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No second law of entanglement manipulation after all

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No second law of entanglement manipulation after all
— Supplementary Information —

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CONTENTS

I. GENERAL DEFINITIONS

Throughout the Supplementary Information, we work in the full generality of Hilbert spaces which can be infinite dimensional. An operator $X : \mathcal{H} \to \mathcal{H}$ on a Hilbert space X is said to be bounded if $\|X\|_\infty := \sup_{|\psi\rangle\in\mathcal{H},\, \||\psi\rangle\|\leq 1} \|X\,|\psi\rangle\| < \infty$. The expression on the left-hand side is called the operator norm of X. The Banach space of bounded operators on a Hilbert space $\mathcal{H}$ will be denoted with $\mathcal{B}(\mathcal{H})$. Its pre-dual is the Banach space of trace class operators, denoted with $\mathcal{T}(\mathcal{H})$. We remind the reader that an operator $T : \mathcal{H} \to \mathcal{H}$ is said to be of trace class if it is bounded and moreover $\sum_{j=0}^{\infty} \langle e_j|\sqrt{T^\dagger T}|e_j\rangle < \infty$ converges for some — and hence for all — orthonormal bases $\{|e_j\rangle\}_{j\in\mathbb{N}}$. Here, $\sqrt{T^\dagger T}$ is the unique positive square root of the positive semi-definite bounded operator $T^\dagger T$, with $T^\dagger$ being the adjoint of T. Positive semi-definite trace-class operators inside

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$\mathcal{T}(\mathcal{H})$ form a cone, indicated with $\mathcal{T}_+(\mathcal{H})$. When normalised to have trace equal to one, operators in $\mathcal{T}_+(\mathcal{H})$ form the set of density operators, which we will denote by $\mathcal{D}(\mathcal{H})$. Hereafter, the subscript sa (e.g. $\mathcal{B}_{\text{sa}}$) indicates a restriction to self-adjoint operators.

A linear map $\Lambda : \mathcal{T}(\mathcal{H}_A) \to \mathcal{T}(\mathcal{H}_B)$, i.e. from system A to system B, is said to be:

(i) positive, if $\Lambda\left(\mathcal{T}_+(\mathcal{H}_A)\right) \subseteq \mathcal{T}_+(\mathcal{H}_B))$;

(ii) completely positive, if $\mathrm{id}_k \otimes \Lambda : \mathcal{T}\left(\mathbb{C}^k \otimes \mathcal{H}_A\right) \to \mathcal{T}\left(\mathbb{C}^k \otimes \mathcal{H}_B\right)$ is positive for all $k \in \mathbb{N}$, where id_k denotes the identity channel on the space of $k \times k$ complex matrices;

(iii) trace preserving, if $\operatorname{Tr} \Lambda(X) = \operatorname{Tr} X$ for all X.

We will denote the set of positive (respectively, completely positive) trace preserving maps from A to B with $\mathrm{PTP}_{A\to B}$ (respectively, $\mathrm{CPTP}_{A\to B}$). Given a bounded linear map $\Lambda : \mathcal{T}(\mathcal{H}_A) \to \mathcal{T}(\mathcal{H}_B)$,[1] we can consider its adjoint $\Lambda^\dagger : \mathcal{B}(\mathcal{H}_B) \to \mathcal{B}(\mathcal{H}_A)$, defined by the identity

$$\operatorname{Tr}\left[X\Lambda^\dagger(Y)\right] = \operatorname{Tr}\left[\Lambda(X)Y\right], \qquad \forall\, X \in \mathcal{T}(\mathcal{H}_A), \quad \forall\, Y \in \mathcal{B}(\mathcal{H}_B) \tag{S1}$$

If $\Lambda : \mathcal{T}(\mathcal{H}_A) \to \mathcal{T}(\mathcal{H}_B)$ is a positive and trace preserving linear map, then it satisfies that

$$\|\Lambda(T)\|_1 \leq \|T\|_1 \qquad \forall\, T \in \mathcal{T}(\mathcal{H}). \tag{S2}$$

This in particular implies that Λ is bounded in the Banach space sense. Its adjoint $\Lambda^\dagger$ is positive and unital, meaning that $\Lambda^\dagger(\mathbb{1}_B) = \mathbb{1}_A$, and more generally

$$\left\|\Lambda^\dagger(X)\right\|_\infty \leq \|X\|_\infty \qquad \forall\, X \in \mathcal{B}(\mathcal{H}). \tag{S3}$$

A. Robustness measures

Let AB be a quantum system with Hilbert space $\mathcal{H}_{AB} \coloneqq \mathcal{H}_A \otimes \mathcal{H}_B$. The set of separable states on AB can be defined as the closed convex hull of all product states, in formula

$$\mathcal{S}^1_{AB} \coloneqq \mathrm{cl}\left(\mathrm{conv}\left\{|\psi\rangle\!\langle\psi|_A \otimes |\phi\rangle\!\langle\phi|_B :\ |\psi\rangle_A \in \mathcal{H}_A,\ |\phi\rangle_B \in \mathcal{H}_B,\ \langle\psi|\psi\rangle = 1 = \langle\phi|\phi\rangle\right\}\right). \tag{S4}$$

As mentioned in the main text, Werner, Holevo, and Shirokov [1] have shown that a state σ_{AB} is separable if and only if it can be expressed as

$$\sigma_{AB} = \int |\psi\rangle\!\langle\psi|_A \otimes |\phi\rangle\!\langle\phi|_B \,\mathrm{d}\mu(\psi,\phi), \tag{S5}$$

where μ is a Borel probability measure on the product of the sets of local (normalised) pure states.

The cone generated by the set of separable states will be denoted with

$$\mathcal{S}_{AB} \coloneqq \mathrm{cone}\left(\mathcal{S}^1_{AB}\right) \coloneqq \left\{\lambda\sigma_{AB} :\ \lambda \geq 0,\ \sigma_{AB} \in \mathcal{S}^1_{AB}\right\}. \tag{S6}$$

As an outer approximation to $\mathcal{S}_{AB}$, one often employs the cone of positive operators that also have a positive partial transpose (PPT). In formula, this is given by

$$\mathcal{PPT}_{AB} \coloneqq \left\{T_{AB} \in \mathcal{T}_+(\mathcal{H}_{AB}) :\ T_{AB}^\Gamma \geq 0\right\}, \tag{S7}$$

and Γ stands for the partial transpose. As recalled already in the main text, this is defined by the expression

$$\Gamma(X_A \otimes Y_B) = (X_A \otimes Y_B)^\Gamma \coloneqq X_A \otimes Y_B^\intercal \tag{S8}$$

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[1] Here, 'bounded' is intended in the Banach space sense; that is, we require that $\|\Lambda(T)\|_1 \leq C\|T\|_1$ for some constant $C < \infty$ and all $T \in \mathcal{T}(\mathcal{H}_A)$.

on simple tensors, and is extended to the whole $\mathcal{T}(\mathcal{H}_{AB})$ by linearity and continuity. Some subtleties related to the infinite-dimensional case are discussed in the Supplementary Note VII. It has been long known that [2]

$$\mathcal{S}_{AB} \subseteq \mathcal{PPT}_{AB} \tag{S9}$$

for all bipartite systems AB. Leveraging this fact, one can introduce an easily computable entanglement measure known as the ***logarithmic negativity***, given by [3, 4]

$$E_N(\rho_{AB}) := \log_2 \left\|\rho_{AB}^\Gamma\right\|_1 = \log_2 \sup\left\{\operatorname{Tr} X\rho :\ \left\|X^\Gamma\right\|_\infty \leq 1\right\}. \tag{S10}$$

Note. Hereafter, unless otherwise specified, $\mathcal{K}_{AB}$ will denote one of the two cones $\mathcal{K}_{AB} = \mathcal{S}_{AB}$ or $\mathcal{K}_{AB} = \mathcal{PPT}_{AB}$ defined by (S6) and (S7), respectively.

