Amplifying asymmetry with correlated catalysts

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We investigate the basic constraint on amplifying the asymmetry in quantum states with correlated catalysts. Here a correlated catalyst is a finite-dimensional auxiliary, which exactly preserves its reduced state while allowed to become correlated to the quantum system. Interestingly, we prove that under translationally invariant operations, catalysts in pure states are useless in any state transformation, while with a correlated catalyst in a mixed state, one can enlarge the set of accessible states from an initially asymmetric state. Moreover, we show that the power of a catalyst increases with its dimension, and further, with a large enough catalyst, a qubit state with arbitrarily small amount of asymmetry can be converted to any mixed qubit state. In doing so, we build a bridge between two important results concerning the restrictions on coherence conversion, the no-broadcasting theorem and the catalytic coherence. Our results may also apply to the constraints on coherence evolution in quantum thermodynamics, and to the distribution of timing information between quantum clocks.

Introduction.— Finding out whether a quantum state can be converted to another under a set of restricted operations is a problem originated from the entanglement theory [1], and has recently been studied in a variety of resource theories [2–5]. Moreover, in resource theories such as entanglement [6], athermality [7], coherence [8–11], and quantum randomness [12], catalysts are employed to enhance the ability of state conversion. A catalyst is an ancilla which interacts with the system and then returns to the exact original state. Conventionally, the catalyst requirement is to be uncorrelated to the system after the process [4, 13–15]. Nevertheless, recent researches suggest that the creation of correlations may greatly extends the set of accessible states [16–20]. In particular, in the resource theory of athermality, if this uncorrelation requirement is lifted, the catalyst becomes more powerful, namely, it enables state conversions which are not achievable using an uncorrelated catalyst [16].

Surprisingly, evidences have been uncovered that catalysts might be useless in the resource theory of asymmetry. The no-catalysis theorem, as proved in Ref. [21], states that if a pure state cannot be converted to another pure state using operations which are symmetric under a compact Lie group, then any finite-dimensional catalyst in a pure state can not enable this conversion. Furthermore, by the no-broadcasting theorem of asymmetry [22, 23], the creation of asymmetry in an initially symmetric state is impossible even with a correlated finite-dimensional catalyst, in comparison to the protocol of catalytic coherence [24] where arbitrary amount of coherence between energy levels can be created by interacting the system with an infinite-dimensional catalyst. In order to explore the crossover between the no-broadcasting theorem and the catalytic coherence, we ask the following question: to what extent can a finite-dimensional catalyst enlarge the set of accessible states under symmetric transformations?

Because the creation of correlations between the system and the catalyst may ease the state transformation, and this correlation does not reduce the ability of the catalyst in other state transformations, we allow the catalyst to become correlated to the system on which it act. In this paper, we only impose two restrictions on the catalytic system: (1) it is finite-dimensional; (2) its reduced state is exactly identical before and after the state conversion. Here we first prove a general result that a catalyst can extends the set of accessible states only if it is in a mixed state, which generalizes the no-catalysis theorem in Ref. [21]. Then we show that, in contrast to other resource theories, there is no bound on amplifying the asymmetry with correlated catalysts. That is, any qubit state with arbitrarily small amount of asymmetry can be boosted to a state arbitrarily close to the state with maximal asymmetry, as long as the dimension of the catalyst is large enough. The applications of our results to the constraints on catalytic coherence evolution in quantum thermodynamics, and to the distribution of timing information, are also discussed.

Notions.— In the resource theory of asymmetry, the free states are the so-called symmetric states ρsym which is invariant during the evolution under its Hamiltonian $H$, i.e., $e^{-iHt}ρ_{sym}e^{iHt} = ρ_{sym}$. The free operations are the translationally invariant operations (TIOs), or covariant operations, which are defined as completely-positive and trace-preserving (CPTP) maps $E$ satisfying $E(e^{-iHt}ρe^{iHt}) = e^{-iHt}E(ρ)e^{iHt}$, $∀ρ$. The set of states that can be converted to from a state $ρ$ via TIO is called the TIO cone of $ρ$, labeled as $C_{TIO}(ρ) ≡ \{ρ'|ρ' = E(ρ), E ∈ TIO\}$.

The correlated catalytic TIO (CCTIO) on a system $S$ with Hamiltonian $H_S$ are implemented by coupling $S$ to a finite-dimensional auxiliary $C$ with Hamiltonian $H_C$ via a global translationally invariant operation which preserves the reduced state of $C$. Here by “correlated”, we mean that we lift the traditional restriction that the catalyst is uncorrelated from the system in the output. Precisely, we say that a state $ρ$ can be transformed to $ρ'$ by CCTIO, if there exist a finite-dimensional auxiliary system in state $σ_C$, and a global covariant operation $E$ satisfying $E(e^{-\iota(H_S+H_C)\frac{t}{2}}⋅e^{\iota(H_S+H_C)\frac{t}{2}}) =$
$$e^{-i(H_S + H_C)t}E(\cdot)e^{i(H_S + H_C)t}$$ such that
$$E(\rho \otimes \sigma) = \rho' |\sigma\rangle.$$  \hfill (1)

Here the label $\rho' |\sigma\rangle$ means a bipartite state of $S$ and $C$, whose reduced states are $\rho'$ on $S$ and $\sigma$ on $C$. Notably, the state of $C$ is exactly identical before and after the action of $E$. The set of states achievable under CCTIO from $\rho$ is called the CCTIO cone of $\rho$, and the auxiliary $C$ is called the correlated catalyst. When the dimension of $C$ is restricted to $d$, the CCTIO cone of $\rho$ is labeled as $C_{\text{CCTIO}}^{(d)}(\rho)$.

Catalysts in pure states are useless.— When the catalyst is in a pure state $\sigma = |\phi\rangle \langle \phi|$, the transformation in Eq. (1) reads
$$E(\rho \otimes |\phi\rangle \langle \phi|) = \rho' \otimes |\phi\rangle \langle \phi|.$$ \hfill (2)

Because it is required that the state of the catalyst is exactly retained, the purity of $\sigma$ ensures that $S$ and $C$ are not correlated in the output. For an asymmetry monotone $I(\rho)$ which is additive on tensor products (such as quantum fisher information and the Wigner-Yanase skew information, see Appendix B for a brief review), Eq. (2) implies that $I(\rho') + I(|\phi\rangle \langle \phi|) = I(\rho' \otimes |\phi\rangle \langle \phi|) \leq I(\rho \otimes |\phi\rangle \langle \phi|) = I(\rho) + I(|\phi\rangle \langle \phi|)$, and hence, $I(\rho') \leq I(\rho)$. It means that with a finite-dimensional pure catalytic state, the asymmetry monotone $I$ can never be increased.

Yet, it is not as simple to see whether other asymmetry monotones, which are not additive on tensor products, are also monotonic under the catalytic transformation in Eq. (2). In the following theorem, we show a stronger result. Namely, any catalyst in a pure state cannot enable state transformations which are not achievable by TIO. The proof of this theorem is in Appendix A.

**Theorem 1.** If the state $\rho$ cannot be transformed to $\rho'$ under a TIO, then transformation $\rho \otimes |\phi\rangle \langle \phi| \rightarrow \rho' \otimes |\phi\rangle \langle \phi|$ under TIO is also not achievable for any choice of pure catalytic state $|\phi\rangle$.

This theorem is our first main result. It generalizes the no-catalysis theorem [21], in which the states $\rho$ and $\rho'$ are restricted to pure states, and indicates that catalysts in pure states are useless in any state transformation under covariant operations.

Extending the set of accessible states with correlated catalysts.— When the catalyst is in a mixed state $\sigma$, it may become correlated to the system after the transformation. Because there are correlated states $\rho' |\sigma\rangle$ such that $I(\rho' |\sigma\rangle) < I(\rho') + I(\sigma)$ [22], it is possible that $I(\rho') > I(\rho)$, i.e., a correlated catalyst may enable state transformations which are not achievable under TIOs.

In Ref. [19], a stationary machine was designed to control and amplify the energetic coherence in quantum systems. It gives an evidence that with the help of a correlated catalyst (the stationary machine), one can achieve state transformations which can not be realized via TIOs. Nevertheless, it is not quite straightforward to see whether the state of the catalyst (machine) is exactly identical after each round, due to the approximations in deriving the master equations.

Here we give an analytic example, which shows that a global TIO acting on $S$ and $C$ can transform the state of $S$ to a state not achievable under TIOs, while strictly preserve the reduced state of $C$. Consider a system qubit with Hamiltonian $H_S = \frac{1}{2} \sigma_z^2$ and a catalytic qubit with Hamiltonian $H_C = \frac{1}{2} \sigma_z^2$, where $\sigma_x, \sigma_y, \sigma_z$ denote the Pauli matrices and $\Delta > 0$ is the energy gap. Initially, the two-qubit state of the system and the catalyst reads $\rho(\eta) \otimes \sigma_C^{\perp}(\eta)$, where
$$\rho(\eta) = \frac{1}{2}(1 + \eta \sigma^x), \; 0 < \eta < 1$$ \hfill (3)
$$\sigma_C^{\perp}(\eta) = \frac{1}{2} + \frac{\sqrt{3}\eta}{4} \sigma^x + \frac{4 - \eta^2}{12} \sigma^z.$$ \hfill (4)

After the application of the global covariant operation $E^{\dagger}(\cdot) = K(\cdot)K_1$, we obtain a correlated two-qubit state, whose reduced states are
$$\rho' = \frac{1}{2}(1 + \frac{25\eta - \eta^3}{48} \sigma^x + \frac{1 - \eta^2}{6} \sigma^z)$$ \hfill (6)

on $S$ and $\sigma_C^{\perp}(\eta)$ on $C$.

