


Fundamental Limits on Correlated Catalytic State Transformations

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Determining whether a given state can be transformed into a target state using free operations is one of the fundamental questions in the study of resource theories. Free operations in resource theories can be enhanced by allowing for a catalyst system that assists the transformation and is returned unchanged, but potentially correlated, with the target state. While this has been an active area of recent research, very little is known about the necessary properties of such catalysts. Here, we prove fundamental limits applicable to a large class of correlated catalytic transformations by showing that a small residual correlation between a catalyst and target state implies that the catalyst needs to be highly resourceful. In fact, the resource required diverge in the limit of vanishing residual correlation. In addition, we establish that in imperfect catalysis a small error generally implies a highly resourceful embezzling catalyst. We develop our results in a general resource theory framework and discuss its implications for the resource theory of athermality, the resource theory of coherence, and entanglement theory.

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Introduction.—A quantum resource theory is defined by a set of *free operations* [1] and a set of *free states* with the property that free operations are closed under composition and map free states into free states [2,3]. Resource theories offer a general and versatile framework to quantify the usefulness of different quantum states and their interconvertibility using free operations. Prominent examples of resource theories include entanglement theory [4–6] (where local operations and classical communication are free and entanglement is considered a resource), athermality in thermodynamics [7–9] (where transformations that preserve the thermal state are free and states out of thermal equilibrium are resourceful), and coherence [10–12] (where incoherent states are free and coherence is a resource).

In the following, we will use the notation $\rho \rightarrow \rho'$ to indicate that a free transformation exists which maps a quantum state ρ to a quantum state ρ' . Given a fixed state ρ , a fundamental question in any resource theory is to find the set of states $\mathcal{FO}(\rho)$ of all ρ' such that $\rho \rightarrow \rho'$, i.e., all states that can be reached from ρ using free operations. More precisely, we are often interested in its closure, $\overline{\mathcal{FO}(\rho)}$, which also contains quantum states that can be arbitrarily well approximated by free operations from ρ [13]. The set of free operations can be enlarged by allowing for *catalytic transformations*, $\rho \otimes \nu \rightarrow \rho' \otimes \nu$, where the catalyst ν is returned unchanged. The set $\mathcal{CO}(\rho)$ then contains all states ρ' for which such a catalytic transformation from ρ exists. Its closure is denoted $\overline{\mathcal{CO}(\rho)}$. More recently, a further

relaxation has been studied where correlations between the catalyst and the target state after the transformation are allowed and can be used as a resource in catalytic transformations [16–24]. We say that a state ρ' can be reached by a *correlated catalytic transformation*, or $\rho' \in \mathcal{CCO}(\rho)$, if there exists a catalyst ν such that $\rho \otimes \nu \rightarrow \tau$ where τ is any state that has marginals ρ' (for the target system) and ν (for the catalyst system). We will similarly be concerned with its closure, $\overline{\mathcal{CCO}(\rho)}$.

To the best of our knowledge, the idea of residual correlations between the system and the catalyst in the output state while the catalyst returns exactly to its original form was first introduced in Ref. [25]. In Ref. [26] the authors first discussed whether the free energy completely characterizes correlated catalytic transformations in resource theory of athermality. This question was answered positively in the classical case and conjectured for the quantum case in Ref. [19]. The conjecture for the quantum case has been recently resolved in the affirmative in Ref. [22] using the previously known construction that allows one to reduce the problem to asymptotic interconvertibility [27]. This was recently generalized for any resource theory in Ref. [24].

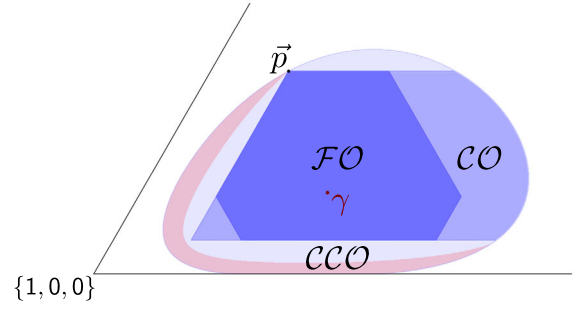
The sets \mathcal{FO} , \mathcal{CO} , and \mathcal{CCO} are generally difficult to characterize, but they take on a natural form for certain resource theories where they are fully characterized by *resource monotones*. Let \mathfrak{R} be a function from quantum states to positive reals that measures the resourcefulness of