All states from now are understood to be on a bipartite system AB, although we will often drop the subscripts for the sake of readability. The ***(standard) $\mathcal{K}$-robustness*** of a state ρ is defined as

$$R^s_{\mathcal{K}}(\rho) := \inf\left\{\operatorname{Tr}\delta :\ \delta \in \mathcal{K},\ \rho + \delta \in \mathcal{K}\right\}. \tag{S11}$$

Here, it is understood that the variable δ is a trace-class operator. Much of the appeal of the expression in (S11) is that it is a convex optimisation program, and even a semi-definite program (SDP) for the special case of $\mathcal{K} = \mathcal{PPT}$ (however, it is an infinite-dimensional optimisation when ρ acts on an infinite-dimensional space). Note that $R^s_{\mathcal{PPT}}(\rho) \leq R^s_{\mathcal{S}}(\rho)$ holds for all states ρ, as a simple inspection of (S11) reveals.

Note. Our definition of robustness $R^s_{\mathcal{K}}$ follows the convention of Vidal and Tarrach [5]. The robustness as constructed in Ref. [6, 7], instead, would be expressed as $1 + R_{\mathcal{K}}$ in our current notation.

It turns out that $1 + 2R^s_{\mathcal{K}}(\rho)$ is nothing but the base norm of $\rho = \rho_{AB}$ as computed in the base norm space $(\mathcal{B}_{\text{sa}}(\mathcal{H}_{AB}), \mathcal{K}_{AB}, \operatorname{Tr}_{AB})$. This is proved in Ref. [6, Lemma 25] for the case $\mathcal{K} = \mathcal{S}$, and in Supplementary Note VII for the case $\mathcal{K} = \mathcal{PPT}$. Leveraging this correspondence, one can establish the dual representation [6, Eq. (23) and Lemma 25]

$$1 + 2R^s_{\mathcal{K}}(\rho) = \sup\left\{\operatorname{Tr} X\rho :\ X \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*}\right\} =: \|\rho\|_{\mathcal{K}}, \tag{S12}$$

where

$$\mathcal{K}^* := \left\{Z_{AB} \in \mathcal{B}_{\text{sa}}(\mathcal{H}_{AB}) :\ \operatorname{Tr}[Z_{AB}W_{AB}] \geq 0 \quad \forall\, W_{AB} \in \mathcal{K}_{AB}\right\} \subset \mathcal{B}_{\text{sa}}(\mathcal{H}_{AB}), \tag{S13}$$

$$[-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*} := \left\{Z_{AB} \in \mathcal{B}_{\text{sa}}(\mathcal{H}_{AB}) :\ |\operatorname{Tr}[Z_{AB}W_{AB}]| \leq \operatorname{Tr} W_{AB} \quad \forall\, W_{AB} \in \mathcal{K}_{AB}\right\} \subset \mathcal{B}_{\text{sa}}(\mathcal{H}_{AB}). \tag{S14}$$

The notation $[-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*}$ is motivated by the fact that this set can be understood as an operator interval with respect to the cone $\mathcal{K}^*$, in the sense that $[-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*} = (\mathbb{1} - \mathcal{K}^*) \cap (-\mathbb{1} + \mathcal{K}^*)$. Combining expressions (S11) and (S12) allows us to compute the robustness exactly in some cases. For instance, for a pure state $\Psi_{AB} = |\Psi\rangle\!\langle\Psi|_{AB}$ with Schmidt decomposition $|\Psi\rangle_{AB} = \sum_{j=0}^\infty \sqrt{\lambda_j}\, |e_j\rangle_A\, |f_j\rangle_B$ it holds that [5–7]

$$R^s_{\mathcal{K}}(\Psi) = \left(\sum\nolimits_{j=0}^\infty \sqrt{\lambda_j}\right)^2 - 1\,. \tag{S15}$$

The special case of this formula where $\Psi = \Phi_2^{\otimes k}$ is made of k copies of an entanglement bit $|\Phi_2\rangle = \frac{1}{\sqrt{2}}(|00\rangle + |11\rangle)$ is especially useful. We obtain that

$$R^s_{\mathcal{K}}\left(\Phi_2^{\otimes k}\right) = 2^k - 1\,. \tag{S16}$$

On a different note, by combining the expression in (S12) with the elementary estimate $\|\rho\|_{\mathcal{K}} \geq \left\|\rho^\Gamma\right\|_1$ one deduces the following.

Lemma S1. *For all states ρ, it holds that*

$$R^s_{\mathcal{S}}(\rho) \geq R^s_{\mathcal{PPT}}(\rho) \geq \frac{\left\|\rho^\Gamma\right\|_1 - 1}{2}\,. \tag{S17}$$

B. Distillable entanglement and entanglement cost

We will also need some notation for the set of quantum channels that preserve separability and PPT-ness, or — in short — $\mathcal{K}$-ness. Remember that we denote with (C)PTP$_{A\to A'}$ the set of (completely) positive and trace-preserving maps from A to A'. Since our results actually hold for all positive transformations, even those that are not completely positive, we will drop the complete positivity assumption from now on.

Thus, let us we define ***$\mathcal{K}$-preserving operations*** between two bipartite systems AB and $A'B'$ by

$$\mathrm{KP}\,(AB\to A'B') := \{\Lambda \in \mathrm{PTP}\,(AB\to A'B') :\ \Lambda(\mathcal{K}_{AB}) \subseteq \mathcal{K}_{A'B'}\}\,. \tag{S18}$$

When $\mathcal{K}=\mathcal{S}$, the above identity defines the set of non-entangling (or separability-preserving) operations employed in the main text. When $\mathcal{K}=\mathcal{PPT}$, we obtain the family of PPT-preserving operations instead. Note that the name of 'PPT-preserving' maps has been used in the literature to refer to several distinct concepts; we stress that here we only impose that $\Lambda(\sigma)$ is PPT whenever σ is.

Let us comment briefly on this choice of transformations. As discussed in the main text, the intention here is to be as general as possible — it would be beside the point to ask ourselves whether all non-entangling transformations are physically implementable in any given theory; in general this might not always be the case. In exactly the same way, not all transformations that obey the second law of thermodynamics are physical: consider e.g. one that does not preserve electric charge or angular momentum. The assumption of no entanglement generation is merely a necessary condition for a transformation to be physical — any practical process used for entanglement manipulation should not create extra entanglement from nothing, and hence should be in KP$(AB\to A'B')$.

We record the following elementary yet important observation.

Lemma S2. *For $\mathcal{K}=\mathcal{S},\mathcal{PPT}$, the $\mathcal{K}$-robustnesses* (S11) *is monotonic under $\mathcal{K}$-preserving operations.*

Proof. The proof follows standard arguments, and indeed an even stronger variant of the monotonicity of the robustnesses (selective monotonicity) has been shown e.g. in Ref. [8]; we repeat the basic argument here only for the sake of convenience. For a bipartite state ρ_{AB}, an arbitrary $\varepsilon>0$, and some $\Lambda\in\mathrm{KP}\,(AB\to A'B')$, let $\delta_{AB}\in\mathcal{K}_{AB}$ be such that $\rho_{AB}+\delta_{AB}\in\mathcal{K}_{AB}$ and $\operatorname{Tr}\delta_{AB}\le R^s_{\mathcal{K}}(\rho_{AB})+\varepsilon$. Then $\Lambda(\delta_{AB})\in\mathcal{K}_{A'B'}$ and also $\Lambda(\rho_{AB})+\Lambda(\delta_{AB})=\Lambda(\rho_{AB}+\delta_{AB})\in\mathcal{K}_{A'B'}$, so that

$$R^s_{\mathcal{K}}\left(\Lambda(\rho_{AB})\right)\le \operatorname{Tr}\Lambda(\delta_{AB})=\operatorname{Tr}\delta_{AB}\le R^s_{\mathcal{K}}(\rho_{AB})+\varepsilon\,.$$

