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Letter

No-go theorem for entanglement distillation using catalysis

Ludovico Lami [*]
QuSoft, Science Park 123, 1098 XG Amsterdam, the Netherlands;
Korteweg–de Vries Institute for Mathematics, University of Amsterdam, Science Park 105-107, 1098 XG Amsterdam, the Netherlands;
and Institute for Theoretical Physics, University of Amsterdam, Science Park 904, 1098 XH Amsterdam, the Netherlands

Bartosz Regula
Mathematical Quantum Information RIKEN Hakubi Research Team, RIKEN Cluster for Pioneering Research (CPR)
and RIKEN Center for Quantum Computing (RQC), Wako, Saitama 351-0198, Japan

Alexander Streltsov
Institute of Fundamental Technological Research, Polish Academy of Sciences, Pawińskiego 5B, 02-106 Warsaw, Poland
and Centre for Quantum Optical Technologies, Centre of New Technologies, University of Warsaw, Banacha 2c, 02-097 Warsaw, Poland

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The use of ancillary quantum systems known as catalysts is known to be able to enhance the capabilities of entanglement transformations under local operations and classical communication. However, the limits of these advantages have not been determined and in particular it is not known if such assistance can overcome the known restrictions on asymptotic transformation rates—notably the existence of bound entangled (undistillable) states. Here we establish a general limitation on entanglement catalysis: we show that catalytic transformations can never allow for the distillation of entanglement from a bound entangled state with positive partial transpose, even if the catalyst may become correlated with the system of interest and even under permissive choices of free operations. This precludes the possibility that catalysis may make entanglement theory asymptotically reversible. Our methods are based on asymptotic bounds for the distillable entanglement and entanglement cost assisted by correlated catalysts.

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Introduction. The study of quantum entanglement as a resource has been one of the most fundamental problems in the field of quantum information ever since its inception [1]. To utilize this resource efficiently, it is often required to transform and manipulate entangled quantum systems, which leads to the well-studied question of how quantum states can be converted using only local operations and classical communication (LOCC) [2,3]. The limits of such conversion capability are represented by asymptotic transformation rates: given many copies of an input quantum state ρ, how many copies of a desired target state can we obtain per each copy of ρ? Such rates are particularly important in the context of purifying noisy quantum states into singlets Φ_2, a task known as entanglement distillation, as well as for the reverse task of using such singlets to synthesize noisy quantum states. This leads to the notions of *distillable entanglement* $E_d(\rho)$ [2], which tells us how many copies of Φ_2 we can extract from a given state ρ, and of *entanglement cost* $E_c(\rho)$ [3], which tells us how many copies of a pure singlet are needed to produce ρ.

A phenomenon that can severely restrict our ability to extract entanglement is known as *bound entanglement* [4]: there exist states from which no entanglement can be distilled, even though their entanglement cost is nonzero. A consequence of this is the irreversibility of entanglement theory—after performing a transformation $\rho \to \omega$, one may not be able to realize the reverse process $\omega \to \rho$ and recover all of the supplied copies of ρ. This contrasts with the asymptotic reversibility of theories such as classical and quantum thermodynamics [4–6]. Although reversibility may still hold in some restricted cases (e.g., for all bipartite pure quantum states [3,5]), and there are even approaches that may enable reversibility by suitably relaxing the restrictions on the allowed physical transformations [7–10], irreversibility is often a fundamental property of the theory of quantum entanglement that may not be easily evaded [11]. It is then important to understand how, if at all, irreversibility can be overcome.

A promising approach to increase the capabilities of entanglement transformations is the use of so-called *catalysts* [12], that is, ancillary systems that can be employed in the conversion protocol, but must eventually be returned in an unchanged state. Although this phenomenon has been shown to be remarkably powerful in the context of single- and many-copy transformations [12–19], it is unknown whether catalysis can enhance asymptotic conversion rates. This motivates in particular an important question: is the use of catalysis enough to facilitate the reversibility of entanglement theory?