The TIO cone of $\rho(\eta)$ reads $C_{\text{TIO}}[\rho(\eta)] = \{ |\xi\rangle \langle \xi| = \frac{1}{2}(1 + r \cos \phi \sigma^x + \sin \phi \sigma^y) + r_z \sigma_z, r_z \in [-1, 1], \phi \in [0, 2\pi], 0 \leq r \leq \eta \sqrt{1 - |r_z|^2} \}$ (see Appendix D for a proof). Hence for any state $|\xi\rangle \in C_{\text{TIO}}[\rho(\eta)]$, it holds that $\text{Tr}(|\xi\rangle \langle \xi|) \leq \eta$. Because $\text{Tr}[(\rho') |\sigma_C^{\perp}(\eta)\rangle \langle \sigma_C^{\perp}(\eta)|] > \eta$, we have $\rho' \notin C_{\text{TIO}}[\rho(\eta)]$. It means that the state transformation from $\rho(\eta)$ to $\rho'(\eta)$, which is not achievable by TIO, can be enabled by employing a correlated catalytic qubit. Notably, the reduced state $\sigma_C^{\perp}(\eta)$ of $C$ is exactly identical before and after the action of the global TIO $E^{\dagger}$, which excludes the phenomenon of embezzlement.

Next we analyse whether the power of catalysis is affected by the dimension $d$ of $C$. Here we focus on the whole set of accessible states from a given initial state, instead of the amplification of some asymmetry measure, in order to avoid the dependence on the choice of asymmetry measure. Specifically, we numerically calculate the CCTIO cone with $d = 2, 3$ of state $\rho(\eta)$. The numerical method in obtaining this result is detailed in Appendix E. In the Bloch presentation, the basic structure of the CCTIO cone of a qubit state is that it is rotationally symmetric about the $z$ axis. For state $\rho(\eta)$, it is also symmetric about the $xy$ plane (see Appendix D for a proof). In Fig. 1, we plot $C_{\text{TIO}}[\rho(\eta)]$, $C_{\text{CCTIO}}^{(2)}[\rho(\eta)]$, and $C_{\text{CCTIO}}^{(3)}[\rho(\eta)]$ within the $xz$ plane with $x \geq 0$ and $z \geq 0$. Clearly, $C_{\text{CCTIO}}^{(3)}$ is strictly larger than $C_{\text{CCTIO}}^{(2)}$. It means that, with a correlated catalyst of higher dimension, more state conversions can be realized.
Unbounded amplification of asymmetry.—Here we propose a protocol for amplifying the asymmetry of a qubit using a finite-dimensional correlated catalyst. This protocol shows that, for any input qubit state ρ satisfying [ρ, HS] ≠ 0, it can be transformed to a state arbitrarily close to ρ† = 1/2(I + σz) via CCTIO, given that the dimension of catalyst is large enough (but still finite). Notice that ρ† is the qubit state with the maximum amount of asymmetry, when a variety of asymmetry measures, including those based on the skew information, relative entropy, robustness, etc., are employed.

Our protocol is a generalization of the analytic example in the last section. Here the catalyst consists of N particles, labeled as C1, C2, . . . , CN, each of which contains two qubits C1,i and C2,i. The Hamiltonian of the i-th particle Ci reads HCl = HC1,i + HC2,i, with HC1,i = 1/2σz and HC2,i = 1/2 2σz. The reduced state of Ci is set in the form

\[ \sigma(\eta_i) = \frac{1}{2} \sigma^+(\eta_i) \otimes |\uparrow\rangle\langle \uparrow| + \frac{1}{2} \sigma^-(\eta_i) \otimes |\downarrow\rangle\langle \downarrow|, \]

where \( \sigma^+(\eta_i) \) as in Eq. (4) and \( \sigma^-(\eta_i) \equiv \sigma^z \sigma^-(\eta_i) \sigma^z \) are states of qubit C1,i, the parameter \( \eta_i \) depends on the state of the system qubit, and |\uparrow\rangle \) and |\downarrow\rangle \) are energy eigenstates of qubit C2,i.

To start with, we convert the initial asymmetric state of the system qubit to \( \rho(\eta_i) \) with \( \eta_i > 0 \) via TIO. Then the system qubit is coupled to each of the N particles via TIO consequently. In each turn, the TIO reads

\[ E_i = E_i^+ \otimes \Pi^+ + E_i^- \otimes \Pi^-, \]

where the two-qubit TIOs \( E_i^+ \) and \( E_i^- \) are applied to \( S \) and \( C_{i1} \), and \( \Pi^+ \) and \( \Pi^- \) are projectors to the energy eigenstates of qubit C1,i. Here \( E_i^- \) is the form of Eq. (5), and \( E_i^+ \equiv U^\dagger \circ E_i^- \circ U \), with \( U^\dagger(\cdot) = \sigma^x \otimes \sigma^x(\cdot) \sigma^x \otimes \sigma^x \) and \( \circ \) denoting the composition of quantum operations. It can be checked by definition that \( E_i \) is a TIO on the composite system SC1,iC2,i.

Effectively, we have

\[ E_i[\rho(\eta_i) \otimes \sigma(\eta_i)] = \frac{1}{2} \rho^+(\eta_i) |\uparrow\rangle\langle \uparrow| + \frac{1}{2} \rho^-(\eta_i) |\downarrow\rangle\langle \downarrow| \]

\[ = \rho(\eta_{i+1}) |\sigma(\eta_i)|, \]

where \( \rho^+(\eta_i) \equiv \sigma^+ \rho^+(\eta_i) \sigma^+ \), and \( \eta_{i+1} = \eta_i + \frac{1}{2}\Delta \eta_i(1 - \eta_i^2) \) for \( i = 1, \ldots, N \).

The operations involved in our protocol are CCTIO. Because \( E_i \) are TIO, the composed operation \( E \equiv E_N \circ \cdots \circ E_1 \) is also TIO. In the following, we prove that \( \text{Tr}_i[\mathcal{E}(\rho \otimes \sigma)] = \sigma \), i.e., the N-particle catalytic state \( \sigma \) exist such that it is not affected by \( E \). Firstly, the reduced state of particle \( C_i \) is affected neither by \( E_i \) due to Eq. (9), nor by \( E_i' \) with \( i' \neq i \), so we have \( \text{Tr}_i[\mathcal{E}(\rho \otimes \Sigma)] = \sigma(\eta_i), \forall i \), where \( \Sigma \) is any N-partite state satisfying \( \text{Tr}_i(\Sigma) = \sigma(\eta_i), \forall i \), and \( \text{Tr}_i ) \) means partial trace on all systems except \( C_i \). Then let \( \sigma^{(1)} = \sigma(\eta_1) \otimes \cdots \otimes \sigma(\eta_N) \) and \( \sigma^{(j+1)} = \text{Tr}_i[\mathcal{E}(\rho \otimes \sigma^{(j)})] \), and we have \( \text{Tr}_i(\sigma^{(j)}) = \sigma(\eta_j), \forall i, j \). Now we define \( \sigma = \lim_{n \to \infty} \frac{1}{n} \sum_{j=1}^{n} \sigma^{(j)} \), and then

\[ \text{Tr}_S[\mathcal{E}(\rho \otimes \sigma)] = \lim_{n \to \infty} \frac{1}{n} \sum_{j=0}^{n} \text{Tr}[\mathcal{E}(\rho \otimes \sigma^{(j)})] \]

\[ = \lim_{n \to \infty} \frac{1}{n} \sum_{j=0}^{n} \sigma^{(j+1)} \]

\[ = \sigma + \lim_{n \to \infty} \frac{1}{n} [\rho^{(n+1)} - \sigma^{(1)}] = \sigma. \]

Now let us illustrate that a state arbitrary close to \( \rho^+ \) can be achieved by our protocol, if the initial state is not symmetric and the catalyst is large enough (but still finite). Specifically speaking, from any qubit state \( \rho \) satisfying \( [\rho, HS] \neq 0 \) and \( \forall \epsilon > 0 \), the state \( \rho(\eta') \) with \( 1 - \epsilon < \eta' < 1 \) can be achieved with finite \( N \). From Eq. (9), after the action of \( E_i \), the parameter \( \eta \) is increased by \( \Delta \eta_i \equiv \eta_{i+1} - \eta_i = \frac{1}{2}(1 - \eta_i^2) \). Because \( \eta_i > 0 \) and \( \eta' < 1 \), we have \( 0 < \eta_i < 1 \) for all \( i \), and hence \( \Delta \eta_i > 0 \). Therefore, we need at most \( N_{\text{max}} \equiv \min_{i} \Delta \eta_i \) particles in the catalyst, where \( N_{\text{max}} \equiv \min_{i} \Delta \eta_i \). Apparently, \( N_{\text{max}} \) is finite because \( \eta' - \eta_i < 1 \) and \( \Delta \eta_i > 0 \).

