

Limits to catalysis in quantum thermodynamics [1]

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Quantum thermodynamics aims at fleshing out the ultimate limits of thermodynamic processes. We have seen renewed efforts in understanding fundamental laws of thermodynamics [2–5] in the quantum nano-regime, where finite size effects [6] and quantum coherences become increasingly relevant. In particular, casting thermodynamics as a resource theory [3, 4, 7, 8] allows us to borrow intuition from entanglement and information theory [9–11]. Analogous to the transformations of entangled states via local operations and classical communication (LOCC), in thermodynamics we study state transformations $\rho \rightarrow \sigma$ that can be accomplished via *thermal operations* (TO). Roughly speaking, TO refers to operations that can be implemented by coupling the system to a thermal bath of fixed temperature T , and applying any global energy-preserving unitary. Also similarly to LOCC where state conversion conditions are in terms of majorization [12, 13], the conditions for $\rho \rightarrow \sigma$ via TO take the form of *thermo-majorization* [14]. The closely related mathematical structure allows the application of many insights and tools derived in information theory.

In the context of TO, it is most natural to allow for ancillae to facilitate processes – after all, it is the ultimate limits of thermodynamic operations that are at stake. There are cases where $\rho \rightarrow \sigma$ is not possible, but there exists a state ω_C such that $\omega_C \otimes \rho \rightarrow \omega_C \otimes \sigma$ is possible. This is called *exact catalysis*. The inclusion of catalyst states in thermal operations seems a crucial and necessary step towards obtaining a complete picture of quantum thermodynamics: it allows us to describe thermodynamic transformations where the system interacts with experimental apparatus, for example a clock system (or an actual physical catalyst after all). Mathematical conditions for catalytic majorization [15, 16] have been extended to the thermodynamic setting, giving us entire families of second laws for quantum thermodynamics.

Again specifically in the thermodynamic context, it is very natural indeed to allow for *inexact catalysis*, i.e., situations in which the catalyst is returned except for a slight degradation (as measured, for example, by trace distance). Yet, in entanglement theory there exists a curious effect called *embezzling* [10, 17]: Here a state $|\mu(n)\rangle$ of dimension n , when shared between two parties Alice and Bob, is capable of achieving $|\mu(n)\rangle \xrightarrow{\text{LOCC}} |\mu(n)\rangle |\phi\rangle_{AB}$ for *any* $|\phi\rangle_{AB}$.

What is more, embezzling can be performed with arbitrarily small precision as the dimension of the embezzling state grows. There also exist *universal* embezzling catalysts that can facilitate any process $\rho \rightarrow \sigma$ [17]. When translated to thermodynamics, this means: for any $\varepsilon > 0$ and *any* two states ρ and σ , there exists ω_C such that starting from $\omega_C \otimes \rho$, we get ε -close in terms of trace distance to $\omega_C \otimes \sigma$ via TO. Table 1 lists some similarities and differences between LOCC embezzling and inexact catalysis. Clearly, in the thermodynamic context, this gives rise to a rather unreasonable situation and hence an important puzzle to be solved. In this work, we resolve this puzzle – in both a mathematically and physically plausible fashion – and provide a comprehensive analysis.

	LOCC embezzling	Inexact catalysis
State conversion conditions	Related to majorization	
Phenomena	The usage of a catalyst state of large dimension/energy while tolerating slight degradation allows the preparation of any desired target state to arbitrary precision	
Hamiltonians	Not of interest	Of much physical significance
States	Pure, multipartite states	Mixed states in general
Commonly used measure of closeness	Fidelity of global state (system and embezzling state)	Trace distance between input and output catalyst state
Allowed operations	LOCC/LO operations	Exact catalysis
Accuracy limited by	Dimension	Dimension and energy

Table 1: An overview of differences between LOCC embezzling and inexact catalysis.

Specifically, we investigate the performance of catalyst input/output pairs (ω_C, ω'_C) which are able to achieve $\rho \otimes \omega_C \rightarrow \sigma \otimes \omega'_C$, for any ρ and σ (of fixed dimension). The catalytic error is defined as the trace distance between ω_C and ω'_C . Here, we highlight the power and limitations of inexact catalysis, by reporting these results:

1. We construct an family of catalyst pairs with dimension n , which is able to facilitate any $\rho \rightarrow \sigma$, for ρ and σ being arbitrary m -dimensional states. This is analysed in the simple case where all Hamiltonians involved are trivial (proportional to identity). We show via mathematical induction that our family achieves the optimal catalytic error. The optimal error vanishes as $n \rightarrow \infty$, demonstrating the power of catalysis.
2. We identify physically motivated restrictions on any catalyst pair (ω_C, ω'_C) , such that if the catalyst pair is able to facilitate any state transformation process, then the catalytic error must be lower bounded by a non-zero constant. We analyse two forms of restrictions: (a) catalysts with bounded dimension, and (b) catalysts (possibly infinite dimensional) such that the partition function Z_C is finite, and the average energy of ω_C (input catalyst) is finite.