states. We say that such a map is (i) a resource monotone if it is nonincreasing under free operations, (ii) *tensor-additive* if it is additive under tensor products, and (iii) *super-additive* if $\mathfrak{R}(\rho_{AB}) \geq \mathfrak{R}(\rho_A) + \mathfrak{R}(\rho_B)$ for any joint state ρ_{AB} with marginals ρ_A and ρ_B . Resource monotones play an important role in characterizing the above sets. It is easy to see that a necessary (but not generally sufficient) condition for ρ' to be in the set $\mathcal{FO}(\rho)$ is that $\mathfrak{R}(\rho) \geq \mathfrak{R}(\rho')$ for any resource monotone. For ρ' to be in $\mathcal{CO}(\rho)$ this ordering only needs to be required for tensor-additive resource monotones, and finally for ρ' to be in $\mathcal{CCO}(\rho)$ the ordering only needs to be satisfied for tensor-additive and super-additive resource monotones. Finally, for ρ' to be in the closure of the sets we require in addition the resource monotone to be lower semicontinuous (see Supplemental Material, Sec. IV, Lemma 6 [28], for a proof). In general, it is not known which resource monotones characterize these sets, i.e., what are the necessary and sufficient conditions for ρ' to be in any of the sets. The particular appeal of $\overline{\mathcal{CCO}(\rho)}$ is that for some prominent resource theories it is fully characterized by a single resource monotone, e.g., the nonequilibrium free energy [22] or the relative entropy of entanglement [23]. Moreover, the set $\overline{\mathcal{CCO}(\rho)}$ is also of operational interest since it contains states that are strictly more useful than $\mathcal{CO}(\rho)$ for some information-theoretic tasks, for example quantum teleportation [63]. Figure 1 gives an example of these sets and their full characterization for the resource theory of athermality restricted to states that commute with the Hamiltonian.

While allowing arbitrary correlations between the catalyst and target state arguably goes against the spirit of catalysis, recent works [19,22–24] showed that for some prominent reversible resource theories target states in $\rho' \in \overline{\mathcal{CCO}(\rho)}$ can be achieved with arbitrarily small correlations with the catalyst.

In this Letter, we investigate the fundamental limits of such correlated catalytic transformations. Our results apply to any catalytic transformation between a given pair of “hard-to-transform” states and are applicable to any resource theory in which certain monotones are tensor additive. We focus on the problem of preparing suitable catalysts, and we find that for some target states that lie in the set $\mathcal{CCO} \setminus \mathcal{CO}$, correlated catalytic transformations with small correlations require catalysts that are highly resourceful, and in fact, require unbounded resources in the limit of vanishing correlations. (See Fig. 1 for a depiction of such states.) In particular, we show a quantitative trade-off between the error ε achievable in the transformation and the resources needed for the catalyst.

Formal setting.—We denote by $\mathcal{S}(\mathcal{H})$ the set of quantum states on a d -dimensional Hilbert space \mathcal{H} . We introduce the purified distance [64], which for normalized states is defined as $P(\rho, \sigma) := \sqrt{1 - F(\rho, \sigma)}$, where



Classical resource theory of athermality	
$\frac{\mathcal{FO}}{\mathcal{FO}}$	$(p, \gamma) \succ (p', \gamma)$ [9]
\mathcal{CO}	$D_\alpha(p\ \gamma) > D_\alpha(p'\ \gamma)$ and $D_\alpha(\gamma\ p) > D_\alpha(\gamma\ p')$ for all $\alpha \geq 1/2$ and p' has full support [28]
$\overline{\mathcal{CO}}$	$D_\alpha(p\ \gamma) \geq D_\alpha(p'\ \gamma)$ and $D_\alpha(\gamma\ p) \geq D_\alpha(\gamma\ p')$ for all $\alpha \geq 1/2$ [28, 29]
\mathcal{CCO}	$D(p\ \gamma) > D(p'\ \gamma)$ and p' has full support [17, 18]
$\overline{\mathcal{CCO}}$	$D(p\ \gamma) \geq D(p'\ \gamma)$ [17]

FIG. 1. Example of the sets \mathcal{FO} , \mathcal{CO} and \mathcal{CCO} for classical resource theory of athermality with rational Gibbs states where we fixed the input state $\vec{p} = \{2/3, 1/12, 3/12\}$ and $\gamma = \{7/10, 2/10, 1/10\}$. We show one corner of the probability simplex (which is a triangle in this case). Each point in the triangle corresponds to a (classical) state of a three-dimensional system. The points in the red region satisfy the conditions of Theorem 2. The table contains the conditions characterizing each set, where D_α is the Rényi divergence of order α and D is the Kullback-Leibler divergence.

$F(\rho, \sigma) := (\text{Tr}[\sqrt{\sqrt{\rho}\sqrt{\sigma}}])^2$ is the Uhlmann fidelity. The Umegaki relative entropy is defined as $D(\rho\|\sigma) = \text{Tr}[\rho(\log \rho - \log \sigma)]$. Since both the fidelity and the relative entropy satisfy a data-processing inequality under quantum channels, we can define resource monotones

$$\mathfrak{D}(\rho) = \min_{\sigma \in \mathcal{F}} D(\rho\|\sigma) \quad \text{and} \quad \mathfrak{D}_{1/2}(\rho) := -\log \mathfrak{F}(\rho)$$

with $\mathfrak{F}(\rho) := \max_{\sigma \in \mathcal{F}} F(\rho\|\sigma)$. These are the limiting cases at $\alpha = 1$ and $\alpha = 1/2$, respectively, of a larger family of resource monotones, $\mathfrak{D}_\alpha(\rho) := \min_{\sigma \in \mathcal{F}} \tilde{D}_\alpha(\rho\|\sigma)$ where $\tilde{D}_\alpha(\rho\|\sigma)$ is the sandwiched Rényi divergence [65–67] and is defined for $\alpha \in [\frac{1}{2}, 1) \cup (1, \infty)$ as [65–67]

$$\tilde{D}_\alpha(\rho\|\sigma) := \frac{1}{\alpha - 1} \log \text{Tr}(\sigma^{\frac{1-\alpha}{2\alpha}} \rho \sigma^{\frac{1-\alpha}{2\alpha}})^\alpha.$$