In this paper, we close this question by showing that even very permissive forms of catalytic transformations are

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[*] ludovico.lami@gmail.com

insufficient to distill entanglement from bound entangled states. Specifically, we consider the representative class of bound entangled states known as positive partial transpose (PPT) states and we show that the catalytically distillable entanglement of any such state is zero, which is strictly less than its catalytic entanglement cost. The result relies on the establishment of a general upper bound on distillable entanglement under catalytic LOCC operations, namely, the relative entropy of PPT entanglement, which was known to be an upper bound only in conventional, noncatalytic protocols [20,21]. We show that this limitation persists even if one allows the catalyst to build up correlations with the main system, as well as if one allows sets of operations larger than LOCC, in particular all PPT-preserving transformations. This presents a very general limitation on the power of catalytic transformations of entangled states. We additionally study the applications of resource monotones to constraining asymptotic state conversion with catalytic assistance, obtaining a number of bounds that may be of independent interest.

Preliminaries. We use SEP$(A\!:\!B)$ to denote the set of states σ_{AB}, which are separable across the bipartition $A\!:\!B$. The notation PPT$(A\!:\!B)$ will be used to denote the set of positive partial transpose states, i.e., ones for which the partially transposed operator σ_{AB}^{Γ} is also a valid quantum state. States which are not in PPT will be conventionally called NPT (nonpositive partial transpose).

Even though the choice of LOCC in the context of entanglement transformations is well motivated from a practical perspective, in many settings there exist other possible choices of allowed "free" operations; let us then use $\mathcal{F}$ to denote the chosen set of such permitted protocols. One such choice is the set of so-called PPT operations [22] or the even larger set of all PPT-preserving operations PPTP [23], comprising all maps $\Lambda : AB \to A'B'$ such that $\Lambda(\sigma_{AB}) \in \mathrm{PPT}(A'\!:\!B')$ for all $\sigma_{AB} \in \mathrm{PPT}(A\!:\!B)$. The latter is one of the largest and most permissive sets considered in the study of operational entanglement transformations.

Given two bipartite states ρ_{AB} and $\omega_{A'B'}$, we say that the transformation from ρ_{AB} to $\omega_{A'B'}$ is possible via operations in $\mathcal{F}$ assisted by catalysts if there exists a finite-dimensional state τ_{CD} and an operation $\Lambda \in \mathcal{F}(AC : BD \to A'C : B'D)$ such that

$$\Lambda(\rho_{AB} \otimes \tau_{CD}) = \omega_{A'B'} \otimes \tau_{CD}. \tag{1}$$

We denote this by $\rho_{AB} \xrightarrow{\mathcal{F}^c} \omega_{A'B'}$. More generally, we say that the transformation is possible via operations in $\mathcal{F}$ assisted by *correlated catalysts* [24–26] and we write $\rho_{AB} \xrightarrow{\mathcal{F}^{cc}} \omega_{A'B'}$, if there exists a finite-dimensional state τ_{CD} and an operation $\Lambda \in \mathcal{F}(AC : BD \to A'C : B'D)$ such that

$$\mathrm{Tr}_{CD}\Lambda(\rho_{AB} \otimes \tau_{CD}) = \omega_{A'B'} \tag{2}$$

and

$$\mathrm{Tr}_{A'B'}\Lambda(\rho_{AB} \otimes \tau_{CD}) = \tau_{CD}. \tag{3}$$

This relaxed notion allows for the output state of the protocol to exhibit correlations between the main system $(A'B')$ and catalyst (CD), as long as the marginal systems satisfy the required constraints. Crucially, correlated catalysis is a strictly more powerful framework than standard catalysis and allowing for such correlations can greatly enlarge the set of achievable state transformations already in the single-shot regime [16,17,19,25–29].

