

## Entropy and Reversible Catalysis

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I show that nondecreasing entropy provides a necessary and sufficient condition to convert the state of a physical system into a different state by a reversible transformation that acts on the system of interest and a further “catalyst,” whose state has to remain invariant exactly in the transition. This statement is proven both in the case of finite-dimensional quantum mechanics, where von Neumann entropy is the relevant entropy, and in the case of systems whose states are described by probability distributions on finite sample spaces, where Shannon entropy is the relevant entropy. The results give an affirmative resolution to the (approximate) catalytic entropy conjecture introduced by Boes *et al.* [*Phys. Rev. Lett.* **122**, 210402 (2019)]. They provide a complete single-shot characterization without external randomness of von Neumann entropy and Shannon entropy. I also compare the results to the setting of phenomenological thermodynamics and show how they can be used to obtain a quantitative single-shot characterization of Gibbs states in quantum statistical mechanics.

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A central question in quantum information theory is which quantum states on some physical system may be transformed into which other states on the same (or a different) physical system by a given set of operations. This question underlies quantum resource theories, such as entanglement [1–4], thermodynamics [5–10], or asymmetry [11–13] (see Ref. [14] for a review on quantum resource theories). Common to most resource theories is that they allow for probabilistic mixing of operations, i.e., to use a source of classical randomness (such as a coin toss) to decide on the operation that is implemented. This use of randomness immediately implies that the resource theory is *convex*, which greatly simplifies the mathematical analysis. While often a natural assumption due to the commonplace access to (quasi)randomness and classical communication, one may ask what happens if either one considers the cost of classical or quantum randomness explicitly or simply disallows the use of classical or quantum randomness. In the most extreme limit, one would then end up only allowing the use of unitary operations and the question of which states can be interconverted becomes trivial: namely, all those states that are unitarily invariant, or in other words, all states with the same spectrum (including multiplicities).

In many resource theories the use of a catalyst is operationally well motivated and may greatly enrich the set of possible state transitions [9,15–22]. A catalyst is a system that remains invariant in a given process, but may or may not, depending on the resource theory, build up correlations to other systems. Because its state does not change in a process, no resources are used up and the catalyst may be used again to facilitate further state

transitions on other systems—this is conceptually similar to catalysts in chemistry or the ubiquitous “periodically working machines” in thermodynamics. One may therefore wonder what happens if we consider only unitary operations together with the possibility for catalysts. Interestingly, Ref. [22] found that in this setting the von Neumann entropy plays a special role due to its subadditivity property (see below). It was, in fact, conjectured that the von Neumann entropy is the *only* constraint for state transitions once one allows for arbitrary small errors on the system (but not the catalyst). This conjecture was called “catalytic entropy conjecture” and can also be formulated in the classical setting as a conjecture connecting Shannon entropy to catalytic permutations of probability distributions. The ideas behind this conjecture already have found application in the context of thermodynamics and fluctuation theorems [23,24].

The special role of von Neumann entropy is surprising for several reasons: Typically, the Shannon or von Neumann entropy appears in settings involving many weakly correlated systems due to the phenomenon of “typicality” [25–27]. It was therefore long believed that the von Neumann entropy only plays a special role in asymptotic settings, such as the thermodynamic limit in physics or the limit of many identically and independently distributed signals in information theory. In particular, in resource theories involving free randomness and allowing for catalysts (that, however, may not become correlated to the system of interest), state transitions are usually characterized by an infinite set of constraints [9,18–20]. It was only very recently first conjectured and then proven that these conditions may collapse to the von Neumann entropy

(or similar quantities, such as relative entropy or free energy) if one allows for catalysts that become correlated to the system [28–34]. However, these settings still made use of external randomness—either by allowing for classical randomness explicitly or allowing free access to systems such as heat baths, which can be seen as sources of randomness. It is thus interesting that the catalytic entropy conjecture posits that von Neumann entropy plays such a special role in situations that neither allow for the use of external randomness, nor require an asymptotic limit. This Letter provides an affirmative resolution of the catalytic entropy conjecture.

The catalytic entropy conjecture can also be formulated in full analogy in the classical case by replacing finite-dimensional density matrices with probability distributions on finite sample spaces, unitary operations with permutations (reversible transformations on the sample space), and von Neumann entropy with Shannon entropy. In the main text of this Letter, we will only consider the quantum case and only briefly comment on how to prove the classical version of our main result. A full proof of the classical result is given in the Supplemental Material [35]. While the proof of the classical result also implies the quantum result, it has the drawback of requiring a larger catalyst, in general.