Since this holds for arbitrary $\varepsilon>0$, we deduce that $R^s_{\mathcal{K}}\left(\Lambda(\rho_{AB})\right)\le R^s_{\mathcal{K}}(\rho_{AB})$, as claimed. □

We now recall the definitions of distillable entanglement and entanglement cost of a state ρ_{AB} under $\mathcal{K}$-preserving operations. We present here a slightly more general construction than in the Methods section of the main text, namely, one which incorporates a non-zero asymptotic error. Following e.g. Vidal and Werner [3, Eq. (43)], for an arbitrary $\varepsilon\in[0,1)$ let us set

$$E^{\varepsilon}_{d,\mathrm{KP}}(\rho_{AB}) := \sup\left\{R>0:\ \limsup_{n\to\infty}\ \inf_{\Lambda_n\in\mathrm{KP}\left(A^nB^n\to A_0^{\lceil Rn\rceil}B_0^{\lceil Rn\rceil}\right)} \frac12\left\|\Lambda_n\left(\rho_{AB}^{\otimes n}\right)-\Phi_2^{\otimes\lceil Rn\rceil}\right\|_1\le\varepsilon\right\}, \tag{S19}$$

$$E^{\varepsilon}_{c,\mathrm{KP}}(\rho_{AB}) := \inf\left\{R>0:\ \limsup_{n\to\infty}\ \inf_{\Lambda_n\in\mathrm{KP}\left(A_0^{\lfloor Rn\rfloor}B_0^{\lfloor Rn\rfloor}\to A^nB^n\right)} \frac12\left\|\Lambda_n\left(\Phi_2^{\otimes\lfloor Rn\rfloor}\right)-\rho_{AB}^{\otimes n}\right\|_1\le\varepsilon\right\}. \tag{S20}$$

For a fixed ρ, the function $E^{\varepsilon}_{d,\mathrm{KP}}(\rho)$ is non-decreasing in ε, while $E^{\varepsilon}_{c,\mathrm{KP}}(\rho)$ is non-increasing. Also, note that

$$E_{d,\mathrm{KP}}(\rho_{AB}) := E^{0}_{d,\mathrm{KP}}(\rho_{AB})\,,\qquad E_{c,\mathrm{KP}}(\rho_{AB}) := E^{0}_{c,\mathrm{KP}}(\rho_{AB})\,, \tag{S21}$$

coincide with the quantities discussed in the Methods section of the main text.

A variation on the notions of distillable entanglement and entanglement cost can be obtained by looking only at *exact* transformations. The corresponding modified entanglement measures read

$$E_{d,\,\mathrm{KP}}^{\mathrm{exact}}(\rho_{AB}) := \sup\left\{R>0:\ \exists\, n_0:\ \forall\, n\geq n_0\ \ \exists\, \Lambda_n\in \mathrm{KP}\left(A^nB^n\to A_0^{\lceil Rn\rceil}B_0^{\lceil Rn\rceil}\right):\ \Lambda_n\left(\rho_{AB}^{\otimes n}\right)=\Phi_2^{\otimes\lceil Rn\rceil}\right\}, \tag{S22}$$

$$E_{c,\,\mathrm{KP}}^{\mathrm{exact}}(\rho_{AB}) := \inf\left\{R>0:\ \exists\, n_0:\ \forall\, n\geq n_0\ \ \exists\, \Lambda_n\in \mathrm{KP}\left(A_0^{\lfloor Rn\rfloor}B_0^{\lfloor Rn\rfloor}\to A^nB^n\right):\ \Lambda_n\left(\Phi_2^{\otimes\lfloor Rn\rfloor}\right)=\rho_{AB}^{\otimes n}\right\}. \tag{S23}$$

Although less operationally meaningful than their error-tolerant counterparts (S19)–(S20), the exact distillable entanglement and the exact entanglement cost are nevertheless useful sometimes. For instance, they can come in handy in establishing bounds, thanks to the simple inequalities

$$E_{d,\,\mathrm{KP}}^{\mathrm{exact}}(\rho)\leq E_{d,\,\mathrm{KP}}(\rho)\leq E_{c,\,\mathrm{KP}}(\rho)\leq E_{c,\,\mathrm{KP}}^{\mathrm{exact}}(\rho)\,, \tag{S24}$$

which hold for all bipartite states ρ. We now introduce formally the notion of reversibility for the theory of entanglement manipulation under $\mathcal{K}$-preserving transformations.

Definition S3. *The theory of entanglement manipulation is said to be* **reversible under $\mathcal{K}$-preserving operations** *if $E_{d,\,\mathrm{KP}}(\rho_{AB})=E_{c,\,\mathrm{KP}}(\rho_{AB})$ holds for all bipartite states ρ_{AB}. Otherwise it is said to be* **irreversible under $\mathcal{K}$-preserving operations**.

It is not difficult to realise that neither of the two families of $\mathcal{K}$-preserving operations, for $\mathcal{K}=\mathcal{S}$ and $\mathcal{K}=\mathcal{PPT}$, is a subset of the other. Hence, one would be tempted to deduce that the distillable entanglement and the entanglement cost under non-entangling and PPT-preserving operations do not obey any general inequality. This is however not the case, and the reason must be traced back to the very high degree of symmetry exhibited by the maximally entangled state, and — more precisely — to the fact that for isotropic states the 'PPT criterion' [2] is necessary and sufficient for separability [9]. The operational relation between the two classes can be inferred from [10, Remark on pp. 843–844] already, and we can formalise this observation as follows.

Lemma S4. *For all bipartite states ρ and all $\varepsilon\in[0,1)$ it holds that*

$$E_{d,\,\mathrm{NE}}^{\varepsilon}(\rho)\geq E_{d,\,\mathrm{PPTP}}^{\varepsilon}(\rho)\,,\qquad E_{c,\,\mathrm{NE}}^{\varepsilon}(\rho)\geq E_{c,\,\mathrm{PPTP}}^{\varepsilon}(\rho)\,, \tag{S25}$$

$$E_{d,\,\mathrm{NE}}^{\mathrm{exact}}(\rho)\geq E_{d,\,\mathrm{PPTP}}^{\mathrm{exact}}(\rho)\,,\qquad E_{c,\,\mathrm{NE}}^{\mathrm{exact}}(\rho)\geq E_{c,\,\mathrm{PPTP}}^{\mathrm{exact}}(\rho)\,. \tag{S26}$$

In particular,

$$E_{d,\,\mathrm{NE}}(\rho)\geq E_{d,\,\mathrm{PPTP}}(\rho)\,,\qquad E_{c,\,\mathrm{NE}}(\rho)\geq E_{c,\,\mathrm{PPTP}}(\rho)\,. \tag{S27}$$

Proof. We prove only the first inequality, as all the others are completely analogous. Let R be an achievable rate for $E_{d,\,\mathrm{PPTP}}^{\varepsilon}(\rho)$ at error threshold $\varepsilon\in[0,1)$, and let $\Lambda_n\in\mathrm{PPTP}$ be PPT-preserving operations satisfying that $\limsup_{n\to\infty}\frac12\left\|\Lambda_n\left(\rho^{\otimes n}\right)-\Phi_2^{\otimes\lceil Rn\rceil}\right\|_1\leq\varepsilon$. To proceed further, define the twirling operation $\mathcal{T}_m$ on a $2^m\times 2^m$ bipartite system by [9]

$$\begin{aligned}\mathcal{T}_m(X) &:= \int (U\otimes U^*)\,X\,(U\otimes U^*)^\dagger\,\mathrm{d}U\\ &= \mathrm{Tr}\left[X\Phi_2^{\otimes m}\right]\Phi_2^{\otimes m}+\mathrm{Tr}\left[X\left(\mathbb{1}-\Phi_2^{\otimes m}\right)\right]\frac{\mathbb{1}-\Phi_2^{\otimes m}}{4^m-1}\,,\end{aligned} \tag{S28}$$

where $\mathrm{d}U$ denotes the Haar measure over the (local) unitary group. Note that $\mathcal{T}_m\in\mathrm{NE}\cap\mathrm{PPTP}$ — in fact, $\mathcal{T}_m$ can be physically implemented with local operations and shared randomness — and that the output states of $\mathcal{T}_m$ are all isotropic, that is, they are linear combinations of $\Phi_2^{\otimes m}$ and the maximally mixed state. For states of this form, it is known that the PPT criterion is necessary and sufficient for separability [9].