Given any allowed choice of transformations $\widetilde{\mathcal{F}} \in \{\mathcal{F}, \mathcal{F}^c, \mathcal{F}^{cc}\}$, we write $\rho_{AB} \xrightarrow{\widetilde{\mathcal{F}}} \approx_\varepsilon \omega_{A'B'}$ if there exists a state $\omega'_{A'B'}$ such that the transformation is realizable up to some small error ε, which we quantify with the trace distance:

$$\rho_{AB} \xrightarrow{\widetilde{\mathcal{F}}} \omega'_{A'B'}, \qquad \tfrac{1}{2}\left\| \omega'_{A'B'} - \omega_{A'B'} \right\|_1 \leqslant \varepsilon. \tag{4}$$

A conceptually simplified picture can be obtained by looking at the *ultimate* limitations to which the above transformation is subjected. This intuition can be formalized by investigating the conversion of a large number n of copies of ρ_{AB} into as many copies as possible of $\omega_{A'B'}$, with the transformation error vanishing with growing n. The relevant figure of merit is then the transformation rate, i.e., the ratio between the number of output copies and the number of input copies (that is, n). Mathematically, this can be defined by

$$\begin{aligned} &R_{\widetilde{\mathcal{F}}}(\rho_{AB} \to \omega_{A'B'}) \\ &\quad := \sup\left\{R : \ \rho_{AB}^{\otimes n} \xrightarrow{\widetilde{\mathcal{F}}} \approx_{\varepsilon_n} \omega_{A'B'}^{\otimes \lceil Rn \rceil}, \quad \lim_{n\to\infty} \varepsilon_n = 0\right\}. \end{aligned} \tag{5}$$

The distillable entanglement and entanglement cost under operations in $\widetilde{\mathcal{F}}$ are then defined by

$$E_{d,\widetilde{\mathcal{F}}}(\rho) := R_{\widetilde{\mathcal{F}}}(\rho \to \Phi_2), \quad E_{c,\widetilde{\mathcal{F}}}(\rho) := \frac{1}{R_{\widetilde{\mathcal{F}}}(\Phi_2 \to \rho)}, \tag{6}$$

where $\Phi_2 := |\Phi_2\rangle\langle\Phi_2|$ denotes the maximally entangled two-qubit state, $|\Phi_2\rangle = \frac{1}{\sqrt{2}}(|00\rangle + |11\rangle)$. Conventionally, the notation E_d and E_c is used to refer to $E_{d,\,\mathrm{LOCC}}$ and $E_{c,\,\mathrm{LOCC}}$.

An entangled state σ_{AB} is called bound entangled if $E_{d,\mathrm{LOCC}}(\sigma_{AB}) = 0$. A particularly useful criterion to detect undistillability was established in [4]: if a state σ_{AB} is PPT, then $E_{d,\mathrm{LOCC}}(\sigma_{AB}) = 0$. As $E_{c,\mathrm{LOCC}}(\sigma_{AB}) > 0$ for any entangled state σ_{AB} [30], this means that any PPT σ_{AB} which is not separable has a nonzero entanglement cost, while no entanglement can be extracted from it. Interestingly, it is still an open question whether *every* bound entangled state is PPT [31,32].

Monotones. A very common way to constrain entanglement transformations, also in the asymptotic transformation regime, is to use so-called entanglement monotones, also known as entanglement measures [33]. These are functions M which satisfy $M[\Lambda(\rho_{AB})] \leqslant M(\rho_{AB})$ for all free operations $\Lambda \in \mathcal{F}$. It is well known that, if the monotone satisfies weak additivity, i.e., $M(\rho^{\otimes n}) = nM(\rho)$, as well as a stronger form of continuity known as asymptotic continuity [34,35], then the (noncatalytic) transformation rate is bounded as [23,36]

$$R_{\mathcal{F}}(\rho \to \omega) \leqslant \frac{M(\rho)}{M(\omega)}. \tag{7}$$

Monotones are typically chosen so that they are normalized on the maximally entangled state, i.e., $M(\Phi_2) = 1$. Any such monotone then satisfies [37]

$$E_{d,\mathcal{F}}(\rho) \leqslant M(\rho) \leqslant E_{c,\mathcal{F}}(\rho). \tag{8}$$