We say that \mathfrak{D}_α is additive for the state ρ if $\mathfrak{D}_\alpha(\rho \otimes \nu) = \mathfrak{D}_\alpha(\rho) + \mathfrak{D}_\alpha(\nu)$ for any catalyst state ν .

We are now ready to define correlated catalytic transformations [22–24] as follows:

Definition 1. Let $\rho, \rho' \in \mathcal{S}(\mathcal{H})$ be a pair of quantum states and $\varepsilon > 0$ a small positive constant. We say that ρ can

be transformed into ρ' by an ε -correlated catalytic transformation if there exists a free operation \mathcal{N} and a catalyst state $\nu \in \mathcal{S}(\mathcal{H}')$ such that $\mathcal{N}(\rho \otimes \nu) = \tau$, $\text{Tr}_{\mathcal{H}}[\tau] = \nu$, and $P(\rho' \otimes \nu, \tau) \leq \varepsilon$. If this holds for any $\varepsilon > 0$ we say that ρ is transformable into ρ' by a correlated catalytic transformation.

For the specific resource theories we consider, the quantity \mathfrak{D} completely characterizes the set $\overline{\text{CCO}}$, namely the necessary and sufficient condition for ρ' to be in $\overline{\text{CCO}}(\rho)$ is that $\mathfrak{D}(\rho) \geq \mathfrak{D}(\rho')$ (see the discussion of the individual resource theories below). Motivated by this, we identify \mathfrak{D} as the relevant resource measure to evaluate the resourcefulness of the catalyst. We remark that the dimension of the Hilbert space of the catalyst, without adding any further constraints, does not quantify the resourcefulness of the catalyst. For example, in the resource theory of athermality, states with large free energy can be constructed easily in low dimension using a sufficiently gapped Hamiltonian.

Main result and discussion.—We are now ready to state our main theorem.

Theorem 1. Assume that $\rho, \rho' \in \mathcal{S}(\mathcal{H})$ and $\alpha \in [1/2, 1)$ such that \mathfrak{D}_α is additive for the state ρ' and $\mathfrak{D}_\alpha(\rho) < \mathfrak{D}_\alpha(\rho')$. Then, for any ε -correlated catalytic transformation with catalyst ν mapping ρ into ρ' , we have

$$\mathfrak{D}(\nu) = \Omega\left(\log \frac{1}{\varepsilon}\right).$$

In particular, when $\alpha = 1/2$ and, thus, $\mathfrak{F}(\rho) > \mathfrak{F}(\rho')$, we have the quantitative bound

$$\sqrt{\mathfrak{F}(\nu)} \leq \frac{\varepsilon}{\sqrt{\mathfrak{F}(\rho)} - \sqrt{\mathfrak{F}(\rho')}}.$$

We formulated the above theorem for any resource theories, but it is only meaningful when there exist a pair of states and α satisfying the assumptions. Quantitative bounds for $\alpha \neq 1/2$ can be found in the Supplemental Material, Sec. V [28]. The quantities \mathfrak{D}_α can be interpreted as a measure of distance between a state and the free set. In the following, we refer to the quantity $\sqrt{\mathfrak{F}(\rho)} - \sqrt{\mathfrak{F}(\rho')}$ as fidelity gap.

The condition $\mathfrak{D}_\alpha(\rho) < \mathfrak{D}_\alpha(\rho')$ for some $\alpha \in [1/2, 1)$, together with the additivity assumption, implies that the output state ρ' lies outside the set $\overline{\text{CCO}}(\rho)$ (see the Supplemental Material, Sec. IV, Lemma 6 [28] for a detailed discussion). Hence, catalytic transformation from ρ to ρ' is possible only by allowing correlations (see Fig. 1). For this reason, we say that the pair of states (ρ, ρ') is “hard to transform,” and we will establish the existence of such state pairs for the resource theories we consider.

For correlated catalysis, i.e., when there are nonzero residual correlations between the catalyst and the system in the output state, the theorem implies that, as the error

decreases, the distance between the catalyst and the free set must increase. In particular, in the limit of zero error, the catalyst state must be orthogonal to the set of free states, i.e., its resourcefulness is unbounded. As we discuss in the Supplemental Material, Sec. V [28], we can also derive bounds for the robustness of the catalyst.

We point out that the above theorem actually also holds if we lift the restriction $\text{Tr}_{\mathcal{H}}[\tau] = \nu$, and hence we do not need to exactly recover the catalyst after the transformation. If we allow a small error in the catalyst after the transformation, any state transformation is possible. This phenomenon is called *embezzling* [68–70]. Our result shows that to achieve small errors we need a highly resourceful embezzling catalyst. In particular, we recover the optimal lower bound for embezzlement already established for entanglement theory [69,71,72] and we extend it, in principle, to any resource theory.