*Setting and main result.*—Throughout we consider a system  $S$  described by density matrices  $\rho$  and  $\rho'$  on a Hilbert space of dimension  $d$ . In the following, we write  $H(\rho)$  for von Neumann entropy, which is defined as

$$H(\rho) = -\text{Tr}[\rho \log(\rho)]. \quad (1)$$

Von Neumann entropy is continuous in  $\rho$  [38,39] and has several useful properties, such as unitary invariance [ $H(\rho) = H(U\rho U^\dagger)$  for any unitary  $U$ ], additivity [ $H(\rho \otimes \sigma) = H(\rho) + H(\sigma)$ ], and subadditivity:  $H(\rho_{12}) \leq H(\rho_1) + H(\rho_2)$ , where  $\rho_{12}$  is a bipartite quantum state with marginals  $\rho_1$  and  $\rho_2$ . Finally, denote by  $D(\rho, \rho') := \frac{1}{2}\|\rho - \rho'\|_1$  the trace distance between two density matrices. We then define catalytic state transitions formally as follows (see also Fig. 1).

*Definition 1.*—*Definition 1 (approximate catalytic transformation).*—Consider two finite-dimensional density matrices  $\rho$  and  $\rho'$  on the same system  $S$ . We write  $\rho \rightarrow_\epsilon \rho'$  if there exists a finite-dimensional density matrix  $\sigma$  on a system  $C$  and a unitary  $U$  on  $SC$  such that

$$\text{Tr}_S[U\rho \otimes \sigma U^\dagger] = \sigma \quad (2)$$

and

$$D(\text{Tr}_C[U\rho \otimes \sigma U^\dagger], \rho') \leq \epsilon. \quad (3)$$

The following main result of this Letter then shows that the set of states that are reachable from a given state  $\rho$  is given exactly by the set of states with higher von Neumann entropy:

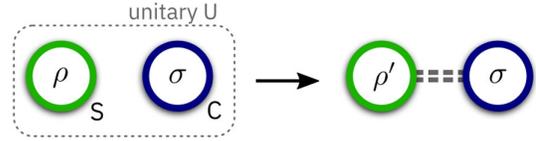


FIG. 1. A catalytic transition: A unitary operation  $U$  is applied to systems  $S$  and  $C$  in the state  $\rho \otimes \sigma$ . The resulting reduced state on  $S$  is (arbitrarily close to)  $\rho'$ , while the reduced state on  $C$  is preserved exactly, but correlated to  $S$  (indicated by the dashed lines). The main result of this Letter shows that a matching  $\sigma$  and  $U$  can be found if and only if  $H(\rho') \geq H(\rho)$ .

*Theorem 2.*—(catalytic transformations characterize von Neumann entropy).—The following are equivalent:

(i)  $\rho \rightarrow_\epsilon \rho'$  for all  $\epsilon > 0$ . (ii)  $H(\rho') \geq H(\rho)$ .

Before coming to the proof of this statement, let us first discuss the formal similarity between the theorem and the corresponding setup in thermodynamics and then give an application for quantum statistical mechanics.

*Analogy with thermodynamics.*—Let us consider the following idealized, but ubiquitous setting of a thermodynamic work process acting on three systems: A system  $S$  composed of a working substance and all other parts that may change in the process (such as heat baths), a collection of systems  $C$  composed of all systems that take part in the process but return to their initial state when the process has finished (gears, pistons, etc.), and finally, an idealized work-storage device  $W$  with vanishing entropy (e.g., a purely mechanical device such as a suspended weight in a uniform gravitational field). Since the system  $C$  is cyclic, it does not contribute any energy or entropy to the process. Similarly,  $W$  only contributes or absorbs the work necessary for the process, but does not act as a source or sink of entropy. Let the initial and final states of  $S$  be labeled by  $a$  and  $b$ , respectively. By the second law of thermodynamics, the considered process can only be possible if the entropy of  $S$  is nondecreasing:  $H(b) \geq H(a)$  (we use the same letter for thermodynamic entropy as for von Neumann or Shannon entropy). Conversely, and importantly, however, it is commonly assumed in setting up the thermodynamic framework that for any two states  $a, b$  of  $S$  there is some such idealized work process connecting the two states (for a very clear, recent exposition, see [40]). Indeed, this is required to be able to consistently define a thermodynamic entropy. Therefore, states  $a$  and  $b$  can be connected by such a process if and only if  $H(b) \geq H(a)$ , in complete analogy with Theorem 2, where the role of  $W$  is played by the external laboratory implementing the unitary  $U$ . In the thermodynamic setting, the system  $C$  is usually left implicit, but is clearly physically necessary and its precise design depends on the states  $a$  and  $b$ . While in the thermodynamic case the source of the increase of entropy is left unspecified, in the microscopic case discussed here it is due to the build up of correlations between  $S$  and  $C$ . Explicitly, we have  $H(\rho') - H(\rho) = I(S':C)$ , where

$I(S':C)$  denotes the mutual information between  $S$  and  $C$  after the unitary evolution.