The condition \( [\rho, HS] \neq 0 \) on the input state \( \rho \) is essential for our protocol. If it is satisfied, we have \( \rho(\eta_i) \in C_{\text{TIO}}(\rho) \) with \( \eta_i > 0 \) (see Appendix D). Otherwise, \( \eta_i = 0 \), and hence, \( \Delta \eta_i = 0, \forall i \). It means that, if we start from a symmetric state, it remains symmetric for arbitrary large \( N \). This is compatible with the no-broadcasting theorem of asymmetry [22, 23]. Namely, by employing a correlated catalyst, one can amplify the asymmetry in an asymmetric state, instead of creating asymmetry in a symmetric state.

Here we mention that in general, it is impossible to reach \( \rho^+ \) exactly with any finite-dimensional catalyst. Suppose there are a finite-dimensional catalytic state \( \sigma_C \) and a global TIO \( E \) such that \( E(\rho \otimes \sigma_C) = \rho^+ \otimes \sigma_C \). Because \( \rho^+ \) is a pure state, the bipartite state in the output is not correlated, i.e., \( \rho^+ \otimes \sigma_C = \rho^+ \otimes \sigma_C \). It follows that \( I(\rho) \geq I(\rho^+) \), which
holds only if \( \rho \) is on the equator of the Bloch sphere. Therefore, it is impossible to transform any state (other than the ones equivalent to \( \rho^+ \) by symmetric unitaries) exactly to \( \rho^+ \) by any CCTIO process.

This protocol can be generalized to transform an asymmetric state to any mixed qubit state. It follows that the CCTIO cone of an asymmetric qubit state includes all but the surface states of the Bloch ball. In other words, let \( \rho \) and \( \rho' \) be two qubit states. The state conversion from \( \rho \) to \( \rho' \) is achievable under CCTIO, as long as \( [\rho, H_S] \neq 0 \) and \( \rho' \) is a mixed state. Because for any state \( \rho \), there is a state \( \rho_1 \) arbitrarily close to \( \rho \) such that \( [\rho_1, H_S] \neq 0 \), and similarly, for any state \( \rho' \), we can find a mixed state \( \rho' \), which is arbitrarily close to \( \rho' \), we arrive at the following theorem.

**Theorem 2.** For any pair of qubit states \( \rho \) and \( \rho' \), the state conversion from \( \rho \) to \( \rho' \) is achievable under CCTIO.

**Application to the resource theory of athermality.**—In the resource theory of athermality [25], the free operations are the thermal operations, which can be implemented by interacting the system to a reservoir at inverse temperature \( \beta \) and then shutting down the interaction after a while. Here the interaction preserves the total energy, which makes sure that thermal operations are translationally invariant.

It was shown in Ref. [16] that, for any pair of symmetric states \( \rho \) and \( \rho' \), there exist a finite-dimensional catalytic system in state \( \sigma \) and a global thermal operation \( \mathcal{E}_{\text{TO}} \) such that \( \mathcal{E}_{\text{TO}}(\rho \otimes \sigma) = \rho' \otimes \sigma \), where \( \rho' \) is arbitrarily close to \( \rho' \), if and only if \( F(\rho) \geq F(\rho') \). Here the free energy \( F(\rho) := \text{Tr}(\rho H) - \frac{k_B}{\beta} \text{S}(\rho) \), with the von Neumann entropy \( \text{S}(\rho) := -\text{Tr}(\rho \log_2 \rho) \). However, this statement cannot be generalized to the fully quantum regime where \( \rho' \) is asymmetric but \( \rho \) may be symmetric, because of the no-broadcasting theorem of asymmetry [23].

Our results show that the restriction of no-broadcasting can be lifted if the initial state has a small amount of asymmetry. Therefore, the following conjecture may hold. For a given pair of two quantum states \( \rho \) and \( \rho' \), there exist a finite-dimensional system in state \( \sigma \) and a global thermal operation \( \mathcal{E}_{\text{TO}} \) such that

\[
\mathcal{E}_{\text{TO}}(\rho_1 \otimes \sigma) = \rho'_1 \otimes \sigma,
\]

with \( \rho_1 \) arbitrarily close to \( \rho \), if and only if \( F(\rho) \geq F(\rho') \). We will leave further discussions on this conjecture to future work.

**Application to the quantum clock.**—After a system is prepared in an asymmetric state \( \rho \), it evolves according to its Hamiltonian \( H \) as \( \rho(t) = e^{-iHt} \rho e^{iHt} \). From the asymmetric condition \( [\rho, H] \neq 0 \), the states \( \rho(t) \) are not all the same and thus contain some time information. In this sense, the evolution of an asymmetric state is considered as the pointer of a quantum clock [26]. In general, a quantum clock is identified by the pair \( (\rho, H) \), and its accuracy is quantified by the Fisher timing information \( F(\rho, H) := \text{Tr}(\dot{\rho} \Delta^{-1} \rho) \), where \( \dot{\rho} := i[\rho, H] \) and \( \Delta B := \frac{1}{2}(\rho B + B \rho) \).

Previous result has shown that [23], it is impossible to distribute the timing information of a quantum clock into a system with zero timing information, without affecting the quantum state of the clock. Formally, let \( (\rho_1, H_1) \) be a finite-dimensional quantum clock \( S_1 \), and the pair \( (\rho_2, H_2) \) with \( [\rho_2, H_2] = 0 \) denote a system \( S_2 \) without timing information. Then the no-broadcasting theorem of asymmetry implies that there does not exist a global covariant operation \( \mathcal{E} \) such that \( \mathcal{E}(\rho_1 \otimes \rho_2) = \rho_1' \otimes \rho_2' \) and \( [\rho_2', H_2] \neq 0 \).

In some realistic circumstances, completely dephasing operations are difficult to implement exactly [27, 28]. Therefore, when initializing the second system \( S_2 \) to satisfy \( [\rho_2, H_2] = 0 \), one might obtain a pair \( (\rho_2', H_2) \) with arbitrarily small but positive Fisher timing information \( F(\rho_2, H_2) = \epsilon > 0 \). Theorem 1 indicates that, if \( \rho_1 \) is pure, the timing information in \( S_2 \) is still negligible after the action of a global covariant operation that preserves \( \rho_1 \), and thus generalizes the no-distributing principle of timing information to this noisy case.

Nevertheless, if \( \rho_1 \) is mixed, then it is possible to make the clock \( S_2 \) more accurate without affecting the state of the clock \( S_1 \). Further, the obtainable maximum accuracy of \( S_2 \) depends on the dimension of \( S_1 \). Still, it should be noticed that, the design of \( S_1 \) depends heavily on the initial state \( \rho_2 \). Therefore, in order to deterministically amplify the accuracy of \( S_2 \), one must know the exact form of \( \rho_2 \).

**Conclusion and discussion.**—We have investigated the ability of correlated catalyst in amplifying the asymmetry of a quantum system. While a catalyst in a pure state cannot extend the set of accessible states from any input state, a large enough catalyst in a mixed state can enable the conversion from a qubit state, which is arbitrarily close to (but not in) the set of symmetric states, to a state arbitrarily close to \( \rho^+ \), which is the qubit state with maximum asymmetry. The asymmetry in the initial state is essential due to the no-broadcasting theorem [22, 23]. Besides, in the limit of infinite-dimensional catalysis, \( \rho^+ \) can be reached as in catalytic coherence [24]. Hence, our result bridges these two important results concerning the restrictions on the coherence dynamics under translationally invariant operations.

It is also of interest to study the amplification of asymmetry in higher dimensional systems. The main difficulty in solving this problem is that it is numerically hard to calculate the cones of a high-dimensional state. An alternative way in dealing with this problem is to calculate the amount of asymmetry amplified by correlated catalysts, but such results would depend on the choice of asymmetry measure. A more meaningful question to ask is: can we generalize Theorem 2 to any finite-dimensional system? We conjecture that the answer is “yes”. One clue is to use the elementary framework as in Ref. [29], i.e., to operate on two energy levels at a time. This may be feasible, because the bounds on coherence dynamics under TIOs [30, 31] does not eliminate the possibility of engineering the coherence between two energy levels while preserving the coherence between other energy levels.

We have seen that, in resource theories of athermality [16] and asymmetry, correlated catalyst are strictly more powerful
than the uncorrelated ones. A related open problem is as follows. Is there any resource theory, in which the creation of correlations between the catalyst and the system can never extend the set of accessible states? A sufficient condition is that for any given bipartite state (whose form is known) of two resourceful systems, one can decouple the two systems while preserve their reduced states by free operations. This free-decoupling condition is potentially an interesting problem on its own, and to our knowledge, has been discussed only in the resource theory of athermality [32]. If a resource theory satisfies the free-decoupling condition, then any resource monotone are super-additive. However, the converse is not obvious: a variety of coherence monotones are proved to be superadditive [33–36], but it is not straightforward to prove that the resource theory of coherence satisfies the free-decoupling condition.

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APPENDICES

A. Proof of Theorem 1

Here we first prove a series of lemmas and then present the proof of Theorem 1.