Sketch of the proof of Theorem 1.—We only give a sketch of the proof below but leave the formal derivation to the Supplemental Material, Sec. V [28], and Appendix A. We will need the *smoothed sandwiched quantum Rényi divergence*, which is defined for two states $\rho, \sigma \in \mathcal{S}(\mathcal{H})$ and $\alpha \in [1/2, 1)$ as

$$\tilde{D}_\alpha^\varepsilon(\rho \parallel \sigma) := \max \{ \tilde{D}_\alpha(\tilde{\rho} \parallel \sigma) : \tilde{\rho} \in \mathcal{S}_\varepsilon(\mathcal{H}), P(\tilde{\rho}, \rho) \leq \varepsilon \},$$

where $\mathcal{S}_\varepsilon(\mathcal{H})$ is the set of subnormalized states. An important ingredient in the proof of the above theorem is the data-processing inequality for this quantity. We believe this result to be of independent interest. In the Supplemental Material, Sec. II [28], and Appendix B, we give a proof and we also argue why a similar result does not hold for some other generalizations of Rényi divergence. We note that the use of subnormalized states in the definition of the smoothed sandwiched quantum Rényi divergence turns out to be crucial for $\alpha \in [\frac{1}{2}, 1)$, which is in contrast to the case $\alpha > 1$.

Theorem 2. Let $\rho, \sigma \in \mathcal{S}(\mathcal{H})$ be two states and \mathcal{E} a quantum channel. For any $\alpha \in [1/2, 1)$

$$\tilde{D}_\alpha^\varepsilon(\rho \parallel \sigma) \geq \tilde{D}_\alpha^\varepsilon(\mathcal{E}(\rho) \parallel \mathcal{E}(\sigma)).$$

Another key ingredient of our proof is the following continuity bound for the quantum sandwiched Rényi divergences in the interval $\alpha \in (0, 1)$.

Proposition 1. Let $\alpha \in (0, 1)$ and $\rho, \sigma \in \mathcal{S}_\varepsilon(\mathcal{H})$. Then for any $\tilde{\rho} \in \mathcal{S}_\varepsilon(\mathcal{H})$ such that $\Delta(\rho, \tilde{\rho}) \leq \varepsilon \leq \tilde{Q}_\alpha(\rho \parallel \sigma)^{(1/\alpha)}$ we have

$$|\tilde{D}_\alpha(\rho \parallel \sigma) - \tilde{D}_\alpha(\tilde{\rho} \parallel \sigma)| \leq \frac{1}{\alpha - 1} \log \left(1 - \frac{\varepsilon^\alpha}{\tilde{Q}_\alpha(\rho \parallel \sigma)} \right)$$

where we introduced generalized trace distance [67] which for normalized states is defined as $2\Delta(\rho, \sigma) := \|\rho - \sigma\|_1$ and the function $\tilde{Q}_\alpha(\rho \parallel \sigma) = \exp(\alpha - 1)\tilde{D}_\alpha(\rho \parallel \sigma)$. We remark

that the previous bound does not depend explicitly on the dimension of the Hilbert space of the states. Moreover, the previous proposition implies that the resource monotones \mathfrak{D}_α are also continuous (see the Supplemental Material, Corollary 4 [28]).

The main idea of the proof of the main theorem is that we choose a pair of states (ρ, ρ') such that $\mathfrak{D}_\alpha(\rho) < \mathfrak{D}_\alpha(\rho')$, and hence, since \mathfrak{D}_α is tensor additive by assumption, the data-processing inequality for \mathfrak{D}_α for any (uncorrelated) catalytic transformation taking ρ to ρ' is strictly violated. Moreover, for any ε -correlated catalytic transformation with catalyst ν mapping ρ into ρ' , we have both

$$\begin{aligned} \mathfrak{D}_\alpha(\rho \otimes \nu) &< \mathfrak{D}_\alpha(\rho' \otimes \nu) \quad \text{and} \\ \mathfrak{D}_\alpha^\varepsilon(\rho \otimes \nu) &\geq \mathfrak{D}_\alpha^\varepsilon(\tau) \geq \mathfrak{D}_\alpha(\rho' \otimes \nu), \end{aligned}$$

where the inequalities on the second line are due to the monotonicity for the transformation (Theorem 2), including the catalyst and our assumption that τ is ε -close to $\rho' \otimes \nu$.

However, these two inequalities lead to a tension with the continuity of \mathfrak{D}_α , which ensures that $\mathfrak{D}_\alpha(\rho \otimes \nu)$ and $\mathfrak{D}_\alpha^\varepsilon(\rho \otimes \nu)$ are arbitrarily close as ε decreases. We then show that this tension can only be relieved if $\mathfrak{D}_\alpha(\nu)$ grows large when ε decreases.