A particularly important example of an LOCC monotone is the relative entropy of (NPT) entanglement [33]

$$D_{\rm PPT}(\rho) := \min_{\sigma\in{\rm PPT}(A:B)} D(\rho\|\sigma), \tag{9}$$

with the quantum relative entropy defined by $D(\omega\|\tau) := {\rm Tr}\,\omega(\log_2\omega - \log_2\tau)$ [38,39]. However, this measure is not additive: it is merely *subadditive*, in the sense that

$$D_{\rm PPT}(\rho_{A:B}\otimes\omega_{A':B'}) \leqslant D_{\rm PPT}(\rho_{A:B}) + D_{\rm PPT}(\omega_{A':B'}), \tag{10}$$

and the inequality may be strict for some states [40]. This issue is circumvented through the procedure of *regularization*, which considers the asymptotic limit

$$D^{\infty}_{\rm PPT}(\rho) := \lim_{n\to\infty}\frac{1}{n}D_{\rm PPT}(\rho^{\otimes n}). \tag{11}$$

The resulting regularized relative entropy of entanglement is weakly additive [36] and constitutes one of the most fundamental and commonly used bounds on entanglement transformation rates: in particular, $E_{d,{\rm LOCC}}(\rho) \leqslant D^{\infty}_{\rm PPT}(\rho)$.

The situation is much more intricate when it comes to catalytic transformations [17,41,42]. To establish a similar bound, it appears that several more assumptions about the given monotone are needed. In particular, if we also assume full additivity [i.e., $M(\rho_{AB}\otimes\omega_{A'B'}) = M(\rho_{AB}) + M(\omega_{A'B'})$ for any $\rho_{AB}, \omega_{A'B'}$] and strong superadditivity [i.e., $M(\rho_{AA':BB'}) \geqslant M(\rho_{AB}) + M(\rho_{A'B'})$], then we analogously obtain

$$R_{\mathcal{F}^{cc}}(\rho\to\omega) \leqslant \frac{M(\rho)}{M(\omega)} \tag{12}$$

(see [43] for a proof). However, to date, there are only two LOCC monotones that are known to satisfy all the required assumptions: the squashed entanglement $E_{\rm sq}$ [44] and the conditional entanglement of mutual information E_I [45,46], both of which are however typically difficult to evaluate or even estimate.

Importantly, as the regularized relative entropy $D^{\infty}_{\rm PPT}$ is not known to satisfy the above required properties, we do not yet know whether it is monotone under asymptotic correlated–catalytic protocols. This entails that we cannot straightforwardly use it to bound $E_{d,{\rm LOCC}^{cc}}$ or $E_{c,{\rm LOCC}^{cc}}$ and thus to alleviate the issue of the lack of readily computable bounds on transformation rates. Any attempt to derive such asymptotic bounds on transformations with correlated catalysts therefore requires a completely different approach than conventional, noncatalytic bounds.

Results. Our main technical contribution is the establishment of very general bounds on correlated catalytic transformation rates and in particular the recovery of the regularized relative entropy as an upper bound on the rate of distillation:

$$E_{d,\,{\rm LOCC}^{cc}}(\rho_{AB}) \leqslant D^{\infty}_{\rm PPT}(\rho_{AB}) \leqslant D_{\rm PPT}(\rho_{AB}). \tag{13}$$

As $D_{\rm PPT}(\rho_{AB}) = 0$ for any PPT state, a key consequence of this result is the fact that correlated catalysis is not sufficient to break the fundamental irreversibility of PPT bound entangled states.

Theorem 1. All PPT entangled states ρ_{AB} are bound entangled under LOCC operations assisted by correlated catalysts, but have nonzero cost. More formally, if ρ_{AB} is PPT entangled, then

$$E_{d,\,{\rm LOCC}^{cc}}(\rho_{AB}) = 0 < E_{c,\,{\rm LOCC}^{cc}}(\rho_{AB}). \tag{14}$$

Consequently, entanglement theory is irreversible even under LOCC assisted by correlated catalysts.