*Quantitative single-shot characterization of Gibbs states.*—Let us now illustrate the use of the main result with an application in quantum statistical mechanics. We first have to establish some background material. A “passive state” is any quantum state  $\rho$  whose energy (with respect to some fixed Hamiltonian  $H$ ) cannot be lowered by a unitary transformation. Equivalently, we may say that no work can be extracted from a passive state using unitary operations. Even though  $\omega$  is passive, it is not necessarily *completely* passive, meaning that a positive amount of energy

$$\bar{W}(\rho) := \lim_{n \rightarrow \infty} \frac{1}{n} \sup_U (\text{Tr}[\rho^{\otimes n} \hat{H}_n] - \text{Tr}[U \rho^{\otimes n} U^\dagger \hat{H}_n])$$

may be extracted *per copy* if many copies of  $\rho$  are available. Here,  $\hat{H}_n := \sum_{j=1}^n H_j$  with  $H_j \equiv H$  is the total Hamiltonian on  $n$  copies. Let us assume for simplicity that the ground state of  $H$  is unique. It has been proven that [41]

$$\bar{W}(\rho) = \text{Tr}[\rho H] - \text{Tr}[\omega_{\beta(\rho)}(H)H], \quad (4)$$

where  $\omega_{\beta}(H) := \exp(-\beta H)/\text{Tr}[\exp(-\beta H)]$  denotes a Gibbs state and  $\beta(\rho) \geq 0$  is chosen such that  $H[\omega_{\beta(\rho)}(H)] = H(\rho)$  [if  $H(\rho) = 0$ , one has to take the limit  $\beta \rightarrow +\infty$ ]. Thus, the only completely passive states are ground states and Gibbs states with positive temperature [42,43].

The above characterization of Gibbs states relies on a thermodynamic limit. It was observed in Ref. [23], and discussed in detail for the particular case of three-level systems, that the energy of passive states may, nevertheless, in general, be reduced using catalytic transitions instead of unitary operations, unless the state in question is a Gibbs state. This may be surprising, since the catalyst, by virtue of not changing its state, cannot compensate for energetic changes. It was an open problem to determine how much work can be extracted from an arbitrary state using a single catalytic transformation [44]. Let us denote this quantity by

$$W_{\text{cat}}(\rho) := \sup_{\rho \rightarrow \rho'} (\text{Tr}[\rho H] - \text{Tr}[\rho' H]). \quad (5)$$

Then Theorem 2 immediately leads to the following corollary, which shows that catalysts allow one to extract the same amount of energy from a single passive states as, on average, from asymptotically many copies:

*Corollary 3.*— $W_{\text{cat}}(\rho) = \bar{W}(\rho)$ .

*Proof.*—By Theorem 2 and continuity, we have

$$W_{\text{cat}}(\rho) = \sup_{\rho': H(\rho') \geq H(\rho)} (\text{Tr}[\rho H] - \text{Tr}[\rho' H]). \quad (6)$$

By concavity of von Neumann entropy, the optimizer must have  $H(\rho') = H(\rho)$ . But by Gibbs variational

principle,  $\omega_{\beta(\rho)}(H)$  is the state with lowest energy among all states with entropy  $H(\rho)$ . Hence  $W_{\text{cat}}(\rho) = \text{Tr}[\rho H] - \text{Tr}[\omega_{\beta(\rho)}(H)H] = \bar{W}(\rho)$ . ■

*Proof of Theorem 2.*—The direction (i)  $\Rightarrow$  (ii) follows directly from subadditivity, unitary invariance, and continuity of von Neumann entropy: For a given  $\epsilon$ , denote by  $\rho'_\epsilon$  the final state on  $S$ . Then we have

$$H(\rho'_\epsilon) + H(\sigma) \geq H[U(\rho \otimes \sigma)U^\dagger] = H(\rho) + H(\sigma), \quad (7)$$

and hence  $H(\rho'_\epsilon) \geq H(\rho)$ . By continuity, we thus find  $H(\rho') \geq H(\rho)$ .

