considered transformations with multiple catalysts where correlation can be present *among* the catalysts at the end of the transformation, i.e., from $\rho \otimes \tau_{C^{(0)}} \ldots \otimes \tau_{C^{(K-1)}}$ to $\rho' \otimes \tau_{C^{(0)} \ldots C^{(K-1)}}$ while the marginal state of $\tau_{C^{(0)} \ldots C^{(K-1)}}$ on each catalytic system $C^{(j)}$ remains as the original catalyst $\tau_{C^{(j)}}$. They showed that quasiclassical transformations by thermal operations [43] in this form are characterized solely by the free energy, surpassing the enhancement provided by uncorrelated catalysts [22]. Although the perfect reusability is generally lost due to the correlation generated among the final state of the catalysts, we follow the terminology in Ref. [31] and call such a finite set of states $\otimes_{i=0}^{K-1} \tau_{C^{(j)}}$ *marginal catalysts*.

Characterizing the capability of covariant operations with marginal catalysts will provide insights into an ultimate coherence manipulability, as well as differences in operational capability of covariant operations and thermal operations, the latter of which is a subclass of the former. Although significant progress has been made for qubit coherence transformation [29], the potential of marginal catalysts in general coherence transformation has still been left unclear.

Here, we show that correlation among catalysts can completely remove the aforementioned limitations and even provide unlimited power to coherence manipulation. We prove that covariant operations assisted by marginal catalysts enable *any* state transformations with arbitrary precision, making a high contrast to coherence transformation with the other catalytic settings. Furthermore, we discuss the underlying mechanism of this phenomenon from the viewpoint of general resource theories of quantum states [9,44–58]. We show that an arbitrary state transformation is forbidden in a wide class of resource theories, establishing the peculiarity of quantum coherence among other quantum resources. We also relate single-shot catalytic transformations to the asymptotic transformation in general resource theories and exactly characterize feasible state transformations for several important settings such as quantum thermodynamics, entanglement, and speakable coherence [59,60] with the resource measures based on the relative entropy [61], offering them with an operational meaning in terms of extended classes of single-shot catalytic transformations.

Arbitrary state transformation.—For an arbitrary system X with dimension d_X, let $\mathcal{D}(X)$ be the set of quantum states defined in X and $H_X = \sum_{i=0}^{d_X-1} E_{X,i}|i\rangle\langle i|_X$ be its Hamiltonian where $|i\rangle_X$ is an energy eigenstate. When multiple systems $X_0, X_1, \ldots, X_{N-1}$ are involved, we consider the total Hamiltonian over the systems in the additive form as $H_{X_0\ldots X_{N-1}} = \sum_{i=0}^{N-1} H_{X_i} \otimes \mathbb{I}_{\bar{i}}$ where $\bar{i}$ refers to the systems other than the ith system. Coherence between eigenstates with distinct energies can be quantitatively analyzed in the resource theory of asymmetry with U(1) group [62]. Resource theories are frameworks accounting for the quantification and manipulation of precious quantities with respect to freely accessible quantum states and dynamics under given physical settings [44]. The resource theory of asymmetry considers states without coherence, i.e., invariant under time translation, as free states and *covariant* channels as free operations. We call a channel $\mathcal{E}\colon \mathcal{D}(A) \to \mathcal{D}(B)$ covariant if its action is invariant under time translation, i.e., $e^{-iH_B t}\mathcal{E}(\rho)e^{iH_B t} = \mathcal{E}(e^{-iH_A t}\rho e^{iH_A t})$ for any ρ and t. Importantly, covariant operations cannot create coherence from incoherent states, making coherence a precious quantum resource under the situation where only covariant operations are accessible. Such a situation arises when energy-conserving dynamics are concerned. It is known that a map $\mathcal{E}$ is covariant if and only if it can be

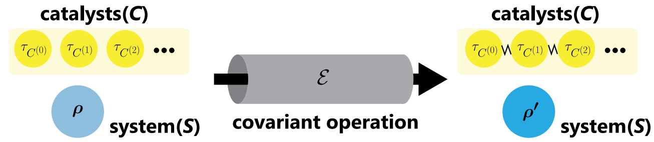

FIG. 1. Schematic of marginal-catalytic covariant transformations. Each catalyst should get back to the original state, while it can correlate with the system and other catalysts.

implemented by an energy-conserving unitary U_{SE} satisfying $[U_{SE}, H_{SE}] = 0$ as $\mathcal{E}(\rho) = \mathrm{Tr}_E[U_{SE}(\rho \otimes \sigma)U_{SE}^\dagger]$ where ρ is an arbitrary state and σ is an ancillary incoherent state [17,63]. If the ancillary state is restricted to the Gibbs state in E, the channels in this form coincide with the thermal operations [43]. Therefore, covariant operations can be seen as an operation that focuses on the coherence part of the resource in quantum thermodynamics, and clarifying the difference in operational power between covariant operations and thermal operations under the same catalytic setting will help pinpoint the roles played by classical athermality and quantum coherence [64].