**Lemma 1.** (Neother’s theorem [21]). For two pure states $|\psi\rangle$ and $|\psi'\rangle$, there exist a covariant unitary $V$ such that $V|\psi\rangle = |\psi'\rangle$ if and only if

$$
\langle\psi|e^{-iHt}\psi\rangle = \langle\psi'|e^{-iHt}\psi'|\rangle, \forall t.
$$

**Lemma 2.** (Marvian and Spekkens [21]). For two pure states $|\psi\rangle, |\psi'\rangle \in \mathcal{H}_S$ and a pure catalytic state $|\phi\rangle \in \mathcal{H}_C$, if a global covariant unitary $U$ induces the transformation

$$
U(|\psi\rangle \otimes |\phi\rangle) = |\psi'\rangle \otimes |\phi\rangle,
$$

then there exist a covariant unitary $V$ acting on $\mathcal{H}_S$ such that $V|\psi\rangle = |\psi'\rangle$.

**Proof.** From Lemma 1 and Eq. (13), we have $\langle\psi|e^{-iH_{ct}t}\psi\rangle\langle\phi|e^{-iH_{ct}t}\phi\rangle = \langle\psi'|e^{-iH_{ct}t}\psi'|\rangle\langle\phi|e^{-iH_{ct}t}\phi\rangle, \forall t$. Because $\langle\phi|e^{-iH_{ct}t}\phi\rangle \neq 0$, it follows that $\langle\psi|e^{-iH_{ct}t}\psi\rangle = \langle\psi'|e^{-iH_{ct}t}\psi'|\rangle, \forall t$. Then from Lemma 1, $|\psi\rangle$ can be transformed to $|\psi'\rangle$ via a covariant unitary. \hfill \Box

**Lemma 3.** Let $|\tilde{a}_0\rangle, \ldots, |\tilde{a}_{n-1}\rangle, |\tilde{b}\rangle$ be state vectors in Hilbert space $\mathcal{H}$ which need not be normalized. If for a given unitary $V_1$ the following equations hold

$$
\langle\tilde{a}_j|\tilde{b}\rangle = \langle\tilde{a}_j|V_1|\tilde{b}\rangle, \forall j,
$$

then there exist a unitary $V$ such that $V|\tilde{a}_j\rangle = |\tilde{a}_j\rangle$, $\forall j$ and $V|\tilde{b}\rangle = V_1|\tilde{b}\rangle$.

**Proof.** Here we first consider two trivial cases.

Case 1. $|\tilde{b}\rangle = 0$. In this case we simply set $V = 1$.

Case 2. $|\tilde{a}_j\rangle = 0, \forall j$. In this case, one can set $V = V_1$.

In the situation where the above two cases are excluded, let $\{|a_0\rangle, \ldots, |a_{d-1}\rangle\}$ be an orthonormal basis for the subspace $\mathcal{A} \equiv \text{span}\{|\tilde{a}_0\rangle, \ldots, |\tilde{a}_{n-1}\rangle\}$, and $\mathcal{A}_\perp \subset \mathcal{H}$ be the subspace orthogonal to $\mathcal{A}$. Then $|\tilde{b}\rangle$ can be written as

$$
|\tilde{b}\rangle = \sum_{k=0}^{d-1} b_k |a_k\rangle + b_d |b_\perp\rangle,
$$

where $|b_\perp\rangle \in \mathcal{A}_\perp$. From Eq. (14), for any state $|a\rangle \in \mathcal{A}$, it holds that $\langle a|V_1|\tilde{b}\rangle = \langle a|\tilde{b}\rangle$, and hence, $\langle a_k|V_1|\tilde{b}\rangle = \langle a_k|\tilde{b}\rangle = b_k$, for $k = 0, \ldots, d - 1$. Therefore,

$$
V_1|\tilde{b}\rangle = \sum_{k=0}^{d-1} b_k |a_k\rangle + b_d |b'_\perp\rangle,
$$

where $|b'_\perp\rangle \in \mathcal{A}_\perp$ and $\langle b'_\perp|b'_\perp\rangle = \langle b_\perp|b_\perp\rangle$. It follows that there exist a unitary $V_1$ acting on $\mathcal{A}_\perp$ such that $V_1|b_\perp\rangle = |b'_\perp\rangle$. Now we set $V = \Pi_\mathcal{A} \oplus V_1$, where $\Pi_\mathcal{A}$ is the projection to subspace $\mathcal{A}$. \hfill \Box
Lemma 4. Let $|\phi\rangle$ be a state in $\mathcal{H}_C$ and $\{|\psi_j\rangle\}_{j=0}^{k-1}$ be a set of states in $\mathcal{H}_S$. If a covariant unitary $U$ acting on $\mathcal{H}_S \otimes \mathcal{H}_C$ induces the following state conversion

$$U(|\psi_j\rangle \otimes |\phi\rangle) = |\psi'_j\rangle \otimes |\phi\rangle, \quad j = 0, \ldots, k - 1,$$

then there is a covariant unitary $V$ acting on $\mathcal{H}_S$ such that $V|\psi_j\rangle = |\psi'_j\rangle$, $\forall j = 0, \ldots, k - 1$.

Proof. For $k = 1$, it is obvious from Lemma 2.

Now we assume it holds for $k = n$ ($n \leq \dim(H_S) - 1$) and prove that it holds for $k = n + 1$. From the assumption, a covariant $V_a$ exists such that $|\psi'_j\rangle = V_a|\psi_j\rangle$ for $0 \leq j \leq n - 1$. The condition as in Eq. (17) is then written as

$$U_a(|\psi_j\rangle \otimes |\phi\rangle) = |\psi_j\rangle \otimes |\phi\rangle, \quad j = 0, \ldots, n - 1,$$

$$U_a(|\psi_n\rangle \otimes |\phi\rangle) = |\psi''_n\rangle \otimes |\phi\rangle,$$

where $U_a = (V_a^\dagger \otimes 1_C)U$ is still a global covariant unitary, and $|\psi''_n\rangle = V_a^\dagger|\psi_n\rangle$. Now we define a state $|\psi\rangle = c_a|\psi_j\rangle + c_b|\psi_n\rangle$, where $c_a, c_b \neq 0$ and $|\psi_j\rangle$ is chosen arbitrarily from $\{\psi_j\}^{n-1}_{j=0}$. From Eqs. (18) and (19), we have

$$U_a(|\psi\rangle \otimes |\phi\rangle) = |\psi'\rangle \otimes |\phi\rangle,$$

where $|\psi'\rangle = c_a|\psi_j\rangle + c_b|\psi''_n\rangle$. From Lemma 2, Eqs. (19) and (20) imply that $|\psi''_n\rangle = V_b|\psi_n\rangle$ and $|\psi'\rangle = V_0|\psi\rangle$, respectively, where $V_b$ and $V_0$ are covariant unitary operations. Hence,

$$V_0(c_a|\psi_j\rangle + c_b|\psi_n\rangle) = c_a|\psi_j\rangle + c_bV_a|\psi_n\rangle.$$

Let $\mathcal{H}^{(i)}$ be the $i$th energy eigenspace of the system, and then $\mathcal{H}_S = \oplus_i \mathcal{H}^{(i)}$. One can write $|\psi_j\rangle = \sum_i |\tilde{a}^{(i)}_j\rangle$, $|\psi_n\rangle = \sum_i |\tilde{b}^{(i)}_j\rangle$ where $|\tilde{a}^{(i)}_j\rangle, |\tilde{b}^{(i)}_j\rangle \in \mathcal{H}^{(i)}$ are not necessarily normalized. Because $[V_b, H_S] = [V_0, H_S] = 0$, we have $V_b = \oplus_i V^{(i)}_b$ and $V_0 = \oplus_i V^{(i)}_0$, where $V^{(i)}_b$ and $V^{(i)}_0$ are unitary operators acting on $\mathcal{H}^{(i)}$. Eq. (21) is then rewritten as

$$V^{(i)}_0(c_a|\tilde{a}^{(i)}_j\rangle + c_b|\tilde{b}^{(i)}_j\rangle) = c_a|\tilde{a}^{(i)}_j\rangle + c_bV^{(i)}_b|\tilde{b}^{(i)}_j\rangle, \quad \forall i.$$

It means that the states $c_a|\tilde{a}^{(i)}_j\rangle + c_b|\tilde{b}^{(i)}_j\rangle$ and $c_a|\tilde{a}^{(i)}_j\rangle + c_b|\tilde{b}^{(i)}_j\rangle$ are unitarily equivalent, so their norms equal. By noticing that this argument holds for all coefficients $c_a$ and $c_b$, and for arbitrary choice of $|\psi_j\rangle$, we arrive at

$$\langle \tilde{a}^{(i)}_j | \tilde{b}^{(i)}_j \rangle = \langle \tilde{a}^{(i)}_j | V^{(i)}_b | \tilde{b}^{(i)}_j \rangle, \quad \forall i, j.$$

Then by Lemma 3, a covariant unitary $V_1$ exists such that $V_1|\psi_j\rangle = |\psi'_j\rangle = V_a^\dagger|\psi'_j\rangle$ for $j = 0, \ldots, n - 1$, and $V_1|\psi_n\rangle = |\psi''_n\rangle = V_a^\dagger|\psi''_n\rangle$. By setting $V = V_aV_1$, we find that $|\psi'_j\rangle = V|\psi_j\rangle$, for $j = 0, \ldots, n$, i.e., this lemma holds for $k = n + 1$. This completes the proof. \qed

Lemma 5. For any two states $\rho$ and $\rho'$, and a given pure catalytic state $|\phi\rangle$ of finite dimension, if there exist a covariant unitary $U$, which satisfies $[U, H_S + H_C] = 0$, such that

$$U(\rho \otimes |\phi\rangle\langle\phi|)U^\dagger = \rho' \otimes |\phi\rangle\langle\phi|,$$

then there exist a covariant unitary $V$ satisfying $[V, H_S] = 0$, such that $V\rho V^\dagger = \rho'$.