In the following, we quickly summarize the consequences of Theorem 1 for the resource theory of athermality, entanglement theory, and resource theory of coherence. In each resource theory we will specify the set of free states; our results apply to any resource theory compatible with this choice of free states. To apply Theorem 1, for each resource theory we first discuss the additivity of \mathfrak{D}_α , and we then check if there exist states in $\overline{\mathcal{CCO}}$ that satisfy the conditions of the theorem. In particular, in each resource theory we find states that satisfy both $\mathfrak{D}(\rho) \geq \mathfrak{D}(\rho')$ and $\mathfrak{F}(\rho) > \mathfrak{F}(\rho')$.

Resource theory of athermality.—In resource theory of athermality the thermal or Gibbs state $\gamma = e^{-\beta H}/Z$ is the only free state. Here, β is the inverse temperature, H is the Hamiltonian of the system and Z is the normalisation factor (partition function). The relevant resource measure is the non-equilibrium free energy [1], $\mathfrak{D}(\rho) = D(\rho||\gamma)$. We remark that our results apply to both the resource theory of athermality with thermal operations and resource theory of athermality under Gibbs preserving maps since in both resource theories free operations keep the Gibbs state invariant [8,73]. However since for the former we do not know the resource monotone characterizing the set $\overline{\mathcal{CCO}}$, for our considerations we will mainly focus on the latter where the relevant resource monotone is the nonequilibrium free energy [19,22].

The resource monotones \mathfrak{D}_α are trivially additive, and we prove in the Supplemental Material, Sec. VI [28], that there exist states in $\overline{\mathcal{CCO}}$ satisfying conditions of Theorem 1. In particular, we find numerically pairs of

qubit states with a nonzero fidelity gap, and we construct analytically pairs of classical qutrit states with a fidelity gap arbitrarily close to 1.

From Theorem 1, we get that the nonequilibrium free energy of any catalyst must satisfy $\mathfrak{D}(\nu) = \Omega[\log(1/\varepsilon)]$. Therefore, a correlated catalytic transformation between any two states would require one to prepare a catalyst with an unbounded amount of free energy as the error vanishes. Moreover, we show that the protocol discussed in Ref. [22] is optimal, i.e., $\mathfrak{D}(\nu) = \Theta[\log(1/\varepsilon)]$. With this protocol, any state in $\overline{\mathcal{CCO}}$ can be reached up to arbitrary accuracy. This means that it reaches exactly some states in $\mathcal{CCO} \setminus \overline{\mathcal{CO}}$ (with finite residual correlations for finite resourceful catalysts). This method generalizes to correlated catalytic transformations the already known construction of the catalyst introduced in Ref. [27] for (uncorrelated) catalytic transformations. This method provides a recipe to construct the catalyst whenever the states are asymptotically transformable.

To prove that it is optimal we use the exponential upper bound for the convergence of the error in approximate asymptotic pairwise state transformation [74]. In Ref. [74] the authors provided a method to obtain a lower bound for the error exponent which controls the exponential convergence of the error to zero with the number of copies. In this Letter, we give a qualitative first order expansion of the error exponent for small relative entropy gaps. We find that, under some mild regularity conditions, the error exponent γ satisfies $\gamma \geq \Delta D^2 \log e/8(V_1 + V_2) + O(\Delta D^3)$ where $\Delta D := D(\rho_1||\sigma_1) - D(\rho_2||\sigma_2)$ is the relative entropy gap and $V_i := V(\rho_i||\sigma_i) = \text{Tr}[\rho_i(\log \rho_i - \log \sigma_i)^2] - D(\rho_i||\sigma_i)^2$ is the relative entropy variance. We remark that the above expression shows the appropriate scaling behavior with the number of copies of the states (see the Supplemental Material, Sec. VI [28], and Appendix C for more details).

Entanglement theory.—In this case, the separable states are the free states of the theory [4,6]. In the following, we consider input and output bipartite pure states $|\psi_{AB}\rangle, |\psi'_{AB}\rangle$, but allow general mixed catalysts during the protocol. The resource monotone characterizing the set of pure states in $\overline{\mathcal{CCO}}$ is the relative entropy of entanglement [23]. Moreover, the resource monotones \mathfrak{D}_α are additive when one state is pure [75].

Therefore, our main theorem implies that for pairs of pure states satisfying the conditions of the main theorem any correlated catalytic transformation needs a catalyst with a diverging amount of relative entropy of entanglement as the error approaches zero. We then construct states with a fidelity gap arbitrarily close to 1 (see the Supplemental Material, Sec. VII [28], for more details).

Resource theory of coherence.—Fixing a basis $\{|i\rangle, i = 1, \dots, d\}$, we say that a state is free if it is diagonal in such a basis [10]. We consider output pure states where the monotone that characterizes the set $\overline{\mathcal{CCO}}$ is the relative entropy of coherence [10,24,76]. All the monotones \mathfrak{D}_α

are additive [[77], Theorem 3]. In our work, we give an independent proof of additivity of $\mathfrak{D}_{1/2}$ by finding an Alberti's form of the fidelity of coherence $\mathfrak{F}(\rho) := \max_{\sigma \in \mathcal{F}} F(\rho, \sigma)$ through semidefinite program (SDP) formulation.