We first formally define the marginal-catalytic covariant transformation as follows. (See also Fig. 1).

Definition 1.—$\rho \in \mathcal{D}(S)$ is transformable to $\rho' \in \mathcal{D}(S')$ by a marginal-catalytic covariant transformation if there exists a constant K and a state $\otimes_{j=0}^{K-1} \tau_{C^{(j)}}$ in a finite-dimensional system $\otimes_{j=0}^{K-1} C^{(j)}$ and a covariant operation $\mathcal{E}\colon \mathcal{D}(SC^{(0)}\ldots C^{(K-1)}) \to \mathcal{D}(S'C^{(0)}\ldots C^{(K-1)})$ such that

$$\mathcal{E}(\rho \otimes \tau_{C^{(0)}} \ldots \otimes \tau_{C^{(K-1)}}) = \rho' \otimes \tau_{C^{(0)}\ldots C^{(K-1)}}$$
$$\mathrm{Tr}_{\overline{C^{(j)}}}\tau_{C^{(0)}\ldots C^{(K-1)}} = \tau_{C^{(j)}}, \quad \forall\ j, \tag{1}$$

where $\mathrm{Tr}_{\bar{X}}$ denotes the partial trace over the systems other than X.

Marginal catalysts can be seen as an extension of a catalytic transformation in the sense that the final state keeps some properties of the initial state in an exact form. Our main focus here is not to keep the repeatable property of catalytic transformations but to investigate how the state transformability could be enhanced by the nontrivial change of the setting regarding the correlation. Nevertheless, we can also motivate this specific setting operationally; although the final catalyst as a whole is not reusable in the next round, *some parts of it* can be reused multiple times without the degradation of performance in the desired state transformation. We discuss this *partial reusability* of marginal catalysts in the Supplemental Material [65].

Our main result shows that, despite the apparent limitations in catalytic coherence transformation with uncorrelated and correlated catalysts [28,29,40,41], marginal catalysts can provide extraordinary power—in fact, any state transformation can be accomplished by a marginal-catalytic covariant transformation with arbitrary accuracy.

Theorem 2.—For any $\rho \in \mathcal{D}(S)$, $\rho' \in \mathcal{D}(S')$, and $\epsilon > 0$, ρ can be transformed to a state $\rho'_\epsilon \in \mathcal{D}(S')$ such that $\frac{1}{2}\|\rho' - \rho'_\epsilon\|_1 \le \epsilon$ by a marginal-catalytic covariant transformation.

We sketch our proof in a later section, while deferring the detailed proof to the Supplemental Material [65].

Theorem 2 implies that marginal catalysts can trivialize coherence transformations, fully generalizing the result in Ref. [29] established for the qubit state transformations to those involving arbitrary Hilbert spaces of finite dimensions. Notably, the result can be extended to the implementation of an arbitrary *quantum channel* (see the Supplemental Material [65]).

Our result makes a high contrast to quantum thermodynamics, in which state transformations by marginal-catalytic thermal operations respect the ordering of the free energy [42]. A related phenomenon is known as embezzlement [67], where a negligibly small error in a catalyst enables an arbitrary transformation. We stress that marginal-catalytic transformations recover the marginal states *exactly* and are fundamentally different from the mechanism of the well-known embezzlement. In fact, as discussed below, the trivialization of state transformations by marginal catalysts is an unusual phenomenon, which shows a clear contrast to the embezzlement seen in a broad class of resource theories from entanglement [67] to quantum thermodynamics [22].