Proof. Given $\rho = \sum_j p_j |\psi_j\rangle\langle\psi_j|$, Eq. (24) is equivalent to

$$\sum_j p_j |\Psi_j\rangle\langle\Psi_j| = \rho' \otimes |\phi\rangle\langle\phi|,$$

where $|\Psi_j\rangle = U(|\psi_j\rangle \otimes |\phi\rangle)$. By taking partial trace on $S$, we have $\sum_j p_j \text{Tr}_S(|\Psi_j\rangle\langle\Psi_j|) = |\phi\rangle\langle\phi|$, and hence, $\text{Tr}_S(|\Psi_j\rangle\langle\Psi_j|) = |\phi\rangle\langle\phi|$ for each $j$. It means that each $|\Psi_j\rangle$ is a product state, i.e., $U(|\psi_j\rangle \otimes |\phi\rangle) = |\psi'_j\rangle \otimes |\phi\rangle$, $\forall j$. By Lemma 4, a covariant unitary $V$ exist such that $|\psi'_j\rangle = V|\psi_j\rangle$, $\forall j$. It is obvious that $\rho' = \sum_j p_j |\psi'_j\rangle\langle\psi'_j|$, so we have $\rho' = V\rho V^\dagger$. \qed

Now we are ready to present the proof of theorem 1.
Direct calculations lead to the following results

$$\rho' \otimes |\phi\rangle \langle \phi| = \text{Tr}_E[U(\rho \otimes |\phi\rangle \langle \phi| \otimes \tau_E)U^\dagger],$$  \hspace{1cm} (26)

where $[\tau_E, H_E] = 0$ and $[U, H_S + H_C + H_E] = 0$. By taking partial trace on $S$, we have $\text{Tr}_{SE}[U(\rho \otimes |\phi\rangle \langle \phi| \otimes \tau_E)U^\dagger] = |\phi\rangle \langle \phi|$, and hence $U(\rho \otimes |\phi\rangle \langle \phi| \otimes \tau_E)U^\dagger = \rho'_{SE} \otimes |\phi\rangle \langle \phi|$. By Lemma 5, a covariant unitary $V$ exist such that $V(\rho \otimes \tau_E)V^\dagger = \rho'_{SE}$. By taking partial trace on $E$, we have $\rho' = \text{Tr}_E(\rho'_{SE}) = \text{Tr}_E(V(\rho \otimes \tau_E)V^\dagger)$, which means that $\rho'$ can be prepared from $\rho$ via TIO.

B. Asymmetry monotones of qubit states

Here we briefly review several measures of asymmetry (which are monotonic under TIOs), and then compare the ordering of them for qubit states.

Consider a system with Hamiltonian $H$ and in state $\rho$. Generally, the measure of asymmetry is a function of both $\rho$ and $H$. In the regime we concerned here, the Hamiltonian $H$ is fixed. Hence in the following, we express the measures of asymmetry as functions of $\rho$.

The quantum Fisher information is defined as

$$I_F(\rho) := \text{Tr}(\dot{\rho} \Delta^{-1} \dot{\rho}),$$  \hspace{1cm} (27)

where $\Delta, B := (\rho B + B \rho)/2$ and $\dot{\rho} := i[\rho, H]$. It quantifies the accuracy of a quantum clock specified by the pair $(\rho, H)$. A related measure is the Wigner-Yanase skew information, which is defined as

$$I_{WY}(\rho) := -\frac{1}{2} \text{Tr}(|\rho|^2, H^2),$$  \hspace{1cm} (28)

It has been proved that both $I_F$ and $I_{WY}$ are additive on tensor products. Further, these two measures become equivalent for pure states, i.e., $I_{WY}(|\psi\rangle\langle\psi|) = \frac{1}{2}I_F(|\psi\rangle\langle\psi|) = |\langle\psi|H|\psi\rangle|^2 - (|\langle\psi|H|\psi\rangle|^2).

Notice that in the resource theory of asymmetry, the set of free states is convex. Hence, one can employ some general resource measures to quantify the amount of asymmetry, e.g., the robustness and the distance-based measure. The robustness of asymmetry [38] is defined as

$$R(\rho) := \inf_{\gamma \in D} \left\{ s : \frac{\rho + s\gamma}{1 + s} \in \mathcal{F} \right\},$$  \hspace{1cm} (29)

where $D$ is the set of all states of the system and $\mathcal{F} = \{ \rho : \rho = e^{-iHt} \rho e^{iHt} \}$ is the set of symmetric states. It is quantitatively related to the task of state discrimination. The distance-based measure is defined as the minimum distance from the state $\rho$ to the set of symmetric states $\mathcal{F}$. Formally, let $D(\cdot|\cdot)$ be a distance measure of states, and the distance-based measure of asymmetry is defined as $A_D(\rho) := \min_{\gamma \in F} D(\rho||\gamma)$. When the distance measure $D(\rho||\gamma)$ is chosen to be the relative entropy $S(\rho||\gamma) := \text{Tr}(\rho \log_2 \rho - \rho \log_2 \gamma)$, the corresponding asymmetry measure is called the relative entropy of asymmetry. It has been shown that the relative entropy of asymmetry can be expressed as

$$A_r(\rho) = S[\Pi(\rho)] - S(\rho),$$  \hspace{1cm} (30)

where $S(\rho) := -\text{Tr}(\rho \log_2 \rho)$ is the von Neumann entropy, and $\Pi(\rho) \equiv \sum_j \Pi_j \rho \Pi_j$ with $\Pi_j$ the projection to the $j$-th eigenspace of $H$.

When the system under consideration is a qubit with fixed Hamiltonian $H = \frac{\Delta}{2} \sigma^z$, its asymmetry is relevant to the quantum coherence between the two eigenstates of $H$. In general, a qubit state can be expressed in the Bloch representation as $\rho = \frac{1}{2} \mathbf{1} + r \cdot \mathbf{\sigma}$, where $r \in [0, 1]$, $\mathbf{\sigma} = (\sigma^x, \sigma^y, \sigma^z)$ and $r = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$ is a normalized 3-dimensional real vector. Direct calculations lead to the following results

$$R(\rho) = r \sin \theta,$$  \hspace{1cm} (31)

$$I_F(\rho) = \Delta^2 (r \sin \theta)^2 = \Delta^2 [R(\rho)]^2,$$  \hspace{1cm} (32)

$$I_{WY}(\rho) = \frac{\Delta^2}{4} (1 - \sqrt{1 - r^2}) \sin^2 \theta = \frac{\Delta^2}{4} [R(\rho)]^2,$$  \hspace{1cm} (33)

$$A_r(\rho) = h(r \cos \theta) - h(r),$$  \hspace{1cm} (34)

Proof. From Proposition 2 of Ref. [37], every TIO can be implemented by coupling the system to an ancilla $E$ prepared in a symmetric state via a covariant unitary, so we have

$$\rho' \otimes |\phi\rangle \langle \phi| = \text{Tr}_E[U(\rho \otimes |\phi\rangle \langle \phi| \otimes \tau_E)U^\dagger],$$  \hspace{1cm} (26)
where the function \( h(x) \equiv -\frac{1+x}{2} \log_2 \frac{1+x}{2} - \frac{1-x}{2} \log_2 \frac{1-x}{2} \). From these results, we have the following observations.

**Observation 1.** Orderings of states. Let \( \rho_1 \) and \( \rho_2 \) be two qubit states. Then \( I_F(\rho_1) \geq I_F(\rho_2) \) is equivalent to \( R(\rho_1) \geq R(\rho_2) \). However, it is possible that \( I_{WY}(\rho_1) < I_{WY}(\rho_2) \) and/or \( A_r(\rho_1) < A_r(\rho_2) \). It means that, for qubit states with fixed Hamiltonian, the quantum Fisher information \( I_F \) and the robustness \( R \) give the same ordering of states, while the Wigner-Yanase skew information \( I_{WY} \) and the relative entropy of asymmetry \( A_r \) give other orderings of states. Therefore, the monotonicity of any measure is a necessary but not sufficient condition for state transformations under TIOs.

**Observation 2.** The maximally asymmetric states. All of the measures discussed above reach maximum for the set of states \( \{ \rho | \rho = \frac{1}{2} (I + \cos \phi \sigma^x + \sin \phi \sigma^y), \phi \in [0, 2\pi) \} \), which we call the maximally asymmetric states. Notice that each maximally asymmetric state can be obtained from the state \( \rho^+ = \frac{1}{2} (I + \sigma^x) \) by a covariant unitary operation. Also notice that the TIO cone of \( \rho^+ \) does not include all the qubit states.