Our first result concerning the construction of a family of catalysts partially contributes to the search for optimal embezzling states in LOCC setting [10, 11]. From a more physical viewpoint, the second part of our results strongly limits the possibility of drastically enlarging the set of allowed processes in quantum thermodynamics, when systems with physically relevant Hamiltonians are considered. Our methodology makes use of tools from quantum information, furthermore we use convex relaxation techniques to arrive at part of our second result, which is a novel approach towards investigating such problems. We hope that the use of such information theoretic approaches continue to unearth the reality of quantum thermodynamical processes.

The power of inexact catalysis. We first explore the case of trivial Hamiltonians, the simplest case of thermal operations when the Hamiltonian is proportional to the identity operator. The conditions governing a transition $\rho \rightarrow \sigma$ is that the eigenvalue vector of ρ majorizes that of σ [3], denoted as $\rho \succ \sigma$. Given fixed integers m, n , we ask what is the smallest $\varepsilon(m, n)$ such that there exists a catalyst state pair (ω_C, ω'_C) that facilitates $\rho \rightarrow \sigma$ for any ρ, σ being m -dimensional states, while the trace distance $d(\omega_C, \omega'_C) \leq \varepsilon(m, n)$. This is used as a measure of catalytic error throughout our analysis. We show that this can be simplified by considering a single transformation

$$\omega_C \otimes \mathbb{I}/m \succ \omega'_C \otimes |0\rangle\langle 0|. \quad (1)$$

This is because if some catalyst pair (ω_C, ω'_C) satisfies Eq. (1), then it also facilitates $\omega_C \otimes \rho \rightarrow \omega'_C \otimes \sigma$ for any m -dimensional states ρ, σ . Therefore, this example is uniquely interesting since the optimal error for this transformation is also optimal for inducing arbitrary state transformations within this fixed Hilbert space.

Since majorization conditions depend solely on the eigenvalues of the density matrices ω_C and ω'_C , we show that finding the optimal $\varepsilon(m, n)$ can be casted as the solution of a linear minimization program over catalyst states diagonal and ordered in the same basis. By running this linear program for small m and n , we were able to guess the form of the optimal catalyst pair. We then prove that this construction is indeed optimal and obtained the following theorem.

Theorem 1 (Power of inexact catalysis) *For $m \geq 2$ and $n = m^a$ where $a \geq 1$ is an integer, the optimal catalytic error is $\varepsilon(m, n) = \frac{m-1}{1+(m-1)\log_m n}$. Let $\omega_C = \sum_{i=1}^n \omega_i |i\rangle\langle i|$, where $\omega_1 = m/[1 + (m-1)a]$,*

$$\omega_i = \begin{cases} \omega_1 m^{-\lceil \log_m i \rceil} & \text{if } 2 \leq i \leq n/m, \\ 0 & \text{if } i > n/m. \end{cases} \quad (2)$$

Define ω'_C to be such that $\omega'_1 = \omega_1/m$, $\omega'_i = \omega_i$ for $i \in [2, n/m]$ and $\omega'_i = m^{1-a}/[1 + (m-1)a]$ for $i > n/m$. Then (ω_C, ω'_C) achieves $\varepsilon(m, n)$.

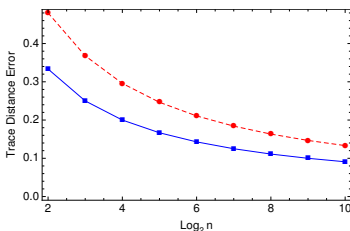


Figure 1: The comparison of trace distance error for our state ω_C (blue, solid) and $\tilde{\omega}_C$ (red, dashed) for $m = 2$.

In Fig. 1 we compare the catalytic error achieved by our catalyst, compared to the state proposed in Ref. [17]. We see that for small dimensions ω_C outperforms $\tilde{\omega}_C$, however asymptotically the error scales with $\log n$ for both catalysts.