The result can be strengthened in several ways. First, our methods immediately apply also to PPT-preserving operations assisted by correlated catalysis, showing that even under such extended transformations no entanglement can be extracted from PPT states: $E_{d,\,{\rm PPTP}^{cc}}(\rho_{AB}) = 0$.

Even more strongly, we can show that a PPT state can never be converted to an NPT state by means of PPT-preserving operations assisted by correlated catalysts, including all catalytic LOCC protocols. This implies in particular that not only is the rate of distillation equal to zero, but not even a single copy of Φ_2 can be distilled with error $\varepsilon < 1/2$, no matter how many copies of a given PPT entangled state are at our disposal.

This conclusively shows that bound entanglement, and thus the irreversibility of entanglement theory, cannot be circumvented or even alleviated through the use of catalysts.

Let us remark here that a different notion of "catalytic irreversibility" was previously considered in the seminal work of Vidal and Cirac [47]. However, the transformations considered there are much more restricted than the ones allowed in our approach—indeed, they are not truly "catalytic" in the sense that the preservation of the assisting ancillary system is not actually required, and furthermore no correlations are permitted between the main and the ancillary systems. Our setting is thus strictly more general than that of [47] and as far as we know it is not possible to retrieve our findings on catalytic bound entanglement using results from [47] only.

Proof idea. A crucial ingredient in our proofs is the *measured relative entropy of entanglement* $D^{\mathbb{PPT}}_{\rm PPT}$, which belongs to a family of entanglement measures first studied by Piani in a pioneering work [48]. It is defined as

$$D^{\mathbb{PPT}}_{\rm PPT}(\rho) := \inf_{\sigma\in{\rm PPT}}\ \sup_{\mathcal{M}\in PPT} D(\mathcal{M}(\rho)\|\mathcal{M}(\sigma)), \tag{15}$$

where $\mathbb{PPT}$ denotes the set of PPT measurements—that is, POVMs $\{M_i\}_i$ such that each operator M_i is PPT and $\mathcal{M}(\rho) = \sum_i {\rm Tr}[M_i\,\rho]\,|i\rangle\!\langle i|$ is the corresponding measurement channel. The difference between this quantity and the relative entropy of entanglement $D_{\rm PPT}$ is that the relative entropy is evaluated not between quantum states, but rather the probability distributions constituted by the measurement outcomes.

While (15) seemingly adds a further layer of complication to (10), it is in many respects a more natural and well-behaved quantity. Most importantly for us, the measured relative entropy satisfies strong superadditivity and in fact it allows for the establishment of a superadditivitylike relation for the relative entropy of entanglement $D_{\rm PPT}$ itself: it holds that [48]

$$D_{\rm PPT}(\rho_{AA':BB'}) \geqslant D_{\rm PPT}(\rho_{A:B}) + D^{\mathbb{PPT}}_{\rm PPT}(\rho_{A':B'}). \tag{16}$$

This remarkable relation allows us to avoid having to rely solely on the properties of $D_{\rm PPT}$, which—as we discussed before—are not sufficient to use this quantity in the catalytic setting.

Let us then derive the upper bound on catalytic distillable entanglement announced in Eq. (13).

Assume that R is an achievable rate for entanglement distillation under operations in PPTP$^{\mathrm{cc}}$; that is, that there exists a sequence of catalysts $\tau_n = (\tau_n)_{C_nD_n}$ on the finite-dimensional systems C_nD_n and a sequence of operations $\Lambda_n \in \mathrm{PPTP}(\mathrm{A^nC_n : B^nD_n \to A_0^{\lceil Rn\rceil}C_n : B_0^{\lceil Rn\rceil}D_n})$, with A_0 and B_0 being single-qubit systems, such that