Theorem 3. Let $\rho \in \mathcal{S}(\mathcal{H})$. The fidelity of coherence is the solution of the minimization problem

$$\mathfrak{F}(\rho) = \inf_{R>0} \text{Tr}[\rho R^{-1}] \|\Delta(R)\|_{\infty}$$

where Δ is the dephasing operator $\Delta(\cdot) = \sum_i |i\rangle\langle i| \cdot |i\rangle\langle i|$.

We believe that this result is of independent interest since it allows one to efficiently compute this quantity for which, to the best of our knowledge, an analytic form is known only for pure states [77].

Also in this case, we prove the existence of states in $\overline{\mathcal{CO}}$ satisfying the conditions of Theorem 1 with a fidelity gap arbitrarily close to 1. We obtain from Theorem 1 for the relative entropy of coherence of the catalyst $\mathfrak{D}(\nu) = D(\nu \| \Delta(\nu)) = \Omega[\log(1/\varepsilon)]$. Hence, we establish that to perform the correlated catalytic transformation we would need, at least for some states, to prepare a catalyst with a diverging amount of coherence as the error vanishes. (See the Supplemental Material, Sec. VIII [28], for more details).

Conclusion and open questions.—In this Letter, we established that for some correlated catalytic processes a small residual correlation between the system and the catalyst implies a highly resourceful catalyst. We also show similarly how in the context of imperfect catalysis a small error is only possible with a highly resourceful embezzling catalyst. Our results apply to resource theories for which certain resource monotones are tensor additive. We point out that a characterization of the sets $\overline{\mathcal{CO}}$ and $\overline{\mathcal{CO}}$, and therefore of the set $\overline{\mathcal{CO}} \setminus \mathcal{CO}$, is not known for many resource theories. Hence, the range of applicability of our main theorem and whether unbounded resources for the catalyst are required in such theories are still open questions.

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- [1] For our purposes, quantum operations or quantum channels are completely positive and trace-preserving maps from linear operators on an input to linear operators on an output Hilbert space.
- [2] E. Chitambar and G. Gour, Quantum resource theories, *Rev. Mod. Phys.* **91**, 025001 (2019).

- [3] F. G. S. L. Brandao and G. Gour, Reversible Framework for Quantum Resource Theories, *Phys. Rev. Lett.* **115**, 070503 (2015).
- [4] R. Horodecki, P. Horodecki, M. Horodecki, and K. Horodecki, Quantum entanglement, *Rev. Mod. Phys.* **81**, 865 (2009).
- [5] M. B. Plenio and S. S. Virmani, An Introduction to Entanglement Theory, in *Quantum Information and Coherence*, edited by E. Andersson and P. Ohberg (Springer, Cham, 2014), 10.1007/978-3-319-04063-9_8.
- [6] V. Vedral, M. B. Plenio, M. A. Rippin, and P. L. Knight, Quantifying Entanglement, *Phys. Rev. Lett.* **78**, 2275 (1997).
- [7] F. G. S. L. Brandao, M. Horodecki, J. Oppenheim, J. M. Renes, and R. W. Spekkens, Resource Theory of Quantum States Out of Thermal Equilibrium, *Phys. Rev. Lett.* **111**, 250404 (2013).
- [8] P. Faist, J. Oppenheim, and R. Renner, Gibbs-preserving maps outperform thermal operations in the quantum regime, *New J. Phys.* **17**, 043003 (2015).
- [9] M. Horodecki and J. Oppenheim, Fundamental limitations for quantum and nanoscale thermodynamics, *Nat. Commun.* **4**, 2059 (2013).
- [10] A. Winter and D. Yang, Operational Resource Theory of Coherence, *Phys. Rev. Lett.* **116**, 120404 (2016).
- [11] J. Aberg, Quantifying superposition, [arXiv:quant-ph/0612146](https://arxiv.org/abs/quant-ph/0612146).
- [12] T. Baumgratz, M. Cramer, and M. B. Plenio, Quantifying Coherence, *Phys. Rev. Lett.* **113**, 140401 (2014).
- [13] We consider the closure within the same fixed output space. Note that other definitions might lead to different necessary and sufficient conditions [14, 15].
- [14] M. Klimesh, Inequalities that collectively completely characterize the catalytic majorization relation, [arXiv:0709.3680](https://arxiv.org/abs/0709.3680).
- [15] G. Aubrun and I. Nechita, Catalytic majorization and ℓ_p norms, *Commun. Math. Phys.* **278**, 133 (2008).
- [16] M. Lostaglio, M. P. Müller, and M. Pastena, Stochastic Independence as a Resource in Small-Scale Thermodynamics, *Phys. Rev. Lett.* **115**, 150402 (2015).
- [17] M. P. Müller and M. Pastena, A generalization of majorization that characterizes Shannon entropy, *IEEE Trans. Inf. Theory* **62**, 1711 (2016).
- [18] F. Sapienza, F. Cerisola, and A. J. Roncaglia, Correlations as a resource in quantum thermodynamics, *Nat. Commun.* **10**, 2492 (2019).
- [19] M. P. Müller, Correlating Thermal Machines and the Second Law at the Nanoscale, *Phys. Rev. X* **8**, 041051 (2018).
- [20] S. Rethinasamy and M. M. Wilde, Relative entropy and catalytic relative majorization, *Phys. Rev. Research* **2**, 033455 (2020).
- [21] H. Wilming, Entropy and Reversible Catalysis, *Phys. Rev. Lett.* **127**, 260402 (2021).
- [22] N. Shiraishi and T. Sagawa, Quantum Thermodynamics of Correlated-Catalytic State Conversion at Small Scale, *Phys. Rev. Lett.* **126**, 150502 (2021).
- [23] T. V. Kondra, C. Datta, and A. Streltsov, Catalytic Entanglement, *Phys. Rev. Lett.* **127**, 150503 (2021).
- [24] R. Takagi and N. Shiraishi, Correlation in Catalysts Enables Arbitrary Manipulation of Quantum Coherence, *Phys. Rev. Lett.* **128**, 240501 (2022).