The limits of inexact catalysis. Here, our goal is to identify physical restrictions that prevent the existence of universal catalysts with arbitrarily small catalytic error. Given Hamiltonians H_S, H_C , we ask what is the smallest ε such that there exists (ω_C, ω'_C) where

$$\omega_C \otimes \tau_S \rightarrow \omega'_C \otimes \Pi_{\max}^S \quad (3)$$

is possible via TO, where $\Pi_{\max}^S = |E_{\max}^S\rangle\langle E_{\max}^S|$ is the pure energy eigenstate with energy E_{\max}^S , and $d(\omega_C, \omega'_C) \leq \varepsilon$. Intuitively, this process is the hardest case of state transformation between diagonal states in the energy eigenbasis. More precisely, if Eq. (3) holds, then for any ρ_S and ρ'_S diagonal in H_S , $\omega_C \otimes \rho_S \rightarrow \omega'_C \otimes \rho'_S$ is also possible.

It is shown [2] that the monotonicity of quantum Rényi divergences [18] are necessary conditions for state conversions. More precisely, given some H_S and T , for any ρ_S and ρ'_S , if $\rho_S \rightarrow \rho'_S$ via exact catalysis, then for all $\alpha \geq 0$, $D_\alpha(\rho_S \| \tau_S) \geq D_\alpha(\rho'_S \| \tau_S)$ holds, where τ_S is the thermal state of system S , at temperature T of the thermal bath. Therefore, this allows us to lower bound ε by invoking the monotonicity of D_α for any $\alpha \geq 0$, i.e. defining

$$\begin{aligned} \varepsilon_\alpha := \min & \quad \frac{1}{2} \|\omega_C - \omega'_C\|_1 \\ \text{s.t.} & \quad D_\alpha(\omega_C \otimes \tau_S \| \tau_{CS}) \geq D_\alpha(\omega'_C \otimes \Pi_{\max}^S \| \tau_{CS}), \quad 0 \leq \omega_C, \omega'_C \leq \mathbb{I}, \end{aligned} \quad (4)$$

where $\tau_{CS} = \tau_C \otimes \tau_S$ is the thermal state of the catalyst and system. By using Eq. (4), we derive Theorem 2.

Theorem 2 (Finite dimension) Consider system and catalyst Hamiltonians H_S, H_C which are finite-dimensional, and denote $\{E_i^S\}_{i=1}^m, \{E_i^C\}_{i=1}^n$ to be the set of energy eigenvalues respectively. For any catalyst pair (ω_C, ω'_C) where $\omega_C \otimes \tau_S \rightarrow \omega'_C \otimes \Pi_{\max}^S$ is possible via TO, the catalytic error

$$\varepsilon \geq \varepsilon_\infty \geq \left(\frac{Z_S}{e^{-\beta E_{\max}^S}} - 1 \right) \frac{e^{-\beta E_{\max}^C}}{Z_C} \neq 0. \quad (5)$$

To consider infinite-dimensional catalysts, we use Eq. (6) for $\alpha = 1/2$. Method-wise, we first use relaxation techniques to split the non-convex minimization into smaller and significantly easier convex subproblems involving independent variables. By exploiting Lagrange duality in these nested optimization programs we arrive at the following theorem.

Theorem 3 (Finite initial average energy) Consider any system and catalyst Hamiltonian H_S, H_C , and assume that $\dim(H_S) \geq 2$. For any diagonal catalyst pair (ω_C, ω'_C) where $\omega_C \otimes \tau_S \rightarrow \omega'_C \otimes \Pi_{\max}^S$ is possible via TO, and if $\text{tr}(H_C \omega_C) \leq E$, then the catalytic error is lower bounded by

$$\varepsilon \geq \varepsilon_1^2 / (9Z_C), \quad (6)$$

where $\varepsilon_1 = \max_{W \in (0,1)} W \gamma^{E_j^C(W)}$ and $j(W) = \min\{j : E_{j+1}^C > E/(1-W)\}$.

Theorems 2 and 3 prove a strictly positive lower bound on the catalytic error. In particular, Theorem 3 has remarkable implications, i.e. even with infinite dimensional catalysts, as long as the catalyst Hamiltonian corresponds to a finite partition function, and the input catalyst has finite average energy, one cannot find universal catalyst pairs that facilitate arbitrary state transformations with vanishing error. Such a phenomenon is distinctively different from the LOCC setting, which does not consider the role of Hamiltonians in entanglement resources. The lower bounds in Theorem 3 can also be generalized to tighter versions, depending on the form of H_S . We summarize our findings in Table 2. Note that both our case studies above can be modified to study specific state transformations ρ_S to ρ'_S as well, obtaining lower bounds similar to Eq. (6), but state dependent. This is useful if one wants to account for more specific cases of resource generation (instead of studying universal catalyst pairs). Precise details of this work can be found in Ref. [1].

Energy levels \ Dimension	Bounded	Unbounded
	Fully degenerate	No
Bounded	No	True at least for fully degenerate Hamiltonians
Unbounded	N/A	No, if average energy and partition function is finite

Table 2: The possibility of inexact catalysis for all state conversion processes with arbitrary precision.

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