$$\begin{aligned}
\varepsilon_n &:= \tfrac{1}{2}\left\| \mathrm{Tr}_{C_nD_n} \Lambda_n\big(\rho_{AB}^{\otimes n} \otimes \tau_n\big) - \Phi_2^{\otimes \lceil Rn\rceil} \right\|_1 \xrightarrow[n\to\infty]{} 0,\\
\tau_n &= \mathrm{Tr}_{A_0^{\lceil Rn\rceil}B_0^{\lceil Rn\rceil}}\big[\Lambda_n\big(\rho_{AB}^{\otimes n} \otimes \tau_n\big)\big].
\end{aligned} \tag{17}$$

Then

$$\begin{aligned}
&n\, D_{\mathrm{PPT}}(\rho_{AB}) + D_{\mathrm{PPT}}(\tau_n)\\
&\overset{\text{(i)}}{\geqslant} D_{\mathrm{PPT}}\big(\rho_{AB}^{\otimes n}\big) + D_{\mathrm{PPT}}(\tau_n)\\
&\overset{\text{(ii)}}{\geqslant} D_{\mathrm{PPT}}\big(\rho_{AB}^{\otimes n} \otimes \tau_n\big)\\
&\overset{\text{(iii)}}{\geqslant} D_{\mathrm{PPT}}\big(\Lambda_n\big(\rho_{AB}^{\otimes n} \otimes \tau_n\big)\big)\\
&\overset{\text{(iv)}}{\geqslant} D_{\mathrm{PPT}}^{\mathbb{PPT}}\big(\mathrm{Tr}_{\mathrm{C_nD_n}} \Lambda_\mathrm{n}\big(\rho_{\mathrm{AB}}^{\otimes \mathrm{n}} \otimes \tau_\mathrm{n}\big)\big)\\
&\quad + D_{\mathrm{PPT}}\big(\mathrm{Tr}_{\mathrm{A_0^{\lceil Rn\rceil}B_0^{\lceil Rn\rceil}}} \Lambda_\mathrm{n}\big(\rho_{\mathrm{AB}}^{\otimes \mathrm{n}} \otimes \tau_\mathrm{n}\big)\big)\\
&= D_{\mathrm{PPT}}^{\mathbb{PPT}}\big(\mathrm{Tr}_{\mathrm{C_nD_n}} \Lambda_\mathrm{n}\big(\rho_{\mathrm{AB}}^{\otimes \mathrm{n}} \otimes \tau_\mathrm{n}\big)\big) + \mathrm{D_{PPT}}(\tau_\mathrm{n})\\
&\overset{\text{(v)}}{\geqslant} D_{\mathrm{PPT}}^{\mathbb{PPT}}\big(\Phi_2^{\otimes \lceil Rn\rceil}\big) - \varepsilon_n \lceil Rn\rceil - g(\varepsilon_n) + D_{\mathrm{PPT}}(\tau_n)\\
&\overset{\text{(vi)}}{\geqslant} \lceil Rn\rceil - 1 - \varepsilon_n\lceil Rn\rceil - g(\varepsilon_n) + D_{\mathrm{PPT}}(\tau_n).
\end{aligned} \tag{18}$$

Here (i) and (ii) are applications of the tensor subadditivity of D_{PPT} [20]; (iii) comes from its monotonicity under PPT-preserving operations; (iv) descends from Piani's superadditivitylike inequality in (16); (v) follows from asymptotic continuity [49,50], which states that [43]

$$\left| D_{\mathrm{PPT}}^{\mathbb{PPT}}(\rho_{AB}) - D_{\mathrm{PPT}}^{\mathbb{PPT}}(\omega_{AB}) \right| \leqslant \varepsilon \log_2 d + g(\varepsilon) \tag{19}$$