We now claim that $\mathcal{T}_{\lceil Rn \rceil} \circ \Lambda_n \in \mathrm{NE}$. To see why this is the case, note that for all states $\sigma \in \mathcal{S} \subseteq \mathcal{PPT}$ it holds that $\Lambda_n(\sigma) \in \mathcal{PPT}$ and hence $\left(\mathcal{T}_{\lceil Rn \rceil} \circ \Lambda_n\right)(\sigma) \in \mathcal{PPT}$. However, since the latter is an isotropic state, we conclude that in fact $\left(\mathcal{T}_{\lceil Rn \rceil} \circ \Lambda_n\right)(\sigma) \in \mathcal{S}$, proving the claim. Observe also that

$$\left\| \left(\mathcal{T}_{\lceil Rn \rceil} \circ \Lambda_n\right)\left(\rho^{\otimes n}\right) - \Phi_2^{\otimes \lceil Rn \rceil} \right\|_1 = \left\| \left(\mathcal{T}_{\lceil Rn \rceil} \circ \Lambda_n\right)\left(\rho^{\otimes n}\right) - \mathcal{T}_{\lceil Rn \rceil}\left(\Phi_2^{\otimes \lceil Rn \rceil}\right) \right\|_1 \leq \left\| \Lambda_n\left(\rho^{\otimes n}\right) - \Phi_2^{\otimes \lceil Rn \rceil} \right\|_1 , \tag{S29}$$

where the last inequality is a consequence of the contractivity of the trace norm under positive trace preserving maps [11], or more mundanely of the triangle inequality applied to the integral representation (S28) of $\mathcal{T}_{\lceil Rn \rceil}$. The above relation implies that the distillation rate R is achievable at error threshold ε by means of the separability-preserving operations $\mathcal{T}_{\lceil Rn \rceil} \circ \Lambda_n$, i.e. $E_{d,\mathrm{NE}}(\rho) \geq R$. Taking the infimum in R we obtain the sought inequality $E_{d,\mathrm{NE}}(\rho) \geq E_{d,\mathrm{PPTP}}(\rho)$. □

II. TEMPERED ROBUSTNESS AND TEMPERED NEGATIVITY

The main idea is to introduce a modified version of the standard $\mathcal{K}$-robustness in (S11) by modifying the dual program in (S12). Namely, for a pair of states ρ, ω, let us define the ***ω-tempered $\mathcal{K}$-robustness*** by

$$1 + 2R_{\mathcal{K}}^{\tau}(\rho|\omega) := \sup\left\{ \operatorname{Tr} X\rho : \; X \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*} , \; \|X\|_\infty = \operatorname{Tr} X\omega \right\} , \tag{S30}$$

$$R_{\mathcal{K}}^{\tau}(\rho) := R_{\mathcal{K}}^{\tau}(\rho|\rho) . \tag{S31}$$

Here, the operator interval $[-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*}$ is defined by (S14). Note that the constraint $\|X\|_\infty = \operatorname{Tr} X\omega$ can be rewritten as $-(\operatorname{Tr} X\omega)\,\mathbb{1} \leq X \leq (\operatorname{Tr} X\omega)\,\mathbb{1}$. Therefore, the expression in (S30) is a convex program, and even an SDP for the special case $\mathcal{K} = \mathcal{PPT}$. What this additional constraint is trying to tell us is that the support $\operatorname{supp}\omega$ of ω lies entirely within the eigenspace of X corresponding to the eigenvalue with the largest modulus. At this point, a little thought shows that $R_{\mathcal{K}}^{\tau}(\rho|\omega)$ depends in fact only on ρ and $\operatorname{supp}\omega$. Analogously, $R_{\mathcal{K}}^{\tau}(\rho)$ depends only on $\operatorname{supp}\rho$.

We also introduce a further quantity, the ***ω-tempered negativity***, defined by

$$N_\tau(\rho|\omega) := \sup\left\{ \operatorname{Tr} X\rho : \; \left\|X^\Gamma\right\|_\infty \leq 1, \; \|X\|_\infty = \operatorname{Tr} X\omega \right\} , \tag{S32}$$

$$N_\tau(\rho) := N_\tau(\rho|\rho) . \tag{S33}$$

Exactly as above, it does not take long to realise that the expression in (S32) is in fact an SDP, and that $N_\tau(\rho|\omega)$ depends only on ρ and $\operatorname{supp}\omega$, while $N_\tau(\rho)$ depends only on $\operatorname{supp}\rho$. The corresponding ***tempered logarithmic negativity*** is

$$E_N^\tau(\rho) := \log_2 N_\tau(\rho) . \tag{S34}$$

The main elementary properties of the tempered robustness and negativity — related to their monotonicity, multiplicativity, and various bounds between the quantities — are gathered in the following proposition.

Proposition S5. *For $\mathcal{K} = \mathcal{S}, \mathcal{PPT}$ and for all pairs of states ρ, ω on a bipartite system AB, it holds that:*

(a) $0 \leq R_{\mathcal{K}}^{\tau}(\rho|\omega) \leq R_{\mathcal{K}}^{s}(\rho)$, *and* $R_{\mathcal{K}}^{s}(\rho) = \sup_{\omega'} R_{\mathcal{K}}^{\tau}(\rho|\omega')$;

(b) $1 \leq N_\tau(\rho|\omega) \leq \left\|\rho^\Gamma\right\|_1$, *and* $\left\|\rho^\Gamma\right\|_1 = \sup_{\omega'} N_\tau(\rho|\omega')$;

(c) $R_{\mathcal{K}}^{\tau}$ *is monotonic under the simultaneous action of any $\mathcal{K}$-preserving map $\Lambda \in \mathrm{KP}(AB \to A'B')$, in formula*

$$R_{\mathcal{K}}^{\tau}\left(\Lambda(\rho) \middle| \Lambda(\omega)\right) \leq R_{\mathcal{K}}^{\tau}(\rho|\omega) . \tag{S35}$$

(d) *the inequalities*

$$R_{\mathcal{S}}^{\tau}(\rho|\omega) \geq R_{PPT}^{\tau}(\rho|\omega) \geq \frac{N_\tau(\rho|\omega) - 1}{2} \tag{S36}$$

are satisfied.

(e) N_τ *is super-multiplicative and hence* E_N^τ *is super-additive, in formula*

$$N_\tau\left(\rho_1 \otimes \rho_2 \middle| \omega_1 \otimes \omega_2\right) \geq N_\tau(\rho_1|\omega_1)\, N_\tau(\rho_2|\omega_2)\,, \tag{S37}$$

$$E_N^\tau\left(\rho_1 \otimes \rho_2 \middle| \omega_1 \otimes \omega_2\right) \geq E_N^\tau(\rho_1|\omega_1) + E_N^\tau(\rho_2|\omega_2)\,, \tag{S38}$$

for all states $\rho_1, \rho_2, \omega_1, \omega_2$.

Proof. We proceeed one claim at a time.

(a) Taking $X = \mathbb{1}$ in the definition of $R_{\mathcal{K}}^\tau$ (S30) yields immediately that $R_{\mathcal{K}}^\tau(\omega|\tau) \geq 0$. Also, since we obtained (S30) by adding one more constraint to the dual program (S12) for the standard robustness, it is clear that the value of the the supremum can only decrease, implying that $R_{\mathcal{K}}^\tau(\rho|\omega) \leq R_{\mathcal{K}}^s(\rho)$.