### C. Modes of coherence and a general form of TIO operations

Consider a system with Hamiltonian \( H = \sum_j E_j |j\rangle \langle j| \). For a quantum state \( \rho \) expanded in its energy eigenbasis \( \rho = \sum_{i,j} \rho_{ij} |i\rangle \langle j| \), a mode of coherence [30] is defined as [39]

\[
\rho^{(\delta)} := \sum_{i,j; E_i-E_j=\delta} \rho_{ij} |i\rangle \langle j|.
\]

Here we define matrices

\[
P^{(\delta)} := \sum_{i,j; E_i-E_j=\delta} |i\rangle \langle j|,
\]

and then the modes of coherence can be written as

\[
\rho^{(\delta)} = \rho \odot P^{(\delta)},
\]

where the label \( \odot \) denotes the Hadamard product, i.e., the entrywise matrix product.

Let \( \mathcal{U}_t := e^{-iHt} \cdot e^{iHt} \) denote the free evolution of the system under its Hamiltonian \( H \), and it is directly check that

\[
\mathcal{U}_t(\rho^{(\delta)}) = e^{-i\delta t} \rho^{(\delta)},
\]

and then,

\[
\mathcal{U}_t(\rho) = \sum_{\delta} \mathcal{U}_t(\rho^{(\delta)}) = \sum_{\delta} e^{-i\delta t} \rho^{(\delta)} = \sum_{\delta} e^{-i\delta t} P^{(\delta)} \odot \rho = T_t \odot \rho,
\]

where \( T_t = \sum_{\delta} e^{-i\delta t} P^{(\delta)} \).

A TIO operation \( \mathcal{E} \) satisfies \( \mathcal{E} \circ \mathcal{U}_t = \mathcal{U}_t \circ \mathcal{E} \), which is equivalent to

\[
\sum_{\delta} e^{-i\delta t} \mathcal{E}(P^{(\delta)} \odot \rho) = \sum_{\delta} e^{-i\delta t} P^{(\delta)} \odot \mathcal{E}(\rho), \forall \rho, t.
\]

Then we have

\[
\mathcal{E} \left( P^{(\delta)} \odot \rho \right) = P^{(\delta)} \odot \mathcal{E}(\rho), \forall \rho.
\]

It means that, by a TIO operation, each mode in the initial state is independently mapped to the corresponding mode of the final state.

The Choi-Jamiolkowski matrix of operation \( \mathcal{E} \) is defined as

\[
J_{\mathcal{E}} = \begin{pmatrix}
\mathcal{E}(|0\rangle\langle 0|) & \mathcal{E}(|0\rangle\langle 1|) & \cdots \\
\mathcal{E}(|1\rangle\langle 0|) & \mathcal{E}(|1\rangle\langle 1|) & \cdots \\
\vdots & \vdots & \ddots
\end{pmatrix}.
\]


When $\mathcal{E}$ is a TIO, then we have
\[
\mathcal{E}(\langle i | j \rangle) = \mathcal{E}(\mathcal{P}(\delta_{ij}) \otimes | i \rangle \langle j |) = \mathcal{P}(\delta_{ij}) \otimes \mathcal{E}(\langle i | j \rangle),
\]
(43)
where $\delta_{ij} = E_i - E_j$. Here the first equation is from the definition of $\mathcal{P}(\delta)$, and the second equation is from Eq. (41). Now we define a matrix
\[
P := \begin{pmatrix}
P(\delta_{00}) & P(\delta_{01}) & \cdots \\
P(\delta_{10}) & P(\delta_{11}) & \cdots \\
\vdots & \vdots & \ddots
\end{pmatrix},
\]
(44)
Then the Choi-Jamiokowski matrix of a TIO operation $\mathcal{E}$ satisfies
\[
J_\mathcal{E} = J_\mathcal{E} \otimes P.
\]
(45)
This is the general form of a TIO operation.

As an example, we consider a qubit system with Hamiltonian $H = \frac{\Delta}{2} \sigma^z$. The matrix $P$ defined in Eq. (44) reads
\[
P := \begin{pmatrix}
p(0) & p(\Delta) \\
p(-\Delta) & p(0)
\end{pmatrix}
\]
(46)
with $p(0) = 1$ and $p(\pm \Delta) = \frac{1}{2}(\sigma^x \pm i \sigma^y)$. Then from Eq. (45), the Choi-Jamiokowski matrix of a qubit TIO is generally written as
\[
J_\mathcal{E} = \begin{pmatrix}
p_0 & 0 & 0 & \gamma \\
0 & 1-p_0 & 0 & 0 \\
0 & 0 & 1-p_1 & 0 \\
\gamma^* & 0 & 0 & p_1
\end{pmatrix},
\]
(47)
where the parameters satisfy $p_0, p_1 \in [0, 1]$ and $|\gamma| \leq \sqrt{p_0 p_1}$, such that $J_\mathcal{E}$ is positive.

D. TIO cone and CCTIO cone of a qubit state

In the Bloch presentation, a qubit state is generally written as $\rho(\vec{r}) = \frac{1}{2}(1 + \vec{r} \cdot \vec{\sigma})$, where $\vec{r}$ is a 3-dimensional real vector with $|\vec{r}| \leq 1$ and $\vec{\sigma} = (\sigma^x, \sigma^y, \sigma^z)$. The basic structure of its TIO cone or CCTIO cone is that it is rotationally symmetric about the $z$ axis. This is because any set of states which are rotationally symmetric about the $z$ axis are equivalent by covariant unitary operators $U(\phi) = \text{diag}(1, e^{i\phi})$.

Let the Bloch vector $\vec{r} = (\eta \cos \phi, \eta \sin \phi, z)$ with $\eta \in [0, 1], \phi \in [0, 2\pi), z \in [-1, 1]$. The TIO cone of $\rho(\vec{r})$ is written as
\[
C_{\text{TIO}}[\rho(\vec{r})] = \left\{ \rho' \mid \rho' = \frac{1}{2}(1 + \eta' \cos \phi' \sigma^x + \eta' \sin \phi' \sigma^y + z' \sigma^z), \right. \\
0 \leq \eta' \leq \min \left\{ \eta \sqrt{\frac{1+z'}{1+z}}, \eta \sqrt{\frac{1-z'}{1-z}} \right\}, \\
z' \in [-1, 1], \phi' \in [0, 2\pi) \right\}.
\]
(48)
The reason is as follows. After the action of a TIO in the form of Eq. (47), the qubit state $\rho(\vec{r})$ becomes
\[
\rho' = \frac{1}{2} \begin{pmatrix}
1 + z' & \eta' e^{-i\phi'} \\
\eta' e^{i\phi'} & 1 - z'
\end{pmatrix}
\]
(49)
with
\[
z' = -p_1(1-z) + p_0(1+z) - z,
\]
(50)
\[
\eta' e^{i\phi'} = \gamma \eta e^{i\phi}.
\]
(51)
From Eq. (50) and \( p_0, p_1 \in [0, 1] \), we have
\[
p_1 = \frac{(1 + z)p_0 - (z + z')}{1 - z},
\]
\[
p_0 \in [0, 1] \cap \left[ \frac{z + z'}{1 + z}, \frac{1 + z'}{1 + z} \right].
\] (52)

From Eq. (51) and \( |\gamma| \leq \sqrt{p_0p_1} \), we have
\[
\eta' = |\gamma| \eta \leq \eta \sqrt{p_0p_1}
\]
\[
\leq \eta \cdot \min \left\{ \frac{1 + z'}{1 + z}, \frac{1 - z'}{1 - z} \right\}.
\] (53)

Here the last inequality is from Eq. (52). The extreme states, for which the above equality holds, are obtained with \( |\gamma| = \sqrt{p_0p_1} \), \( p_1 \) as in Eq. (52), and
\[
p_0 = \min \left\{ 1, \frac{1 + z'}{1 + z} \right\}.
\] (54)

From the rotational symmetry and the convexity of TIO cone \( C_{\text{TIO}}[\rho(\vec{r})] \), we arrive at Eq. (48). This completes the proof.

Next, we prove the following statement. For state \( \rho(\eta) = \frac{1}{2} (1 + \eta \sigma^z) \), the CCTIO cone \( C_{\text{CCTIO}}^{(d)} \) is symmetric about the \( xy \) plane. The reason is as follows. Suppose \( \rho_\uparrow = \frac{1}{2} (1 + r_+ \sigma_x + r_- \sigma_z) \in C_{\text{CCTIO}}^{(d)}[\rho(\eta)] \), namely, a covariant operation \( \mathcal{E}_\uparrow \) and a \( d \)-dimensional catalyst in state \( \sigma_\uparrow \) exist such that \( \mathcal{E}_\uparrow(\rho(\eta) \otimes \sigma_\uparrow) = \rho_\uparrow \otimes \sigma_\uparrow \). Let \( \sigma_\downarrow = U^x \sigma_\uparrow U^x \) and \( \mathcal{E}_\downarrow = U_x \circ \mathcal{E}_0 \circ U_x \) with \( U_x(\cdot) = \sigma^+ \otimes U^x(\cdot) \sigma^x \otimes U^x \). Here the unitary operator \( U^x \) reverses the energy levels of the catalyst \( C \), i.e., it is anti-diagonal on the eigenbasis of \( H_C \) of each non-zero entries equal to 1. It is directly checked that \( \mathcal{E}_\downarrow \in \text{TIO} \) and \( \mathcal{E}_\downarrow(\rho(\eta) \otimes \sigma_\downarrow) = \rho_\downarrow \otimes \sigma_\downarrow \) with \( \rho_\downarrow = \frac{1}{2} (1 + r_+ \sigma_x - r_- \sigma_z) \), i.e., \( \rho_\downarrow \in C_{\text{CCTIO}}^{(d)}[\rho(\eta)] \).