Comparison to other quantum resource theories.—It may appear odd that one can create unbounded coherence in the main system while keeping the reduced states of the catalysts intact. To get insights into this phenomenon, let us consider whether marginal catalysts could provide similar enhancement in other quantum resource theories. Each resource theory is equipped with a set $\mathcal{F}$ of free states and a set $\mathcal{O}_\mathcal{F}$ of free operations [44]. For given these sets, one can define a resource measure $\mathfrak{R}$, which evaluates zero for any free state, i.e., $\mathfrak{R}(\sigma) = 0$ for any $\sigma \in \mathcal{F}$, and does not increase under free operations, i.e., $\mathfrak{R}(\mathcal{E}(\rho)) \le \mathfrak{R}(\rho)$ for any ρ and for any $\mathcal{E} \in \mathcal{O}_\mathcal{F}$. We particularly call it superadditive if $\mathfrak{R}(\rho_{12}) \ge \mathfrak{R}(\mathrm{Tr}_2[\rho_{12}]) + \mathfrak{R}(\mathrm{Tr}_1[\rho_{12}])$ for any state $\rho_{12} \in \mathcal{D}(S_1 \otimes S_2)$ and tensor-product additive if $\mathfrak{R}(\rho_1 \otimes \rho_2) = \mathfrak{R}(\rho_1) + \mathfrak{R}(\rho_2)$ for any ρ_1 and ρ_2.

The setup of catalytic transformations can be extended to any resource theory. We say that ρ is transformable to ρ' by a *correlated-catalytic free transformation* if there exists a finite-dimensional catalyst τ such that $\rho \otimes \tau$ can be transformed to $\tilde{\rho}_{SC}$ with $\mathrm{Tr}_C\tilde{\rho}_{SC} = \rho'$, $\mathrm{Tr}_S\tilde{\rho}_{SC} = \tau$ by a free operation. Then, we can show that any resource measure satisfying the above two properties remains a valid resource measure under the two catalytic transformations involving correlation. (See the Supplemental Material [65] for a proof).

Proposition 3.—For any given $\mathcal{F}$ and $\mathcal{O}_\mathcal{F}$, suppose that a resource measure $\mathfrak{R}$ satisfies the superaddtivity and the tensor-product additivity. Then, if ρ is transformable to ρ' by a marginal-catalytic or correlated-catalytic free transformation, $\mathfrak{R}(\rho) \ge \mathfrak{R}(\rho')$ holds.

We remark that a related observation was made in the context of quantum thermodynamics [31]. This puts a severe constraint on the possibility of arbitrary state transformation. If there exists even a *single* resource measure satisfying the superadditivity and the tensor-product additivity, then marginal catalysts do not enable an arbitrary state transformation as long as the resource measure is faithful, i.e., any nonfree state takes a nonzero value. (See also Refs. [41,68] and discussion below.) In fact, one can find such measures in many resource theories, including quantum thermodynamics [31], entanglement [69,70], and speakable coherence (superposition between given orthogonal states) [59,60,71], prohibiting the anomalous resource transformation with marginal or correlated catalysts.

On the other hand, Theorem 2 and Proposition 3 imply that there *never* exists a coherence measure that is superadditive, tensor-product additive, and faithful. Our results parallel previous observations; recent analytic proofs of the violation of superadditivity of coherence measures employ covariant operations that can amplify the sum of local coherence indefinitely [41,68]. Examples of tensor-product additive and faithful coherence measures include the Wigner-Yanase skew information [16,72] and other metric-adjusted skew informations [73,74], which indeed violate the superadditivity [68,75,76]. More generally, it was shown that any faithful measure of asymmetry cannot be superadditive [41]. These results together with Theorem 2 and Proposition 3 indicate an intimate connection between the anomalous coherence amplification and the violation of the superadditivity of coherence measures.

Besides the necessary conditions established in Proposition 3, we can also formulate sufficient conditions using a general method of converting asymptotic transformations to one-shot correlated-catalytic transformations [36]. (See the Supplemental Material [65] for a proof.)

Proposition 4.—For any given $\mathcal{F}$ and $\mathcal{O}_\mathcal{F}$, suppose that $\mathcal{O}_\mathcal{F}$ includes the relabeling of the classical register and free operations conditioned on the classical register. Then, if ρ is asymptotically transformable to ρ', there exists a free transformation from ρ to ρ' with a correlated catalyst as well as marginal catalysts with an arbitrarily small error.

Proposition 4 shows that sufficient conditions for asymptotic transformations are directly carried over to single-shot catalytic transformations. This particularly implies that, in a general class of convex resource theories, the regularized relative entropy measure provides a sufficient condition for these single-shot catalytic transformations under asymptotically resource nongenerating operations, given the generalized quantum Stein's lemma holds [46,77] (see also Ref. [78] for the recent argument about the incompleteness in the proof of the generalized quantum Stein's lemma).

Combining Propositions 3 and 4, we arrive at the complete characterizations of marginal- and correlated-catalytic free transformations for various settings in which the resource measures governing asymptotic transformations satisfy the tensor-product additivity and superadditivity. These include several well-known relative entropy based measures, such as the free energy in quantum thermodynamics with Gibbs-preserving operations [36,54,58,79], the entanglement entropy with pure state transformations under local operations and classical communication [37,38,80], and the relative entropy of speakable coherence with several free operations [81–84]. Notably, Propositions 3 and 4 imply the equivalence in the power of correlated and marginal catalysts for these scenarios.