On the other hand, it is not difficult to verify that the operators X in the dual formulation of $R_{\mathcal{K}}^s$ (S12) can always be assumed to be compact and in fact even of finite rank. Indeed, thanks to the fact that $\mathcal{H}_A$ and $\mathcal{H}_B$ are separable Hilbert spaces, we can pick sequences of finite-dimensional projectors $(P_A^N)_{N\in\mathbb{N}}$ and $(P_B^N)_{N\in\mathbb{N}}$ such that $(P_A^N \otimes P_B^N)\rho_{AB}(P_A^N \otimes P_B^N) \xrightarrow[N\to\infty]{} \rho_{AB}$ in trace norm. For any given $X_{AB} \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*}$, the finite-rank operators $X_N := (P_A^N \otimes P_B^N) X_{AB} (P_A^N \otimes P_B^N)$ satisfy that $X_N \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*}$, simply because $\sigma_{AB} \mapsto (P_A^N \otimes P_B^N)\sigma_{AB}(P_A^N \otimes P_B^N)$ sends $\mathcal{K}$ into itself and is trace non-increasing. Moreover, $\mathrm{Tr}[\rho X_N] \xrightarrow[N\to\infty]{} \mathrm{Tr}\,\rho X$.

This shows that X in (S12) can be taken to be of finite rank. For any such X, there will exist a state ω_X such that $\|X\|_\infty = \mathrm{Tr}\, X\omega_X$; in fact, it suffices to have the support of ω_X span the eigenspace of X corresponding to the eigenvalue with maximum modulus. Therefore,

$$\sup_{\omega'} R_{\mathcal{K}}^\tau(\rho|\omega') = \sup_{\substack{X\in[-\mathbb{1},\mathbb{1}]_{\mathcal{K}^*},\, \omega':\\ \|X\|_\infty = \mathrm{Tr}\, X\omega'}} \mathrm{Tr}\, X\rho \geq \sup_{\substack{X\in[-\mathbb{1},\mathbb{1}]_{\mathcal{K}^*},\\ \mathrm{rk}\, X<\infty}} \mathrm{Tr}\, X\rho = R_{\mathcal{K}}^s(\rho)\,.$$

(b) The lower bound $N_\tau(\rho|\omega) \geq 1$ can be retrieved by setting $X = \mathbb{1}$ in (S32). The fact that $N_\tau(\rho|\omega) \leq \left\|\rho^\Gamma\right\|_1$ follows by comparing (S32) with the dual form of the negativity on the rightmost side of (S10). The equality $\left\|\rho^\Gamma\right\|_1 = \sup_{\omega'} N_\tau(\rho|\omega')$ is proved as for claim (a). One starts by showing that the operator X in the rightmost side of (S10) can be assumed to have finite rank. To see this, it suffices to observe that $X_N = (P_A^N \otimes P_B^N) X_{AB} (P_A^N \otimes P_B^N)$ defined as before satisfies that

$$\left\|X_N^\Gamma\right\|_\infty = \left\|P_A^N \otimes \left(P_B^N\right)^\intercal X_{AB}^\Gamma\, P_A^N \otimes \left(P_B^N\right)^\intercal\right\|_1 \leq \left\|X^\Gamma\right\|_1\,.$$

Since $\mathrm{Tr}[\rho X_N] \xrightarrow[N\to\infty]{} \mathrm{Tr}\,\rho X$, considering the sequence of finite-rank operators $(X_N)_N$ instead of X in (S10) leads to the same value of the optimisation.

(c) It suffices to write that

$$\begin{aligned} R_{\mathcal{K}}^\tau\left(\Lambda(\rho)\,\middle|\,\Lambda(\omega)\right) &= \sup\left\{\mathrm{Tr}\, X\Lambda(\rho):\ X \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*},\ \|X\|_\infty = \mathrm{Tr}\, X\Lambda(\omega)\right\} \\ &\overset{\text{(i)}}{=} \sup\left\{\mathrm{Tr}\, \Lambda^\dagger(X)\rho:\ X \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*},\ \|X\|_\infty = \mathrm{Tr}\, \Lambda^\dagger(X)\omega\right\} \\ &\overset{\text{(ii)}}{\leq} \sup\left\{\mathrm{Tr}\, Y\rho:\ Y \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*},\ \|Y\|_\infty = \mathrm{Tr}\, Y\omega\right\} \\ &= R_{\mathcal{K}}^\tau(\rho|\omega)\,. \end{aligned} \tag{S39}$$

Note that in (i) we just used the definition of adjoint map. Justifying (ii) requires a bit more care. We start by observing that if $X \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*}$ then $Y = \Lambda^\dagger(X)$ satisfies

$$\sup_{\sigma\in\mathcal{K},\, \operatorname{Tr}\sigma=1} |\operatorname{Tr} Y\sigma| = \sup_{\sigma\in\mathcal{K},\, \operatorname{Tr}\sigma=1} |\operatorname{Tr} X\Lambda(\sigma)| \leq \sup_{\sigma'\in\mathcal{K},\, \operatorname{Tr}\sigma'=1} |\operatorname{Tr} X\sigma'| \leq 1\,,$$

where the inequality holds because $\Lambda(\sigma)$ is a normalised quantum state and belongs to $\mathcal{K}$. (We remind the reader that our definition of $\mathcal{K}$-preserving maps imposes that any such map is also positive and trace preserving.) Moreover, $\|Y\|_\infty \leq \|X\|_\infty = \operatorname{Tr} X\Lambda(\omega) = \operatorname{Tr} Y\omega$ thanks to the positivity and unitality of $\Lambda^\dagger$ (see (S3)); this is in fact an equality, because on the other hand $\operatorname{Tr} Y\omega \leq \|Y\|_\infty \|\omega\|_1 = \|Y\|_\infty$. Since $Y = \Lambda^\dagger(X)$ satisfies that $Y \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*}$ and moreover $\|Y\|_\infty = \operatorname{Tr} Y\omega$, we deduce the inequality in (ii).

(d) It is easy to see from (S30) that $R^\tau_{\mathcal{K}}(\rho|\omega)$ is monotonically decreasing with respect to the inclusion ordering on the cone $\mathcal{K}$ for all fixed ρ and ω, meaning that $\mathcal{K}_1 \subseteq \mathcal{K}_2$ implies that $R^\tau_{\mathcal{K}_1}(\rho|\omega) \geq R^\tau_{\mathcal{K}_2}(\rho|\omega)$. Since $\mathcal{S} \subseteq \mathcal{PPT}$, the first inequality in (S36) follows.

We now move on to the second. Note that $\left\|X^\Gamma\right\|_\infty \leq 1$ entails that $X \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{S}^*}$, simply because for all $\sigma \in \mathcal{S}$ with $\operatorname{Tr}\sigma = 1$ one has that

$$|\operatorname{Tr}[X\sigma]| = \left|\operatorname{Tr}\left[X^\Gamma \sigma^\Gamma\right]\right| \leq \left\|X^\Gamma\right\|_\infty \left\|\sigma^\Gamma\right\|_1 \leq 1\,,$$

where we remembered that $\sigma^\Gamma \geq 0$ and hence $\left\|\sigma^\Gamma\right\|_1 = \operatorname{Tr}\sigma^\Gamma = \operatorname{Tr}\sigma = 1$. Hence, the set on the right-hand side of (S32) is contained in that on the right-hand side of (S30), which shows that $N_\tau(\rho|\omega) \leq 1 + 2R^\tau_{\mathcal{S}}(\rho|\omega)$.