### E. Numerical method on evaluating CCTIO cone of qubit states

Here we set the input states and target states as \( \rho_S \) and \( \rho_S' \), respectively. In the Bloch representation, the extreme states in CCTIO cone are defined as those with maximum distance from the \( z \)-axis for given \( r'_z = \text{tr}(\sigma^z \rho_S') \). Since any state in the cone can be achieved by applying a dephasing map (which is a TIO) on an extreme a extreme state, it is sufficient to solve the extreme states to obtain the whole cone. Because the CCTIO cone of a qubit system is symmetric about \( z \)-axis, we only need to solve the extreme states within \( xz \) plane with \( x \geq 0 \). Our problem then becomes the following optimization task
\[
R_{\text{CC}}^{(d)}(\rho_S, r'_z) = \max_{\sigma_C \in D(H_c^{(d)})} R_{\text{CC}}(\sigma_C; \rho_S, r'_z),
\] (55)

where \( D(H_c^{(d)}) \) is the set of \( d \)-dimensional density matrices, and the function \( R_{\text{CC}}(\sigma_C; \rho_S, r'_z) \) is defined as
\[
R_{\text{CC}}(\sigma_C; \rho_S, r'_z) = \max_{\mathcal{E} \in \text{TIO}} \text{Tr}[\sigma^\sigma \rho_S'],
\]
\[
\text{s.t.} \quad \rho_S' = \text{Tr}_C [\mathcal{E}(\rho_S \otimes \sigma_C)],
\]
\[
\sigma_C \equiv \text{Tr}_S [\mathcal{E}(\rho_S \otimes \sigma_C)],
\]
\[
r'_z = \text{Tr}[\sigma^z \rho_S'] .
\] (56)

Clearly, the function \( R_{\text{CC}}(\rho_S, r'_z) \) embeds a lower-level optimization Eq. (56) into a upper-level optimization Eq. (55). Such optimization task is called bi-level optimization [40], and is generally hard to be solved. Here we first consider the lower level of optimization, and then describe the methods in solving Eq. (55) for \( d = 2, 3 \).

The lower-level optimization as in Eq. (56) can be reformulated as a semi-definite programming (SDP) task, which allows us effectively solve it in polynomial time via interior point methods [41]. Here, we derive the explicit SDP form in the following. Let \( H_S = \frac{\Delta}{2} \sigma^z \) be the Hamiltonian of \( S \) and \( H_C = \sum_{l=0}^{d-1} |\Delta| l \) be the Hamiltonian of \( C \). The eigenbasis of the total Hamiltonian \( H_{SC} = H_S + H_C \) is labeled as \( \{|\psi_j\rangle\}_{j=0}^{2d-1} \), and the eigenvalue of each \( |\psi_j\rangle \) is denoted as \( E_j \). Thus, the explicit form of matrix \( P \) in Eq. (45) is written as
\[
P = \sum_{jk} |\psi_j\rangle \langle \psi_k| \otimes P^{(mjk)}.
\] (57)
where $P^{(m_j \Delta)}$ is defined in Eq. (36) with $m_j \Delta = E_j - E_k$. Notice that $m_j$ are integers and satisfy $-d \leq m_j \leq d$. Then, by setting the optimization variable as the Choi-Jamiolkowski matrix $J_E$ of TIO, we arrive at the SDP form of Eq. (56) as

$$\max_{J_E} \quad \text{Tr} [\sigma^2 \rho_S^\star]$$

s.t. 
$$J_E \geq 0, \quad (\text{CP condition})$$
$$\text{Tr}_{SC'} [J_E] = \mathbb{1}_{SC}, \quad (\text{TP condition})$$
$$J_E \otimes P = J_E, \quad (\text{TIO condition})$$

$$\rho'_{SC} := \text{Tr}_{SC} \left[ (\rho_S \otimes \sigma_C \otimes \mathbb{1}_{SC'})^T \cdot J_E \right],$$

$$\sigma_C = \text{Tr}_{SC'} [\rho'_{SC}], \quad (\text{CC condition})$$

$$\rho'_S = \text{Tr}_{C'} [\rho'_{SC}], \quad r'_z = \text{Tr}(\sigma^2 \rho'_S)$$

where CC condition denotes the correlated catalyst condition, $S'C'$ is the output space of $SC$ and the total target states $\rho_{SC}'$ follows the definition of Choi-Jamiolkowski matrix. In practice, we use the CVX package [42] to numerically solve this SDP task with tolerance at $1.81 \times 10^{-12}$.

For the upper part of optimization, the property of $R_{CC}(\sigma_C; \rho_S, r'_z)$ is essential. In the following, we prove the continuity of $R_{CC}(\sigma_C; \rho_S, r'_z)$ on $\sigma_C$, which will help us to find the optimizer of $R_{CC}(\rho_S, r'_z)$ over $D(H_C)$.  

**Lemma 6** ($R_{CC}(\sigma_C; \rho_S, r'_z)$ has Lipschitz continuity on $\sigma_C$). For any pair of catalytic states $\sigma_C$ and $\sigma'_C$ satisfying $||\sigma_C - \sigma'_C||_1 \leq \epsilon$, we have

$$|R_{CC}(\sigma'_C; \rho_S, r'_z) - R_{CC}(\sigma_C; \rho_S, r'_z)| \leq 4\epsilon(1 + \epsilon).$$

(59)

*Proof.* For convenience, we set

$$R_{CC}(\sigma'_C; \rho_S, r'_z) \geq R_{CC}(\sigma_C; \rho_S, r'_z).$$

(60)

One optimizer of $R_{CC}(\sigma'_C; \rho_S, r'_z)$ is denoted by $E^*$, and the corresponding output state of $S$ is $\rho'_S$.

The proof is sketched as follows. First, we construct a trace preserving (TP) map $N$, which is close to $E^*$, and satisfies

$$N(\rho_S \otimes \sigma_C) = \rho'_S|\sigma_C.$$

(61)

Second, we slightly extend the set of free operation which lead to an upper bound of $R_{CC}(\sigma'_C; \rho_S, r'_z)$. Finally, we prove that the difference between $R_{CC}(\sigma_C; \rho_S, r'_z)$ and the upper bound of $R_{CC}(\sigma'_C; \rho_S, r'_z)$ is no larger than $4\epsilon(1 + \epsilon)$.

Let us construct a trace preserving map (which need not to be completely positive) as

$$N = \mathcal{I} \otimes N_1 \circ E^* \circ \mathcal{I} \otimes N_0.$$

Here $\mathcal{I}$ denotes the identity map, $N_0 := \mathcal{I} + M_0$ and $N_1 := \mathcal{I} + M_1$, where $M_0$ and $M_1$ are constant maps defined as

$$M_0(\cdot) = \sigma'_C - \sigma_C, \quad M_1(\cdot) = \sigma_C - \sigma'_C.$$

(63)

It is easy to check that $N$ satisfies Eq. (61), and that

$$||\mathcal{I} - N_0||_\diamond \leq \epsilon, \quad ||\mathcal{I} - N_1||_\diamond \leq \epsilon,$$

(64)

where $||\cdot||_\diamond$ is the diamond norm [43]. Then, we examine the distance between $N$ and $E^*$ as

$$||N - E^*||_\diamond \leq ||\mathcal{I} \otimes M_1 \circ E^*||_\diamond + ||E^* \circ \mathcal{I} \otimes M_0||_\diamond + ||\mathcal{I} \otimes M_1 \circ E^* \circ \mathcal{I} \otimes M_0||_\diamond \leq 2\epsilon(1 + \epsilon).$$

(65)

Now we define the set of the allowed bipartite operations for given $\rho_S$, $\sigma_C$, and $r'_z$ as

$$O_{(\sigma_C; \rho_S, r'_z)} = \{ E | \text{Tr}_S [E(\rho_S \otimes \sigma_C)] = \sigma_C, \quad \text{Tr} [\sigma^2 \text{Tr}_C (E(\rho_S \otimes \sigma_C))] = r'_z, \quad E \in \text{TIO} \}.$$

(66)

Then Eq. (65) implies that the TP map $N$ is $2\epsilon(1 + \epsilon)$-close to the set $O_{(\sigma_C; \rho_S, r'_z)}$. 