Correlated-catalytic covariant transformations.—Although Theorem 2 reveals the exceptional power of marginal catalysts, the power of correlated catalysts in coherence transformation still remains elusive. In particular, when the initial state has nonzero coherence, neither the coherence no-broadcasting theorem [40,41] nor Proposition 3 prohibits preparing an arbitrary state. We conjecture that a broad class of transformations is possible with correlated catalysts under the presence of initial coherence.

Conjecture 5 (Informal).—Let $\mathcal{C}(\rho)$ be the set of energy differences for which ρ possesses nonzero coherence. Then, for any states ρ and ρ', ρ can be transformed to ρ' with an arbitrarily small error by a correlated-catalytic covariant transformation if and only if every energy difference in $\mathcal{C}(\rho')$ can be written as a sum of integer multiples of energy differences in $\mathcal{C}(\rho)$.

The idea behind this is that correlated-catalytic covariant transformations should be able to amplify and manipulate the coherence of the initial state to realize any degree of coherence for the energy differences that are combinations of the initial ones with nonzero coherence. Thus, if these energy differences cover those of the target state with nonzero coherence, ρ should be transformable to ρ' under a correlated-catalytic covariant transformation.

In the Supplemental Material [65], we present a precise statement of the conjecture and support it by proving the state transformability under a slightly larger class of catalytic covariant operations, together with several other observations.

Proof sketch for Theorem 2.—To prove our claim, it suffices to provide a protocol that prepares a final state from scratch with an arbitrarily small error. Our protocol makes use of the procedure introduced in Ref. [29] as a subroutine, which amplifies coherence in two-level systems using a correlated catalyst by a small amount [Fig. 2(a)]. This can particularly bring a coherent state $\Sigma(\eta) \coloneqq (\mathbb{I}+\eta X)/2$ with $\eta > 0$, $X \coloneqq |0\rangle\langle 1| + |1\rangle\langle 0|$ to another coherent state $\Sigma(\eta')$ with $\eta' > \eta$ using a catalyst $\Gamma(\eta) \coloneqq \frac{1}{2}\{\mathbb{I} + (\sqrt{3}\eta/2)X + [(4-\eta^2)/6]Z\}$, and sequential application of this protocol allows us to realize any coherent state on the X axis of the Bloch sphere (excluding the pure state $\eta = 1$) with marginal-catalytic covariant operations. Although the authors of Ref. [29] claim that the whole sequence of amplification is a correlated-catalytic covariant transformation, their argument is, unfortunately, insufficient—in fact, the total amplification process is marginal catalytic. We extend

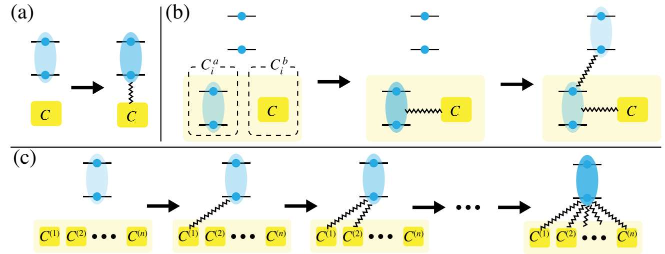

FIG. 2. Schematics for Steps 1 and 2. (a) One cycle of the two-level coherence amplification subroutine. (b) We run the two-level amplification protocol to increase coherence in C^a together with C^b. We transfer this increased coherence to R and restore the state in C^a to the original form. (c) We amplify the coherence generated in R with many rounds of the two-level amplification protocol.

detailed discussions about the two-level coherence amplification subroutine in the Supplemental Material [65].

Our protocol consists of three main steps (Figs. 2 and 3). The first step creates small coherence in an ancillary system, the second step amplifies this coherence and constructs coherent resource states, and the third step uses them to prepare the target state with a covariant operation.