(e) To show the super-multiplicativity of N_τ, it suffices to make a tensor product ansatz inside (S32), obtaining that

$$\begin{aligned} N_\tau\left(\rho_1\otimes\rho_2\middle|\omega_1\otimes\omega_2\right) &= \sup\left\{\operatorname{Tr} X_{12}(\rho_1\otimes\rho_2):\ \left\|X_{12}^\Gamma\right\|_\infty \leq 1,\ \|X_{12}\|_\infty = \operatorname{Tr} X_{12}(\omega_1\otimes\omega_2)\right\} \\ &\overset{\text{(iii)}}{\geq} \sup\left\{\operatorname{Tr}(X_1\otimes X_2)(\rho_1\otimes\rho_2):\ \left\|X_i^\Gamma\right\|_\infty \leq 1,\ \|X_i\|_\infty = \operatorname{Tr} X_i\omega_i\right\} \\ &= N_\tau(\rho_1|\omega_1)\, N_\tau(\rho_2|\omega_2)\,. \end{aligned} \tag{S40}$$

Here, the inequality in (iii) can be proved by noting that $\left\|X_i^\Gamma\right\|_\infty \leq 1$ entails that $\left\|(X_1\otimes X_2)^\Gamma\right\|_\infty = \left\|X_1^\Gamma\right\|_\infty \left\|X_2^\Gamma\right\|_\infty \leq 1$. The super-additivity of E_N^τ in (S38) follows immediately. □

In addition to the basic properties established above, our main results will rely on one more technical property of the tempered quantities. This is a perturbative version of Proposition S5(a), allowing us to relate the robustness $R^s_{\mathcal{K}}(\rho')$ of a given state with the tempered robustness $R^\tau_{\mathcal{K}}(\rho)$ of another state which is sufficiently close to it. The following lemma can quite rightly be regarded as lying at the heart of our method.

Lemma S6 (The ε-lemma)**.** *For all states ρ, ρ' such that*

$$\varepsilon := \frac{1}{2}\|\rho - \rho'\|_1 \leq \frac{1}{2}\,, \tag{S41}$$

it holds that

$$R^s_{\mathcal{K}}(\rho') \geq R^\tau_{\mathcal{K}}(\rho'|\rho) \geq (1-2\varepsilon)R^\tau_{\mathcal{K}}(\rho) - \varepsilon \tag{S42}$$

and also

$$N_\tau(\rho'|\rho) \geq (1-2\varepsilon)\, N_\tau(\rho)\,. \tag{S43}$$

Proof. The first inequality in (S42) is just an application of Proposition S5(a). As for the second, using the definition (S30) of $R_{\mathcal{K}}^{\tau}$ as well as Hölder's inequality we see that

$$
\begin{aligned}
1 + 2R_{\mathcal{K}}^{\tau}(\rho'|\rho) &= \sup\left\{\operatorname{Tr} X\rho' :\ X \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*},\ \|X\|_\infty = \operatorname{Tr} X\rho\right\} \\
&\geq \sup\left\{\operatorname{Tr} X\rho - \|X\|_\infty \|\rho - \rho'\|_1 :\ X \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*},\ \|X\|_\infty = \operatorname{Tr} X\rho\right\} \\
&= \sup\left\{(1-2\varepsilon) \operatorname{Tr} X\rho :\ X \in [-\mathbb{1}, \mathbb{1}]_{\mathcal{K}^*},\ \|X\|_\infty = \operatorname{Tr} X\rho\right\} \\
&= (1-2\varepsilon)\left(1 + 2R_{\mathcal{K}}^{\tau}(\rho)\right),
\end{aligned}
\tag{S44}
$$

which becomes (S42) upon elementary algebraic manipulations. The proof of (S43) is entirely analogous:

$$
\begin{aligned}
N_\tau(\rho'|\rho) &= \sup\left\{\operatorname{Tr} X\rho' :\ \left\|X^\Gamma\right\|_\infty \leq 1,\ \|X\|_\infty = \operatorname{Tr} X\rho\right\} \\
&\geq \sup\left\{\operatorname{Tr} X\rho - \|X\|_\infty \|\rho - \rho'\|_1 :\ \left\|X^\Gamma\right\|_\infty \leq 1,\ \|X\|_\infty = \operatorname{Tr} X\rho\right\} \\
&= \sup\left\{(1-2\varepsilon) \operatorname{Tr} X\rho :\ \left\|X^\Gamma\right\|_\infty \leq 1,\ \|X\|_\infty = \operatorname{Tr} X\rho\right\} \\
&= (1-2\varepsilon)\, N_\tau(\rho).
\end{aligned}
\tag{S45}
$$

This concludes the proof. □

III. MAIN RESULTS: IRREVERSIBILITY OF ENTANGLEMENT MANIPULATION

Here we state our main results concerning the theory of entanglement manipulation for quantum states. The extension of the argument to quantum channels will be tackled in full detail separately [12].

Theorem S7. *For $\mathcal{K} = \mathcal{S}$ or $\mathcal{K} = \mathcal{PPT}$, the entanglement cost under $\mathcal{K}$-preserving operations satisfies that*

$$
\inf_{\varepsilon \in [0, 1/2)} E_{c,\,\mathrm{KP}}^{\varepsilon}(\rho) \geq L_{\mathcal{K}}^{\tau}(\rho)\,,
\tag{S46}
$$

where

$$
L_{\mathcal{K}}^{\tau}(\rho) \coloneqq \limsup_{n\to\infty} \frac{1}{n} \log_2\left(1 + R_{\mathcal{K}}^{\tau}\left(\rho^{\otimes n}\right)\right) \geq E_N^{\tau}(\rho)\,,
\tag{S47}
$$

and the tempered logarithmic negativity is defined by (S34).

Remark S8. An interesting consequence of the above result is that the tempered logarithmic negativity is a lower bound on the standard entanglement cost under local operations and classical communication (LOCC), denoted $E_{c,\,\mathrm{LOCC}}$, in formula

$$
E_{c,\,\mathrm{LOCC}}(\rho_{AB}) \geq E_N^{\tau}(\rho_{AB}) \qquad \forall\ \rho_{AB}\,.
\tag{S48}
$$

The entanglement cost under LOCC is a notoriously hard quantity to compute; it is given by the regularised entanglement of formation [13], and the regularisation is known to be necessary due to Hastings's counterexample to the additivity conjectures [14, 15]. Previously known lower bounds include the regularised relative entropy of entanglement [16] and the squashed entanglement [17–19], both of which are extremely hard to evaluate in general (albeit for different reasons). The former can be in turn lower bounded by either Piani's measured relative entropy of entanglement [20], which has the advantage of doing away with regularisations, or by the E_η measure recently proposed by Wang and Duan [21], which is particularly convenient computationally because it is given by a semi-definite program (SDP). Both of these lower bounds on the LOCC entanglement cost, that inferred by Piani's results and that relying on E_η, are quite useful, but are known to be weaker than that given by the regularised relative entropy of entanglement.

The tempered negativity provides us with an independent lower bound on the LOCC entanglement cost that can strictly improve on the regularised relative entropy one. This latter fact will be apparent from the proof of Theorem S9. Also, since it is also given by an SDP, our bound is still computationally friendly. We are aware of no other quantity possessing these properties.

Proof of Theorem S7. The following argument could be marginally simplified at the level of notation by resorting to the results of Brandão and Plenio [10]. However, for the sake of readability we prefer to give a more direct and self-contained proof.