The converse direction (ii)  $\Rightarrow$  (i) requires several Lemmas. First, we collect a combination of some standard results on typicality and majorization. We write  $\rho \succeq \rho'$  if  $\rho$  majorizes  $\rho'$ , meaning that there exists a probability distribution  $q_i$  over unitaries  $V_i$  such that  $\rho' = \sum_i q_i V_i \rho V_i^\dagger$ . Similarly, we write  $\mathbf{a} \succeq \mathbf{b}$  for two vectors  $\mathbf{a}, \mathbf{b} \in \mathbb{R}^d$  if there exists a probability distribution  $q_i$  over permutation matrices  $\pi_i$  such that  $\mathbf{a} = \sum_i q_i \pi_i \mathbf{b}$ .

*Lemma 4.*—(typicality and majorization).—Let  $\rho$  and  $\rho'$  be two finite-dimensional density matrices of dimension  $d$  with  $H(\rho) < H(\rho')$ . Then for any  $\epsilon > 0$  and large enough  $n$  there exists a state  $\rho'_{\epsilon,n}$  such that  $\rho^{\otimes n} \succeq \rho'_{\epsilon,n}$  and  $D(\rho'_{\epsilon,n}, \rho'^{\otimes n}) \leq \epsilon$ . Moreover, the error  $\epsilon$  can be bounded as

$$\epsilon \leq O[\exp(-n\Delta H^2/4)], \quad (8)$$

with  $\Delta H := H(\rho') - H(\rho)$ , and  $\rho'_{\epsilon,n}$  may be chosen to have the same eigenbasis as  $\rho'^{\otimes n}$ .

A proof sketch of Lemma 4 is given in the Supplemental Material [35]. It is clear that the given error bound is not optimal for every choice of  $\rho$  and  $\rho'$ , since, for example,  $\epsilon = 0$  is possible if  $\rho \succeq \rho'$ . However, Ref. [45] shows that the given error bound is essentially optimal up to constants as a bound that does not take into account detailed information about  $\rho$  and  $\rho'$ . In the Supplemental Material [35], we use this to give an estimate of the size of the catalyst.

The next Lemma will be essential to construct a candidate catalyst by making use of Lemma 4. It is based on the Schur-Horn theorem, which states that, for any  $d \times d$  Hermitian matrix, its vector of eigenvalues  $\boldsymbol{\lambda}$  majorizes the vector of diagonal elements in every orthonormal basis. Conversely, every vector that is majorized by  $\boldsymbol{\lambda}$  may be obtained as the diagonal elements in a suitable orthonormal basis. In particular, if  $\mathbf{p}$  and  $\mathbf{p}'$  denote the ordered vectors of eigenvalues of two density matrices  $\rho$  and  $\rho'$ , respectively, then  $\rho \succeq \rho'$  if and only if  $\mathbf{p} \succeq \mathbf{p}'$ . In the following, we denote by  $\mathcal{D}_{\rho'}$  the “dephasing channel” in the eigenbasis of  $\rho'$  that acts as

$$\mathcal{D}_{\rho'}[\rho] = \sum_i |i\rangle \langle i| \rho |i\rangle \langle i|, \quad (9)$$

where the  $|i\rangle$  constitute an orthonormal eigenbasis of  $\rho'$ .

*Lemma 5.*—(basic lemma).—Let  $H(\rho) < H(\rho')$  and let  $\mathcal{D}_{\rho'}$  represent the dephasing channel in the eigenbasis of  $\rho'$ .

Then, for any  $\epsilon > 0$  there exists an  $n \in \mathbb{N}$  and a unitary  $U$  such that, for any  $1 \leq k \leq n$ ,

$$D(\rho', \mathcal{D}_{\rho'}[\chi_k]) \leq \epsilon, \quad \chi := U\rho^{\otimes n}U^\dagger, \quad (10)$$

where  $\chi_k := \text{Tr}_{\{1, \dots, n\} \setminus \{k\}}[\chi]$ . The error  $\epsilon$  scales as in Lemma 4.