Step 1: Creating small coherence [Fig. 2(b)]. We introduce ancillary system R consisting of two-level subsystems $\{R_i\}_{i=1}^{d_{S'}-1}$ whose Hamiltonians reflect the spectrum of $H_{S'}$ as $H_{R_i} = (E_{S',i} - E_{S',j^\star})|1\rangle\langle 1|_{R_i}$ where $j^\star \in \{0,\ldots,d_{S'}-1\}$ is an arbitrarily chosen integer independent of i. We aim to prepare a coherent state $\Sigma(\eta)$ with $\eta > 0$ for each i. To this end, we introduce catalytic subsystems C_i^a and C_i^b, both of which have the Hamiltonian H_{R_i}. We prepare catalysts $\tau_i^a \coloneqq \Sigma(\eta)$ in C_i^a and $\tau_i^b \coloneqq \Gamma(\eta)$ in C_i^b for some η with $0 < \eta < 1$. We apply a single round of the

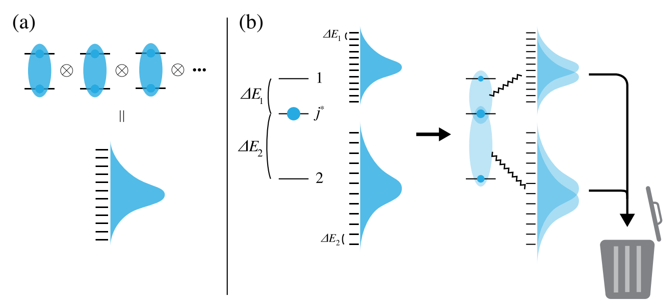

FIG. 3. Schematics for Step 3. (a) Multiple copies of the resource state in R created in Step 1 constitute a state with the binomially distributed energy statistics. (b) We use these coherent states as ancillary coherent resource states to assist the energy transition required for the desired unitary V. The error on the realized unitary from the desired one, V, is quantified by the overlap between the original resource states and the final resource states subject to an energy shift due to the backreaction, which can be made arbitrarily small by creating a sufficiently large resource state. These resource states are discarded at the end of the protocol.

two-level coherence amplification over $\tau_i^a \otimes \tau_i^b$, which increases coherence in C_i^a by a small amount while keeping the reduced state on C_i^b unchanged. We transfer the increased amount of coherence from C_i^a to R_i by applying a covariant unitary over $R_i C_i^a$, creating a nonzero coherence in R_i while bringing the reduced state on C_i^a back to τ_i^a.

Step 2: Amplifying coherence [Fig. 2(c)]. We amplify this nonzero coherence generated in R_i by the two-level coherence amplification using another set of catalysts prepared in $\otimes_{j=0}^{K-1} C_i^{(j)}$ with a large enough integer K. This prepares a state close to $|+\rangle \coloneqq (|0\rangle + |1\rangle)/\sqrt{2}$ in R_i.

Step 3: Prepare the target state (Fig. 3). We repeat Steps 1 and 2 for $L(\gg 1)$ times to prepare a state close to $|+\rangle_{R_i}^{\otimes L}$ for each i, which is a superposition of energy eigenstates with weights according to the binomial distribution. By employing these states as ancillary coherent resource states, we can implement any unitary on S' with arbitrary accuracy by a covariant operation [5,7,12–14,85–87]. Since any pure state on S' can be prepared by applying an appropriate unitary to an incoherent state $|j^\star\rangle_{S'}$, and any mixed state is realized by a probabilistic mixture of pure states, we can obtain the total state whose reduced state on S' is ρ'_ϵ. Finally, the correlation between S' and the catalytic system can be removed by using the technique employed in Ref. [30], where we start with the final state in another catalytic system and swap it with the marginal state created in S'.

The accuracy of the whole protocol is determined by the errors in preparing highly coherent resource states in Step 2 and in approximating unitary in Step 3, both of which can be made arbitrarily small using finite-size catalysts, ensuring any target error $\epsilon > 0$. Also, since the required catalysts except for the final step in removing the correlation do not depend on the target state, they construct a universal family of catalysts applicable to any state transformation if an arbitrarily small correlation is allowed between the main system and catalytic systems.

Conclusions.—We studied catalytic state transformation with marginal catalysts, which allow correlation among multiple catalyst states at the end of the transformation. We showed that marginal catalysts provide exceptional power to coherence transformation, enabling any state transformation with arbitrarily small error. To elucidate the peculiarity of how quantum coherence behaves in catalytic transformations, we compared it to other types of quantum resources by formulating conditions for catalytic state transformations from the perspective of resource quantifiers. We showed that such an anomalous state transformation is impossible in resources such as thermal nonequilibrium, entanglement, and speakable coherence, for which we exactly characterized state transformability with correlated and marginal catalysts in terms of relative entropy resource measures.

An intriguing future direction is to prove or disprove the conjecture on the power of correlated catalysts in coherence transformation, which will provide insights into another interesting problem in quantum thermodynamics, that is, whether the free energy solely determines the state transformability by thermal operations with correlated catalysts if an initial state has finite coherence. Answering this question will pave the way toward a fully general operational characterization of single-shot quantum thermodynamics.

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