- [25] R. Gallego, J. Eisert, and H. Wilming, Thermodynamic work from operational principles, *New J. Phys.* **18**, 103017 (2016).
- [26] H. Wilming, R. Gallego, and J. Eisert, Axiomatic characterization of the quantum relative entropy and free energy, *Entropy* **19**, 241 (2017).
- [27] R. Duan, Y. Feng, X. Li, and M. Ying, Multiple-copy entanglement transformation and entanglement catalysis, *Phys. Rev. A* **71**, 042319 (2005).
- [28] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevLett.129.120506> for the proof of the main theorem and all the technical details of the results mentioned in the main text, which includes Refs. [29–62].
- [29] P. Boes, N. H. Y. Ng, and H. Wilming, Variance of relative surprisal as single-shot quantifier, *PRX Quantum* **3**, 010325 (2022).
- [30] P. Lipka-Bartosik and P. Skrzypczyk, All States are Universal Catalysts in Quantum Thermodynamics, *Phys. Rev. X* **11**, 011061 (2021).
- [31] B. Regula, K. Bu, R. Takagi, and Z.-W. Liu, Benchmarking one-shot distillation in general quantum resource theories, *Phys. Rev. A* **101**, 062315 (2020).
- [32] S. Turgut, Catalytic transformations for bipartite pure states, *J. Phys. A* **40**, 12185 (2007).
- [33] T. Fritz, A local-global principle for preordered semirings and abstract positivstellens” atze, [arXiv:2003.13835](https://arxiv.org/abs/2003.13835).
- [34] L.-H. Shao, Z. Xi, H. Fan, and Y. Li, Fidelity and trace-norm distances for quantifying coherence, *Phys. Rev. A* **91**, 042120 (2015).
- [35] K. Li and A. Winter, Relative entropy and squashed entanglement, *Commun. Math. Phys.* **326**, 63 (2014).
- [36] J. Watrous, *The Theory of Quantum Information* (Cambridge University Press, Cambridge, England, 2018).
- [37] J. Watrous, Simpler semidefinite programs for completely bounded norms, [arXiv:1207.5726](https://arxiv.org/abs/1207.5726).
- [38] W. Zhou and F. Buscemi, General state transitions with exact resource morphisms: A unified resource-theoretic approach, *J. Phys. A* **53**, 445303 (2020).
- [39] X. Wang and M. M. Wilde, Resource theory of asymmetric distinguishability, *Phys. Rev. Research* **1**, 033170 (2019).
- [40] C. T. Chubb, M. Tomamichel, and K. Korzekwa, Beyond the thermodynamic limit: Finite-size corrections to state interconversion rates, *Quantum* **2**, 108 (2018).
- [41] R. Renner, Security of quantum key distribution, *Int. J. Quantum. Inform.* **06**, 1 (2008).
- [42] M. Steiner, Generalized robustness of entanglement, *Phys. Rev. A* **67**, 054305 (2003).
- [43] G. Vidal and R. Tarrach, Robustness of entanglement, *Phys. Rev. A* **59**, 141 (1999).
- [44] N. Datta, Max-relative entropy of entanglement, alias log robustness, *Int. J. Quantum. Inform.* **07**, 475 (2009).
- [45] N. Datta, Min-and max-relative entropies and a new entanglement monotone, *IEEE Trans. Inf. Theory* **55**, 2816 (2009).
- [46] A. Winter, Tight uniform continuity bounds for quantum entropies: Conditional entropy, relative entropy distance and energy constraints, *Commun. Math. Phys.* **347**, 291 (2016).
- [47] C. H. Bennett, S. Popescu, D. Rohrlich, J. A. Smolin, and A. V. Thapliyal, Exact and asymptotic measures of multipartite pure-state entanglement, *Phys. Rev. A* **63**, 012307 (2000).
- [48] M. Christandl and A. Winter, squashed entanglement: An additive entanglement measure, *J. Math. Phys. (N.Y.)* **45**, 829 (2004).
- [49] R. Alicki and M. Fannes, Continuity of quantum conditional information, *J. Phys. A* **37**, L55 (2004).
- [50] C. H. Bennett, H. J. Bernstein, S. Popescu, and B. Schumacher, Concentrating partial entanglement by local operations, *Phys. Rev. A* **53**, 2046 (1996).
- [51] C. H. Bennett, H. J. Bernstein, S. Popescu, and B. Schumacher, Concentrating partial entanglement by local operations, *Phys. Rev. A* **53**, 2046 (1996).
- [52] J. M. Renes, Relative submajorization and its use in quantum resource theories, *J. Math. Phys. (N.Y.)* **57**, 122202 (2016).
- [53] F. Buscemi and G. Gour, Quantum relative Lorenz curves, *Phys. Rev. A* **95**, 012110 (2017).
- [54] D. Petz, Quasi-entropies for finite quantum systems, *Rep. Math. Phys.* **23**, 57 (1986).
- [55] F. Dupuis, L. Kraemer, P. Faist, J. M. Renes, and R. Renner, Generalized entropies, in *XVIIth International Congress on Mathematical Physics* (World Scientific, Singapore, 2014), pp. 134–153.
- [56] V. Vedral and M. B. Plenio, Entanglement measures and purification procedures, *Phys. Rev. A* **57**, 1619 (1998).
- [57] K. M. Audenaert, M. Mosonyi, and F. Verstraete, Quantum state discrimination bounds for finite sample size, *J. Math. Phys. (N.Y.)* **53**, 122205 (2012).
- [58] A. Anshu, M. Berta, R. Jain, and M. Tomamichel, A minimax approach to one-shot entropy inequalities, *J. Math. Phys. (N.Y.)* **60**, 122201 (2019).
- [59] R. Bhatia, *Matrix Analysis*, Graduate Texts in Mathematics Vol. 169 (Springer, New York, 1997), [10.1007/978-1-4612-0653-8](https://doi.org/10.1007/978-1-4612-0653-8).
- [60] E. Carlen, Trace inequalities and quantum entropy: An introductory course, *Entropy Quant.* **529**, 73 (2010).
- [61] A. Marwah and F. Dupuis, Uniform continuity bound for sandwiched Rényi conditional entropy, *J. Math. Phys. (N.Y.)* **63**, 052201 (2022).
- [62] M. A. Nielsen, Conditions for a Class of Entanglement Transformations, *Phys. Rev. Lett.* **83**, 436 (1999).
- [63] P. Lipka-Bartosik and P. Skrzypczyk, Catalytic Quantum Teleportation, *Phys. Rev. Lett.* **127**, 080502 (2021).
- [64] M. Tomamichel, R. Colbeck, and R. Renner, Duality between smooth min-and max-entropies, *IEEE Trans. Inf. Theory* **56**, 4674 (2010).
- [65] M. Müller-Lennert, F. Dupuis, O. Szehr, S. Fehr, and M. Tomamichel, On quantum Rényi entropies: A new generalization and some properties, *J. Math. Phys. (N.Y.)* **54**, 122203 (2013).
- [66] M. M. Wilde, A. Winter, and D. Yang, Strong converse for the classical capacity of entanglement-breaking and hadamard channels via a sandwiched Rényi relative entropy, *Commun. Math. Phys.* **331**, 593 (2014).
- [67] M. Tomamichel, *Quantum Information Processing with Finite Resources: Mathematical Foundations* (Springer, New York, 2015), Vol. 5.
- [68] F. Brandao, M. Horodecki, N. Ng, J. Oppenheim, and S. Wehner, The second laws of quantum thermodynamics, *Proc. Natl. Acad. Sci. U.S.A.* **112**, 3275 (2015).