Call AB the bipartite system where ρ lives. Let R be an achievable rate for the entanglement cost $E_{c,\,\mathrm{KP}}^{\varepsilon}(\rho)$ at some error threshold $\varepsilon \in [0, 1/2)$. Consider a sequence of operations $\Lambda_n \in \mathrm{KP}_{A_0^{\lfloor Rn\rfloor}B_0^{\lfloor Rn\rfloor}\to A^nB^n}$, with A_0, B_0 being single-qubit systems, such that

$$\varepsilon_n := \frac{1}{2}\left\|\Lambda_n\left(\Phi_2^{\otimes\lfloor Rn\rfloor}\right) - \rho^{\otimes n}\right\|_1 \tag{S49}$$

with

$$\limsup_{n\to\infty} \varepsilon_n \leq \varepsilon < \frac{1}{2}\,. \tag{S50}$$

For all sufficiently large n, we then write

$$\begin{aligned}
2^{\lfloor Rn\rfloor} &\overset{\text{(i)}}{=} 1 + R_{\mathcal{K}}^{s}\left(\Phi_2^{\otimes\lfloor Rn\rfloor}\right)\\
&\overset{\text{(ii)}}{\geq} 1 + R_{\mathcal{K}}^{s}\left(\Lambda_n\left(\Phi_2^{\otimes\lfloor Rn\rfloor}\right)\right)\\
&\overset{\text{(iii)}}{\geq} (1-2\varepsilon_n)\left(1 + R_{\mathcal{K}}^{\tau}\left(\rho^{\otimes n}\right)\right) + \varepsilon_n\\
&\geq (1-2\varepsilon_n)\left(1 + R_{\mathcal{K}}^{\tau}\left(\rho^{\otimes n}\right)\right)
\end{aligned} \tag{S51}$$

Here, in (i) we just recalled the value of the standard robustness of maximally entangled states (S16), (ii) comes from the monotonicity of $R_{\mathcal{K}}^{s}$ under $\mathcal{K}$-preserving operations (Lemma S2), and (iii) is an application of the ε-lemma (Lemma S6). Taking the logarithm, dividing by n, and computing the limit for $n \to \infty$ yields

$$\begin{aligned}
R &= \lim_{n\to\infty} \frac{\lfloor Rn\rfloor}{n}\\
&\geq \limsup_{n\to\infty} \frac{1}{n}\log_2\left((1-2\varepsilon_n)\left(1 + R_{\mathcal{K}}^{\tau}\left(\rho^{\otimes n}\right)\right)\right)\\
&\geq \limsup_{n\to\infty} \frac{1}{n}\log_2\left(1 + R_{\mathcal{K}}^{\tau}\left(\rho^{\otimes n}\right)\right) + \liminf_{n\to\infty}\frac{1}{n}\log_2(1-2\varepsilon_n)\\
&\overset{\text{(iv)}}{=} \limsup_{n\to\infty} \frac{1}{n}\log_2\left(1 + R_{\mathcal{K}}^{\tau}\left(\rho^{\otimes n}\right)\right)\\
&= L_{\mathcal{K}}^{\tau}(\rho)\,.
\end{aligned} \tag{S52}$$

where (iv) is a consequence of the fact that ε_n is bounded away from $1/2$, as per (S50). This completes the proof of the first inequality (S46).

As for the second inequality (S47), we observe that

$$
\begin{aligned}
L_{\mathcal{K}}^{\tau}(\rho) &= \limsup_{n\to\infty} \frac{1}{n} \log_2 \left(1 + R_{\mathcal{K}}^{\tau}\left(\rho^{\otimes n}\right)\right) \\
&\overset{\text{(v)}}{\geq} \limsup_{n\to\infty} \frac{1}{n} \log_2 \frac{N_\tau\left(\rho^{\otimes n}\right) + 1}{2} \\
&\geq \limsup_{n\to\infty} \left(\frac{1}{n} \log_2 N_\tau\left(\rho^{\otimes n}\right) - \frac{1}{n} \right) \\
&\overset{\text{(vi)}}{\geq} \limsup_{n\to\infty} \left(\frac{1}{n} \log_2 \left(N_\tau\left(\rho\right)^n\right) - \frac{1}{n} \right) \\
&\overset{\text{(vii)}}{=} E_N^{\tau}(\rho)\,.
\end{aligned}
\tag{S53}
$$

Here, (v) is an application of the lower bound in Proposition S5(d), in (vi) we leveraged the super-multiplicativity of the tempered negativity (Proposition S5(e)), and finally (vii) is just the definition (S34) of tempered logarithmic negativity. □

Before we state and prove our result on the irreversibility of entanglement, we need to recall and discuss two well-known bounds on the distillable entanglement. Lower bounds on $E_{d,\,\mathrm{KP}}(\rho)$ can be obtained by looking at smaller classes of operations included in the set of all $\mathcal{K}$-preserving ones. A typical choice is the set of local operations assisted by one-way classical communication, say from Alice to Bob, denoted with $\mathrm{LOCC}_{\to}$. In this setting, Devetak and Winter's hashing inequality [22] states that

$$
E_{d,\,\mathrm{LOCC}_{\to}}(\rho_{AB}) \geq I_{\mathrm{coh}}(A\rangle B)_\rho := S(\rho_B) - S(\rho_{AB})\,, \tag{S54}
$$

where $S(\omega) := -\operatorname{Tr}\omega\log_2\omega$ is the von Neumann entropy, and $\rho_B := \operatorname{Tr}_A \rho_{AB}$ is the reduced state of ρ_{AB} on Bob's side. Since local operations assisted by one-way classical communication are both non-entangling and PPT-preserving, in formula $\mathrm{LOCC}_{\to} \subseteq \mathrm{KP}$, we see that $E_{d,\,\mathrm{LOCC}_{\to}}(\rho_{AB}) \leq E_{d,\,\mathrm{KP}}(\rho_{AB})$. In particular, the rightmost side of the hashing inequality (S54) lower bounds the distillable entanglement under $\mathcal{K}$-preserving operations, i.e.

$$
E_{d,\,\mathrm{KP}}(\rho_{AB}) \geq I_{\mathrm{coh}}(A\rangle B)_\rho\,. \tag{S55}
$$

To establish an upper bound on $E_{d,\,\mathrm{KP}}(\rho_{AB})$, instead, we can introduce a relative entropy measure defined by [16]

$$
E_{r,\,\mathcal{K}}(\rho_{AB}) := \inf_{\sigma_{AB}\in\mathcal{K}_{AB}\cap\mathcal{D}(\mathcal{H})} D(\rho_{AB}\|\sigma_{AB})\,, \tag{S56}
$$

Since $\mathcal{K}_{AB}\otimes\mathcal{K}_{A'B'} \subseteq \mathcal{K}_{AA'BB'}$, the function $E_{r,\,\mathcal{K}}$ is sub-additive, and then Fekete's lemma [23] implies that its regularisation

$$
E_{r,\,\mathcal{K}}^{\infty}(\rho_{AB}) := \lim_{n\to\infty} \frac{1}{n} E_{r,\,\mathcal{K}}\left(\rho_{AB}^{\otimes n}\right), \tag{S57}
$$

is well defined and satisfies that $E_{r,\,\mathcal{K}}^{\infty}(\rho_{AB}) \leq E_{r,\,\mathcal{K}}(\rho_{AB})$. It does not take long to realise that $E_{r,\,\mathcal{K}}$ is monotonic under $\mathcal{K}$-preserving operations. This amounts to an elementary observation once one remembers that the relative entropy is non-increasing under the simultaneous application of any positive trace preserving map [24]. Since $E_{r,\,\mathcal{K}}$ is also asymptotically continuous [25] (see also [26, Lemma 7]), its regularisation can be shown to be an upper bound on the distillable entanglement under $\mathcal{K}$-preserving operations [27, 28]:

$$
E_{d,\,\mathrm{KP}}(\rho_{AB}) \leq E_{r,\,\mathcal{K}}^{\infty}(\rho_{AB}). \tag{S58}
$$

We are now ready to make use of the above Theorem S7 to prove irreversibility of entanglement manipulation under both non-entangling and PPT-preserving operations. To this end, according to Definition S3 (cf. (S24)) it suffices to exhibit an example of a bipartite state ρ_{AB} for which $E_{d,\mathrm{KP}}(\rho_{AB}) < E_{c,\mathrm{KP}}(\rho_{AB})$. Our candidate is a two-qutrit state, with Hilbert space $\mathcal{H}_A \otimes \mathcal{H}_B = \mathbb{C}^3 \otimes \mathbb{C}^3$. Denote the local computational basis of the two qutrits A, B with $\{|j\rangle\}_{j=1,2,3}$. Define the projector onto the maximally correlated subspace and the maximally entangled state by

$$P_3 := \sum_{j=1}^{3} |jj\rangle\langle jj| \,, \qquad |\Phi_3\rangle = \frac{1}{\sqrt{3}} \sum_{j=1}^{3} |jj\rangle \,, \qquad \Phi_3 := |\Phi_3\rangle\langle\Phi_3| \,, \tag{S59}$$

respectively. Then, construct the state

$$\omega_3 = \omega_3^{AB} := \frac{1}{2}\left(P_3 - \Phi_3\right). \tag{S60}$$

We now show the following, proving and extending Theorem 1 from the main text of the paper.