Next, we define a set of TP maps as
\[ O_{\{\sigma C; \rho S, r'_z\}} = \{ \mathcal{E}' | \inf_{\mathcal{E} \in O_{\{\sigma C; \rho S, r'_z\}}} \| \mathcal{E} - \mathcal{E}' \|_0 \leq \epsilon, \mathcal{E}' \in \mathcal{TP}, \text{Tr}_S [\mathcal{E}' (\rho_S \otimes \sigma_C)] = \sigma_C, \text{Tr} [\sigma^z \text{Tr}_C (\mathcal{E}' (\rho_S \otimes \sigma_C))] = r'_z \}, \] (67)

and a function as
\[ R_{CC}^2(\sigma_C; \rho_S, r'_z) = \max_{\mathcal{E}' \in O_{\{\sigma C; \rho S, r'_z\}}} \text{Tr} [\sigma^z \rho'_S], \]
\[ \text{s.t. } \rho'_S = \text{Tr}_C [\mathcal{E}' (\rho_S \otimes \sigma_C) \sigma_C], \rho'_S \text{ is positive semi-definite}, \]
\[ \sigma_C = \text{Tr}_S [\mathcal{E}' (\rho_S \otimes \sigma_C)], \]
\[ r'_z = \text{Tr}(\sigma^z \rho'_S). \] (68)

Because \( \mathcal{N} \in O_{\{\sigma C; \rho S, r'_z\}} \), we have
\[ R_{CC}(\sigma_C; \rho_S, r'_z) = \text{Tr} [\sigma^z \rho'_S] = \text{Tr} [\sigma^z \text{Tr}_C \mathcal{N} (\rho_S \otimes \sigma_C)] \]
\[ \leq R_{CC}^{2(1+\epsilon)}(\sigma_C; \rho_S, r'_z). \] (69)

Then we turn to the difference between \( R_{CC}(\sigma_C; \rho_S, r'_z) \) and \( R_{CC}^{2(1+\epsilon)}(\sigma_C; \rho_S, r'_z) \). From Eq. (67), for any state \( \rho'_S \) which can be obtained as \( \rho'_S = \text{Tr}_C [\mathcal{E}' (\rho_S \otimes \sigma_C)] \) with \( \mathcal{E}' \in O_{\{\sigma C; \rho S, r'_z\}} \), there exists an operation \( \mathcal{E} \in O_{\{\sigma C; \rho S, r'_z\}} \) such that \( \mathcal{E}(\rho_S \otimes \sigma_C) = \rho'_S | \sigma_C \) and
\[ \| \rho'_S - \rho''_S \|_1 \leq 2\epsilon(1 + \epsilon), \] (70)

It follows that
\[ | \text{Tr}(\sigma^z \rho'_S) - \text{Tr}(\sigma^z \rho''_S) | \leq 2\| \rho'_S - \rho''_S \|_1 \leq 4\epsilon(1 + \epsilon), \] (71)

and hence,
\[ R_{CC}^{2(1+\epsilon)}(\sigma_C; \rho_S, r'_z) \leq R_{CC}(\sigma_C; \rho_S, r'_z) + 4\epsilon(1 + \epsilon) \] (72)

Recalling Eq. (69), we arrive at
\[ R_{CC}(\sigma_C'; \sigma_S, r'_z) \leq R_{CC}(\sigma_C; \rho_S, r'_z) + 4\epsilon(1 + \epsilon). \] (73)

This completes the proof.

The continuity of \( R_{CC}(\sigma_C; \rho_S, r') \) allows us to numerically calculate \( R_{CC}^{(d)}(\rho_S, r'_z) \) (which is the upper bound of \( R_{CC}(\sigma_C^{(d)}; \rho_S, r'_z) \)) with small error, by uniform sampling of \( d \)-dimensional catalytic states. For \( d = 2 \), because states which are rotational symmetric about the \( z \) axis are equivalent under covariant unitaries, it is sufficient to sample within \( z \) plane with \( x \geq 0 \). When \( d > 2 \), due to the enormous sampling cost, we can only obtain the local optimal by gradient descent method, though this continuity property can accelerate the initial sampling process of this optimization task. The technical detail of solving \( c_{\text{CCTIO}}^{(d)} \) is shown in the following.

For solving \( c_{\text{CCTIO}}^{(2)} \), we uniformly sampling on the \( xz \) plane with \( x \geq 0 \), as we have mentioned before. In practice, we sample on 2-dimensional lattice with constant 1/256 (i.e. the size of lattice cell along both \( x \) and \( z \) axis is set to be 1/256). Thus, in a lattice cell, the maximal distance of unsampled points from the sampled point is \( 2\epsilon = 1/(256\sqrt{2}) \). Then, according to Eq. (59), we obtain the Lipschitz error of upper bound of \( c_{\text{CCTIO}}^{(2)} \) less than \( 4\epsilon(1 + \epsilon) \approx 5.553 \times 10^{-3} \).

For obtaining \( c_{\text{CCTIO}}^{(3)} \), we use the gradient descent optimization to find local maximal of \( R_{CC}(\sigma_C^{(3)}; \rho_S, r'_z) \). The initial points for gradient descent are chosen via LIPO algorithm [44], which allows us using Lipschitz condition to accelerate the searching process. Note that we use the HilbertSchmidt ensemble generating method [45] as the sub-task of LIPO to randomly sample the catalysts. Then we parameterize every 3-dimensional catalysts using eight Gell-Mann matrices [46], such that we can calculate the approximate gradient in \( \mathbb{R}^8 \). The termination tolerance of function is set at \( 1 \times 10^{-8} \). Notice that this strategy can be also applied to obtain \( c_{\text{CCTIO}}^{(d)} \) with \( d > 3 \).
F. A generalized protocol

In the main text, we propose a protocol to transform a qubit state $\rho$ satisfying $[\rho, H_S] \neq 0$ to a state arbitrarily close to $\rho^+$ via CCTIO. Nevertheless, because $C_{TIO}(\rho^+)$ does not include all the qubit states, it is not straightforward to see whether one can transform $\rho$ to any qubit state approximately via CCTIO. In the following, we give an affirmative answer to this question by generalizing the above protocol.

In the generalized protocol, we start from a state $\rho(\eta, z) = \frac{1}{2}(1 + \eta \sigma^x + z \sigma^z)$ with $\eta > 0$, and transform it to state

$$\rho(\eta', z') = \frac{1}{2}(1 + \eta' \sigma^x + z' \sigma^z)$$  \hspace{1cm} (74)

with $0 \leq \eta' < \sqrt{1 - z'^2}$ under CCTIO.

The setup of the generalized protocol is the same as the protocol in the main text. Here we make two changes. First, at the beginning of the protocol, we transform the input state $\rho(\eta, z)$ to $\rho(\eta_1, z')$ (instead of $\rho(\eta_1)$) with $\eta_1 > 0$ via TIO. The second change is that, the reduced state of each two-qubit particle $C_i$ in the catalyst is set to be

$$\sigma_i = \sigma(\eta_i, z')$$

$$\equiv \left(\frac{1}{2} + \frac{z'}{3}\right)\sigma^+(\eta_i, z') \otimes |\uparrow\rangle \langle \uparrow|$$

$$+ \left(\frac{1}{2} - \frac{z'}{3}\right)\sigma^+(\eta_i, z') \otimes |\downarrow\rangle \langle \downarrow|,$$  \hspace{1cm} (75)

where

$$\sigma^+(\eta_i, z') \equiv \frac{1}{2} 1 + \frac{\sqrt{3} \eta_i}{2(2 + z')} \sigma^x + \frac{z'(8 + 3z') + 4 - \eta_i^2}{2(2 + z')(3 + 2z')} \sigma^z,$$

$$\sigma^+(\eta_i, z') \equiv \frac{1}{2} 1 + \frac{\sqrt{3} \eta_i}{2(2 + z')} \sigma^x + \frac{z'(8 - 3z') - 4 + \eta_i^2}{2(2 - z')(3 - 2z')} \sigma^z,$$  \hspace{1cm} (76)

are states of qubit $C_{11}$, and $|\uparrow\rangle$ and $|\downarrow\rangle$ are energy eigenstates of qubit $C_{12}$. Here the parameter $z'$ is determined by the target state $\rho(\eta', z')$, and $\eta_i$ depend on both the initial state and the target state.

After the action of $E_i$, the state of the system qubit becomes $\rho(\eta_{i+1}, z')$ with

$$\eta_{i+1} = \eta_i + \frac{\eta_i(1 - z'^2 - \eta_i^2)}{6(4 - z'^2)},$$  \hspace{1cm} (77)

for $i = 1, \ldots, N$. It is directly checked that $\Delta \eta_i \equiv \eta_{i+1} - \eta_i > 0$ for $0 < \eta_i < \sqrt{1 - z'^2}$. Therefore, with finite $N$, we can achieve state $\rho(\eta_N, z')$ with $\eta' < \eta_N < \sqrt{1 - z'^2}$. Clearly, $\rho(\eta', z') \in C_{TIO}[\rho(\eta_N, z')]$. Hence by our protocol, any state as in Eq. (74) can be obtained via CCTIO from state $\rho(\eta, z)$ with $\eta > 0$.

Notice that in the Bloch representation, any mixed state in $xz$ plane with $x \geq 0$ can be presented in the form of Eq. (74). By the rotational symmetry of the CCTIO cone of a qubit state, we conclude that the CCTIO cone of an asymmetric state contains all the mixed qubit states.