- [69] W. van Dam and P. Hayden, Universal entanglement transformations without communication, *Phys. Rev. A* **67**, 060302(R) (2003).
- [70] N. H. Y. Ng, L. Mančinska, C. Cirstoiu, J. Eisert, and S. Wehner, Limits to catalysis in quantum thermodynamics, *New J. Phys.* **17**, 085004 (2015).
- [71] D. Leung and B. Wang, Characteristics of universal embezzling families, *Phys. Rev. A* **90**, 042331 (2014).
- [72] R. Cleve, L. Liu, and V. I. Paulsen, Perfect embezzlement of entanglement, *J. Math. Phys. (N.Y.)* **58**, 012204 (2017).
- [73] M. Lostaglio, Á. M. Alhambra, and C. Perry, Elementary thermal operations, *Quantum* **2**, 52 (2018).
- [74] F. Buscemi, D. Sutter, and M. Tomamichel, An information-theoretic treatment of quantum dichotomies, *Quantum* **3**, 209 (2019).
- [75] R. Rubboli and M. Tomamichel (to be published).
- [76] C. Liu, Q.-M. Ding, and D. Tong, Superadditivity of convex roof coherence measures, *J. Phys. A* **51**, 414012 (2018).
- [77] H. Zhu, M. Hayashi, and L. Chen, Coherence and entanglement measures based on rényi relative entropies, *J. Phys. A* **50**, 475303 (2017).