*Proof.*—We make use of the state  $\rho'_{\epsilon, n}$  guaranteed by Lemma 4. By the Schur-Horn theorem, there exists a unitary  $U$  such that

$$\rho'_{\epsilon, n} = \mathcal{D}_{\rho'}^{\otimes n}[\chi]. \quad (11)$$

However, we have that (writing  $\bar{k} := \{1, \dots, n\} \setminus \{k\}$ )

$$\mathcal{D}_{\rho'}[\chi_k] = \text{Tr}_{\bar{k}}[(\mathcal{D}_{\rho'} \otimes 1_{\bar{k}})[\chi]] = \text{Tr}_{\bar{k}}[\mathcal{D}_{\rho'}^{\otimes n}[\chi]] = \text{Tr}_{\bar{k}}[\rho'_{\epsilon, n}]$$

by locality of quantum mechanics. But since the trace distance is nonincreasing under partial traces, we then find

$$D(\rho', \mathcal{D}_{\rho'}[\chi_k]) \leq D(\rho'^{\otimes n}, \rho'_{\epsilon, n}) \leq \epsilon. \quad (12)$$

The construction of the unitary  $U$  guaranteed by the Schur-Horn theorem is explained in the Supplemental Material [35]. The final Lemma that we require provides a way for us to get rid of unwanted coherences (arising from Lemma 5) in the final state without correlating the catalyst internally (which would spoil the catalyst).

*Lemma 6.*—(no propagation of correlations for mixed unitary channels).—Consider a mixed unitary quantum channel  $\mathcal{C}[\cdot] = \sum_i p_i V_i \cdot V_i^\dagger$  acting on a system  $S$ , where the  $p_i$  denote probabilities and the  $V_i$  are unitary operators. Dilate  $\mathcal{C}$  using an auxiliary system  $C$  and state  $\sigma = \sum_i p_i |i\rangle\langle i|$  using the unitary  $V := \sum_i V_i \otimes |i\rangle\langle i|$  as

$$\mathcal{C}[\rho] = \text{Tr}_2[V\rho \otimes \sigma V^\dagger]. \quad (13)$$

Finally, apply the dilation to a state  $\rho_{S\bar{S}}$  on  $S$  and a further system  $\bar{S}$ . Then

$$\text{Tr}_S[(V \otimes 1_{\bar{S}})\rho_{S\bar{S}} \otimes \sigma(V^\dagger \otimes 1_{\bar{S}})] = \rho_{\bar{S}} \otimes \sigma. \quad (14)$$

That is, the dilating system  $C$  is catalytic and remains uncorrelated to  $\bar{S}$ .

*Proof.*—The result immediately from the unitary invariance of the (partial) trace,

$$\text{Tr}_S[(V \otimes 1_{\bar{S}})\rho_{S\bar{S}} \otimes \sigma(V^\dagger \otimes 1_{\bar{S}})] \quad (15)$$

$$\begin{aligned} &= \sum_i p_i \text{Tr}_S[(V_i \otimes 1_{\bar{S}})\rho_{S\bar{S}}(V_i^\dagger \otimes 1_{\bar{S}})] \otimes |i\rangle\langle i| \\ &= \sum_i p_i \rho_{\bar{S}} \otimes |i\rangle\langle i| = \rho_{\bar{S}} \otimes \sigma. \end{aligned} \quad (16)$$

We are now in position to prove (ii)  $\Rightarrow$  (i) of Theorem 5. The proof proceeds in two parts. First we construct a catalyst  $\sigma_1$  for the exact transition from  $\rho$  to the equal mixture

$$\bar{\chi} := \frac{1}{n} \sum_{k=1}^n \chi_k \quad (17)$$

of the states  $\chi_k = \text{Tr}_{\bar{k}}[\chi]$ , where  $\chi$  is the state from Lemma 5. Then we use a second catalyst  $R$  in state  $\sigma_2$  to implement the dephasing map and obtain  $\mathcal{D}_{\rho'}[\bar{\chi}]$ , which is  $\epsilon$  close to the target  $\rho'$ . The part  $R$  of the catalyst thus effectively acts as a source of randomness. By Lemma 6 and the fact that the dephasing map is a mixed unitary channel, this second part can be done in such a way that the two parts of the catalyst remain uncorrelated. Therefore, the two-step process is still catalytic when both parts of the catalyst are considered as one joint catalyst in state  $\sigma_1 \otimes \sigma_2$ . As a side comment, we mention that the results of [46] imply that  $\sigma_2$  only needs to have a dimension on the order of  $\sqrt{d}$ . Furthermore, note that by perturbing  $\rho'$  arbitrarily slightly, we can always ensure that  $H(\rho) < H(\rho')$  since we allow for arbitrarily small errors and von Neumann entropy is continuous. We thus only need to prove that we can do the transition  $\rho \rightarrow_{\epsilon=0} \bar{\chi}$  in the case  $H(\rho') > H(\rho)$ . To show this, we make use of a trick that was used in recent work by Shiraishi and Sagawa [34]: We denote by  $S = S_1$  the system and by  $S_2, \dots, S_n$  and  $A$  subsystems of the first part of the catalyst. The  $S_i$  all have the same Hilbert space dimension as  $S$ , and  $A$  has Hilbert space dimension  $n$  (see Fig. 2 for an illustration of the structure of the catalyst). Then define