holds for all pairs of states ρ_{AB}, ω_{AB} at trace distance $\varepsilon := \frac{1}{2}\|\rho_{AB} - \omega_{AB}\|_1$ on all systems AB of minimal local dimension $d := \min\{|A|, |B|\}$, where $g(x) := (1+x)\log_2(1+x) - x\log_2 x$; and finally (vi) is a consequence of the quasinormalization of $D_{\mathrm{PPT}}^{\mathbb{PPT}}$, i.e., the fact that $D_{\mathrm{PPT}}^{\mathbb{PPT}}(\Phi_2^{\otimes k}) \geqslant \log_2(2^k + 1) - 1$ [49]. The above chain of inequalities shows that

$$D_{\mathrm{PPT}}\big(\rho_{AB}^{\otimes n}\big) \geqslant (1 - \varepsilon_n)\lceil Rn\rceil - 1 - g(\varepsilon_n), \tag{20}$$

and by dividing by n, taking the limit as $n \to \infty$, and subsequently the supremum over all achievable rates R, we obtain the claimed result.

To complete the proof of Theorem 1, it suffices to use the fact that the squashed entanglement E_{sq}—which, as we remarked before and discuss in more detail in [43], lower bounds the correlated-catalytic entanglement cost—is nonzero for any entangled state [49,51–54]. An approach very similar to the above can be used to derive a corresponding lower bound on the entanglement cost under PPT-preserving operations assisted by catalysts, leveraging once again Piani's superadditivity relation (16). The chain of inequalities in this case reads

$$\begin{aligned}
&\lceil Rn\rceil + D_{\mathrm{PPT}}(\tau_n) && (21)\\
&\quad = D_{\mathrm{PPT}}\big(\Phi_2^{\otimes\lceil Rn\rceil}\big) + D_{\mathrm{PPT}}(\tau_n)\\
&\quad \geqslant D_{\mathrm{PPT}}\big(\Phi_2^{\otimes\lceil Rn\rceil} \otimes \tau_n\big)\\
&\quad \geqslant D_{\mathrm{PPT}}\big(\Lambda_n\big(\Phi_2^{\otimes\lceil Rn\rceil} \otimes \tau_n\big)\big)\\
&\quad \geqslant D_{\mathrm{PPT}}^{\mathbb{PPT}}\big(\mathrm{Tr}_{\mathrm{C_nD_n}} \Lambda_\mathrm{n}\big(\Phi_2^{\otimes\lceil \mathrm{Rn}\rceil} \otimes \tau_\mathrm{n}\big)\big) && (22)\\
&\qquad + D_{\mathrm{PPT}}\big(\mathrm{Tr}_{\mathrm{A_0^{\lceil Rn\rceil}B_0^{\lceil Rn\rceil}}} \Lambda_\mathrm{n}\big(\Phi_2^{\otimes\lceil \mathrm{Rn}\rceil} \otimes \tau_\mathrm{n}\big)\big)\\
&\quad = D_{\mathrm{PPT}}^{\mathbb{PPT}}\big(\mathrm{Tr}_{\mathrm{C_nD_n}} \Lambda_\mathrm{n}\big(\Phi_2^{\otimes\lceil \mathrm{Rn}\rceil} \otimes \tau_\mathrm{n}\big)\big) + \mathrm{D_{PPT}}(\tau_\mathrm{n})\\
&\quad \geqslant D_{\mathrm{PPT}}^{\mathbb{PPT}}\big(\rho_{AB}^{\otimes n}\big) - \varepsilon_n \ln(d^n) - g(\varepsilon_n) + D_{\mathrm{PPT}}(\tau_n). && (23)
\end{aligned}$$

Eliminating $D_{\mathrm{PPT}}(\tau_n)$ on both sides, dividing by n, and taking the limit $n \to \infty$ shows that any rate of dilution R is lower bounded by the regularization of $D_{\mathrm{PPT}}^{\mathbb{PPT}}$. Our technical contributions derived above can be summarized as follows.