$$\sigma_1 = \frac{1}{n} \sum_{k=1}^n \rho^{\otimes k-1} \otimes \chi_{1, \dots, n-k} \otimes |k\rangle\langle k|_A, \quad (18)$$

where  $\chi_{1, \dots, i}$  denotes the reduced density matrix of  $\chi$  consisting of the subsystems 1 to  $i$  and we define  $\chi_0$  and  $\rho^{\otimes 0}$  to be the trivial state 1. We now apply the following sequence of unitaries on  $\rho \otimes \sigma_1$  (see Fig. 3): (1)  $U \otimes |n\rangle\langle n| + 1 \otimes \sum_{k=1}^{n-1} |k\rangle\langle k|$ , with  $U$  the unitary

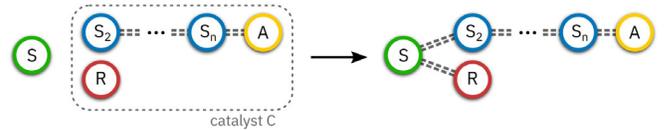


FIG. 2. The structure of the constructed catalyst  $C$ : It contains subsystems  $S_2, \dots, S_n$ , which are copies of the target system  $S$  together with an auxiliary system  $A$  of dimension  $n$  as well as a catalytic source of randomness  $R$ . The dashed lines indicate possible correlations. The source of randomness is utilized to dilate the decoherence channel  $\mathcal{D}_{\rho'}$  on  $S$  in such a way that it does not become correlated to the systems  $S_2, \dots, S_n$  and  $A$  in the process.

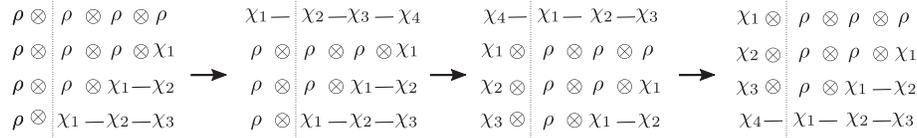


FIG. 3. Illustration of the steps in the first part of the proof for  $n = 4$ . The different columns denote the systems  $S_1, \dots, S_n$  with  $S_1 = S$  and the different rows indicate the state  $|k\rangle$  of the auxiliary system  $A$ . The hyphen indicates that the corresponding subsystems may be correlated. This procedure is essentially identical to the one in Ref. [34]. In the proof of the classical result (see Supplemental Material [35]), each of the subsystems with marginals called  $\chi_i$  here is further correlated to the second part  $R$  of the catalyst, which is not shown.

from Lemma 5, (2) the cyclic shift of subsystems  $S_i \rightarrow S_{i+1}$  with  $S_n \rightarrow S_1$ , and (3) the cyclic shift on  $A$ , acting as  $|i\rangle \rightarrow |i+1\rangle$  with  $|n+1\rangle = |1\rangle$ .

After the three steps, the catalyst is back to its initial state. The state on the system, on the other hand, is given by  $\bar{\chi}$  and, after applying the dephasing map using the system  $R$ , we find

$$D\left(\frac{1}{n} \sum_{k=1}^n \mathcal{D}_{\rho'}[\chi_k], \rho'\right) \leq \frac{1}{n} \sum_{k=1}^n D(\mathcal{D}_{\rho'}[\chi_k], \rho') \leq \epsilon$$

by the triangle inequality and Lemma 5.

*The classical case.*—As mentioned above, we can also formulate the classical version of Theorem 2. To do that, we can define a catalytic transition  $\mathbf{p} \rightarrow_{\epsilon} \mathbf{p}'$  between two probability vectors  $\mathbf{p}, \mathbf{p}' \in \mathbb{R}^d$  as in the quantum case, but with the catalysts' density matrix replaced by a further probability vector  $\mathbf{q} \in \mathbb{R}^{d^c}$  and the unitary  $U$  replaced by a permutation acting on the canonical basis vectors of  $\mathbb{R}^d \otimes \mathbb{R}^{d^c}$ . The corresponding theorem, which is fully proven in the Supplemental Material [35], can then be stated as follows:

*Theorem 7.*—Let  $\mathbf{p}, \mathbf{p}' \in \mathbb{R}^d$  be two probability vectors with Shannon entropies  $H(\mathbf{p})$  and  $H(\mathbf{p}')$ , respectively. The following are equivalent: (1) For all  $\epsilon > 0$ , we have  $\mathbf{p} \rightarrow_{\epsilon} \mathbf{p}'$ . (2)  $H(\mathbf{p}) \leq H(\mathbf{p}')$ .

Let me briefly comment on the main difference in the proof as compared to the quantum case: The essential construction of the catalyst is quite similar to the quantum case, however, clearly we cannot make use of the Schur-Horn theorem, since we do not have access to unitary operations. The proof therefore proceeds by building into the catalyst a source of randomness, which instead of being used to dephase the system, is already correlated with the first part of the catalyst  $S_2, \dots, S_n A$  from the beginning and can be used to implement the transition  $\mathbf{p}^{\otimes n} \rightarrow_{\epsilon=0} \mathbf{p}'_{\epsilon, n}$  by a random permutation in the case that the auxiliary system  $A$  is in state  $n$ . This source of randomness, in general, needs to have a dimension on the order of  $d^n$  in contrast to the quantum case, which only requires a dimension on the order of  $\sqrt{d}$ .

*Conclusion and open problems.*—We have seen that von Neumann and Shannon entropy uniquely characterize state transitions that allow for the use of a catalyst but are

otherwise reversible. There are several natural open problems left for future work: First, in Ref. [22], an exact form of the quantum catalytic entropy conjecture was conjectured, where no error is allowed in the transition from  $\rho$  to  $\rho'$  at the expense of the additional constraint that the rank of  $\rho'$  has to be at least as large as that of  $\rho$ . This form of the conjecture seems much more difficult to prove and probably requires methods that go beyond standard typicality results (which always yield asymptotic statements with vanishingly small, but finite error). A second open problem is to investigate whether a similar result also holds for other standard entropic quantities, such as mutual information or relative entropy. Finally, it will be worthwhile to explore the consequences of the given results for applications. Some further immediate applications of an affirmative resolution of the catalytic entropy conjecture in the context of (quantum) thermodynamics have been explored in Refs. [22,24], but more applications can be expected. In particular, it would be interesting to see whether there are useful applications in the context of (potentially entanglement-assisted) communication scenarios.

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*Note added.*—Recently, Refs. [47,48] have shown similar single-shot characterizations of standard entropic quantities in the context of quantum teleportation and quantum entanglement.

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- [1] C. H. Bennett, H. J. Bernstein, S. Popescu, and B. Schumacher, *Phys. Rev. A* **53**, 2046 (1996).
  - [2] C. H. Bennett, D. P. DiVincenzo, J. A. Smolin, and W. K. Wootters, *Phys. Rev. A* **54**, 3824 (1996).
  - [3] V. Vedral, M. B. Plenio, M. A. Rippin, and P. L. Knight, *Phys. Rev. Lett.* **78**, 2275 (1997).