Proposition 2. For all states ρ_{AB}, the distillable entanglement and the entanglement cost under PPT-preserving operations assisted by correlated catalysts satisfy that

$$E_{d,\,\mathrm{PPTP^{cc}}}(\rho_{AB}) \leqslant D_{\mathrm{PPT}}^{\infty}(\rho_{AB}) \leqslant D_{\mathrm{PPT}}(\rho_{AB}) \tag{24}$$

and

$$E_{c,\,\mathrm{PPTP^{cc}}}(\rho_{AB}) \geqslant D_{\mathrm{PPT}}^{\mathbb{PPT},\infty}(\rho_{AB}) \geqslant D_{\mathrm{PPT}}^{\mathbb{PPT}}(\rho_{AB}). \tag{25}$$

This gives two very general limitations on asymptotic transformation rates with correlated-catalytic assistance, notably ones that can be efficiently computed or bounded as they do not require regularization. For example, a consequence of Proposition 2 coupled with the faithfulness of $D_{\mathrm{PPT}}^{\mathbb{PPT}}$ [55] is that the entanglement cost of any NPT entangled state is nonzero, even under PPT-preserving operations assisted by correlated catalysis. A peculiarity of the bounds in (24) and (25) is that they do not immediately imply that $E_{d,\,\mathcal{F}^{cc}}(\rho_{AB}) \overset{?}{\leqslant} E_{c,\,\mathcal{F}^{cc}}(\rho_{AB})$ for the class of operations $\mathcal{F} = \mathrm{PPTP}$. Such an inequality is central to the logical consistency of the theory, because it tells us that no net entanglement can be generated in a cycle of dilution and distillation of ρ_{AB}, forbidding the existence of a "perpetuum mobile" in entanglement theory [5]. This is essentially a technicality stemming from our use of PPTP$^{\mathrm{cc}}$ operations. For the more operationally grounded classes of free operations $\mathcal{F} = \mathrm{LOCC}, \mathrm{PPT}$, the inequality can be shown (cf. [56]), and we include a complete proof of this that works for all PPT or distillable states when $\mathcal{F} = \mathrm{LOCC}$ and for all states when $\mathcal{F} = \mathrm{PPT}$. Readers interested in a more detailed exposition of the properties of catalytic entanglement monotones are encouraged to consult the Supplemental Material [43]. Therein, we also consider slightly strengthened and generalized variants of the result of Theorem 1.

Extension to quantum coherence. Quantum coherence is another important example of a quantum resource and interestingly it shares many similarities with entanglement theory [57–60]. In this context, incoherent operations (IO) [58,59] have emerged as the main example of a set of operations that are sufficiently powerful to allow for generic coherence distillation, yet not powerful enough to enable full reversibility. It is natural to ask whether one could improve either distillation or dilution under IO via catalysis. Extending our approach from entanglement theory, we can answer this question in the negative in the most general sense: neither the IO distillable coherence nor the IO coherence cost of any state are affected by the introduction of catalysts. As this is beyond the scope of the entanglement-focused discussion in the paper, a thorough consideration of this setting can be found in the Supplemental Material [43].

Discussion. We have established general limitations on asymptotic entanglement transformation rates with correlated catalysis, precluding the possibility of using catalysis to distill entanglement from PPT states.

Although our methods lead to robust and general constraints on the power of catalytic conversion protocols, there are still many open questions in this context. In particular, complementing the no-go results obtained here, is there *any* transformation whose rate can be improved by allowing correlated catalysts? Furthermore, since we have shown that correlated catalysis on its own is not enough to enable the reversibility of entanglement theory, the big open question [54,61] remains: what does it take to achieve reversible entanglement transformations? One context in which our approach is not able to rule out reversibility is the use of nonentangling protocols [8,11] with catalytic assistance, making it an interesting question to investigate such a possibility.

Note added. Recently, a complementary question has been independently studied in [62]: given that an entangled state ρ is distillable, i.e., $E_{d,\mathrm{LOCC}}(\rho) > 0$, can its distillable entanglement be increased by catalytic LOCC protocols? The question is answered in the negative using different methods. Since the results of [62] apply only to distillable states, they cannot be used to derive the results presented in this manuscript.

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