- [4] R. Horodecki, P. Horodecki, M. Horodecki, and K. Horodecki, *Rev. Mod. Phys.* **81**, 865 (2009).
- [5] E. H. Lieb and J. Yngvason, *Phys. Rep.* **310**, 1 (1999).
- [6] D. Janzing, P. Wocjan, R. Zeier, R. Geiss, and T. Beth, *Int. J. Theor. Phys.* **39**, 2717 (2000).
- [7] M. Horodecki and J. Oppenheim, *Nat. Commun.* **4**, 2059 (2013).
- [8] F. G. S. L. Brandao, M. Horodecki, J. Oppenheim, J. M. Renes, and R. W. Spekkens, *Phys. Rev. Lett.* **111**, 250404 (2013).
- [9] F. G. S. L. Brandao, M. Horodecki, N. H. Y. Ng, J. Oppenheim, and S. Wehner, *Proc. Natl. Acad. Sci. U.S.A.* **112**, 3275 (2015).
- [10] M. Lostaglio, *Rep. Prog. Phys.* **82**, 114001 (2019).
- [11] S. D. Bartlett, T. Rudolph, and R. W. Spekkens, *Rev. Mod. Phys.* **79**, 555 (2007).
- [12] I. Marvian and R. W. Spekkens, *Phys. Rev. A* **90**, 062110 (2014).
- [13] I. Marvian and R. W. Spekkens, *Nat. Commun.* **5**, 3821 (2014).
- [14] E. Chitambar and G. Gour, *Rev. Mod. Phys.* **91**, 025001 (2019).
- [15] D. Jonathan and M. B. Plenio, *Phys. Rev. Lett.* **83**, 3566 (1999).
- [16] S. Daftuar and M. Klimesh, *Phys. Rev. A* **64**, 042314 (2001).
- [17] W. van Dam and P. Hayden, *Phys. Rev. A* **67**, 060302(R) (2003).
- [18] G. Aubrun and I. Nechita, *Commun. Math. Phys.* **278**, 133 (2008).
- [19] M. Klimesh, [arXiv:0709.3680v1](https://arxiv.org/abs/0709.3680v1).
- [20] S. Turgut, *J. Phys. A* **40**, 12185 (2007).
- [21] N. H. Y. Ng, L. Mančinska, C. Cirstoiu, J. Eisert, and S. Wehner, *New J. Phys.* **17**, 085004 (2015).
- [22] P. Boes, J. Eisert, R. Gallego, M. P. Müller, and H. Wilming, *Phys. Rev. Lett.* **122**, 210402 (2019).
- [23] C. Sparaciari, D. Jennings, and J. Oppenheim, *Nat. Commun.* **8**, 1895 (2017).
- [24] P. Boes, R. Gallego, N. H. Y. Ng, J. Eisert, and H. Wilming, *Quantum* **4**, 231 (2020).
- [25] C. E. Shannon, *Bell Syst. Tech. J.* **27**, 379 (1948).
- [26] J. A. T. Thomas M. Cover, *Elements of Information Theory* (Wiley John & Sons, New York, 2006).
- [27] M. M. Wilde, *Quantum Information Theory* (Cambridge University Press, Cambridge, England, 2009).
- [28] R. Gallego, J. Eisert, and H. Wilming, *New J. Phys.* **18**, 103017 (2016).
- [29] H. Wilming, R. Gallego, and J. Eisert, *Entropy* **19**, 241 (2017).
- [30] M. Lostaglio, M. P. Müller, and M. Pastena, *Phys. Rev. Lett.* **115**, 150402 (2015).
- [31] M. P. Müller and M. Pastena, *IEEE Trans. Inf. Theory* **62**, 1711 (2016).
- [32] M. P. Müller, *Phys. Rev. X* **8**, 041051 (2018).
- [33] S. Rethinasamy and M. M. Wilde, *Phys. Rev. Research* **2**, 033455 (2020).
- [34] N. Shiraishi and T. Sagawa, *Phys. Rev. Lett.* **126**, 150502 (2021).
- [35] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevLett.127.260402> for a proof sketch of Lemma 4, a detailed discussion of how to construct the unitary  $U$ , the formulation and proof of the classical version of the main result, and a discussion of the dimension required for the catalyst, which includes Refs. [36,37].
- [36] P. Boes, N. H. Y. Ng, and H. Wilming, [arXiv:2009.08391](https://arxiv.org/abs/2009.08391).
- [37] A. W. Marshall, I. Olkin, and B. C. Arnold, *Inequalities: Theory of Majorization and Its Applications* (Springer, New York, 2011).
- [38] M. Fannes, *Commun. Math. Phys.* **31**, 291 (1973).
- [39] K. M. R. Audenaert, *J. Phys. A* **40**, 8127 (2007).
- [40] P. Kammerlander and R. Renner, [arXiv:2002.08968](https://arxiv.org/abs/2002.08968).
- [41] R. Alicki and M. Fannes, *Phys. Rev. E* **87**, 042123 (2013).
- [42] W. Pusz and S. Woronowicz, *Commun. Math. Phys.* **58**, 273 (1978).
- [43] A. Lenard, *J. Stat. Phys.* **19**, 575 (1978).
- [44] In [23] it was argued that by concatenating arbitrarily many catalytic transitions the bound in Corollary 3 can be achieved. However, concatenating two catalytic transitions builds up correlations between the catalysts and thus is, in general, not catalytic as a whole. Our main result shows that one can nevertheless always find a catalytic transformation implementing the two steps in one.
- [45] T. Holenstein and R. Renner, *IEEE Trans. Inf. Theory* **57**, 1865 (2011).
- [46] P. Boes, H. Wilming, R. Gallego, and J. Eisert, *Phys. Rev. X* **8**, 041016 (2018).
- [47] T. V. Kondra, C. Datta, and A. Streltsov, *Phys. Rev. Lett.* **127**, 150503 (2021).
- [48] P. Lipka-Bartosik and P. Skrzypczyk, *Phys. Rev. Lett.* **127**, 080502 (2021).