Theorem S9. *The two-qutrit state ω_3 defined by (S60) satisfies that*

$$E_{d,\mathrm{NE}}(\omega_3) = E_{d,\mathrm{PPTP}}(\omega_3) = \log_2 \frac{3}{2} \approx 0.585 \tag{S61}$$

but

$$E^{\varepsilon}_{c,\mathrm{NE}}(\omega_3) = E^{\varepsilon}_{c,\mathrm{PPTP}}(\omega_3) = 1 \tag{S62}$$

for all $\varepsilon \in [0, 1/2)$. In particular, the resource theory of entanglement is irreversible under either non-entangling or PPT-preserving operations.

Remark S10. The above result not only guarantees that the entanglement cost of the state ω_3 under non-entangling operations is 1. It also establishes a 'pretty strong' converse [29] for this value of the rate. Namely, every protocol that attempts to prepare ω_3 from entanglement bits at a rate smaller than 1 must incur an asymptotic error that is not only non-vanishing, but actually larger than a constant. This constant is $1/2$ in the current formulation of Theorem S9. However, we will see in Lemma S20 that a careful analysis actually yields a slightly larger value of $2/3$. An even stronger statement (strong converse) can be shown for distillable entanglement, where no error smaller than 1 can improve the transformation rates whatsoever. For simplicity, we have omitted these extensions from the statement of Theorem S9, and we instead refer the interested reader to Supplementary Note VI B for a more in-depth discussion of (pretty) strong converses and error-rate trade-offs.

Proof of Theorem S9. We have that

$$\log_2 \frac{3}{2} \overset{\text{(i)}}{=} I_{\mathrm{coh}}\left(A\rangle B\right)_{\omega_3} \overset{\text{(ii)}}{\leq} E_{d,\mathrm{KP}}(\omega_3) \overset{\text{(iii)}}{\leq} E^{\infty}_{r,\mathcal{K}}(\omega_3) \overset{\text{(iv)}}{\leq} E_{r,\mathcal{K}}(\omega_3) \overset{\text{(v)}}{\leq} D\left(\omega_3 \| P_3/3\right) \overset{\text{(vi)}}{=} \log_2 \frac{3}{2} \,. \tag{S63}$$

Here, (i) is an elementary computation, (ii) follows from the hashing inequality (S55), (iii) is a consequence of the upper bound on distillable entanglement in (S58), (iv) descends from the aforementioned sub-additivity of the relative entropy of $\mathcal{K}$-ness, (v) is deduced by taking as ansatz in (S56) the state $\sigma_{AB} = P_3/3 \in \mathcal{S}_{AB} \subseteq \mathcal{PPT}_{AB}$, and finally (vi) comes again from a direct calculation. This proves (S61).

As for the entanglement cost, irreversibility of entanglement under $\mathcal{K}$-preserving operations hinges on the crucial inequality $E^{\varepsilon}_{c,\mathrm{KP}}(\omega_3) \geq 1$. Hereafter, $\varepsilon \in [0, 1/2)$ is a fixed constant. Thanks to Theorem S7, it suffices to show that $N_\tau(\omega_3) \geq 2$. To this end, using the notation defined in (S59), let us consider the operator

$$X_3 := 2P_3 - 3\Phi_3 \,, \tag{S64}$$

Its eigenvalues are 2 (with multiplicity 2), 0 (with multiplicity 6) and −1 (with multiplicity 1). Since X_3 is normal (i.e. it commutes with its adjoint — in fact, X_3 is Hermitian), its operator norm coincides with the maximum modulus of an eigenvalue. Therefore,

$$\|X_3\|_\infty = 2\,. \tag{S65}$$

Calling $F_3 := \sum_{i,j=1}^3 |ij\rangle\!\langle ji|$ the swap operator, it does not take long to verify that the partial transpose of X_3 evaluates to

$$X_3^\Gamma = 2P_3 - F_3\,. \tag{S66}$$

Since X_3^Γ has eigenvalues +1 (with multiplicity 6) and −1 (with multiplicity 3),

$$\left\|X_3^\Gamma\right\|_\infty = 1\,. \tag{S67}$$

Also,

$$\operatorname{Tr} X_3\omega_3 = 2 = \|X_3\|_\infty\,. \tag{S68}$$

Thanks to (S67) and (S68), we see immediately that X is a suitable ansatz for (S33). Using it, we find that $N_\tau(\omega_3) \geq 2$. Thanks to Theorem S7, this implies that

$$E_{c,\,\mathrm{KP}}(\omega_3) \geq E_N^\tau(\omega_3) = \log_2 N_\tau(\omega_3) \geq 1\,. \tag{S69}$$

This shows that the theory of entanglement manipulation is irreversible under $\mathcal{K}$-preserving operations, i.e. under either non-entangling or PPT-preserving operations.

For completeness we now show that the inequalities in (S69) are in fact all tight; this will establish (S62) and conclude the proof. Start by observing that

$$E_{c,\,\mathrm{PPTP}}^\varepsilon(\omega_3) \overset{\text{(vii)}}{\leq} E_{c,\,\mathrm{NE}}^\varepsilon(\omega_3) \overset{\text{(viii)}}{\leq} E_{c,\,\mathrm{NE}}^{\mathrm{exact}}(\omega_3)\,, \tag{S70}$$

where (vii) follows from Lemma S4, while (viii) is an application of the elementary inequality (S24). We now argue that $E_{c,\,\mathrm{NE}}^{\mathrm{exact}}(\omega_3) \leq 1$, by providing an explicit example of a non-entangling operation Λ from a two-qubit to a two-qutrit system such that $\Lambda(\Phi_2) = \omega_3$. Construct

$$\Lambda(X) := \operatorname{Tr}[X\Phi_2]\omega_3 + \operatorname{Tr}\left[X(\mathbb{1} - \Phi_2)\right]\tau_3\,, \tag{S71}$$

where $\tau_3 := \frac{\mathbb{1}-P_3}{6} = \frac{1}{6}\sum_{j\neq k} |jk\rangle\!\langle jk|$. Since $\{\operatorname{Tr}[\sigma\Phi_2] : \sigma \in \mathcal{S}\cap\mathcal{D}(\mathcal{H})\} = [0, 1/2]$, to show that Λ is non-entangling it suffices to prove that $\lambda\omega_3 + (1-\lambda)\tau_3 \in \mathcal{S}$ for all $\lambda \in [0, 1/2]$. Since the claim is trivial for $\lambda = 0$, because τ_3 is manifestly separable, by convexity it suffices to prove it for $\lambda = 1/2$. Let us write

$$\frac{1}{2}(\omega_3 + \tau_3) = \frac{1}{12}\mathbb{1} + \frac{1}{6}P_3 - \frac{1}{4}\Phi_3 = \mathcal{P}\left(|+\rangle\!\langle+| \otimes |-\rangle\!\langle-|\right), \tag{S72}$$

where $|\pm\rangle := \frac{1}{\sqrt{2}}(|1\rangle \pm |2\rangle)$, and $\mathcal{P}$ is the non-entangling quantum operation defined by

$$\begin{aligned} \mathcal{P}(X) &:= \frac{1}{6}\sum_{\pi\in S_3}\int_0^{2\pi} \left(U_{\theta,\pi}\otimes U_{-\theta,\pi}\right) X \left(U_{\theta,\pi}\otimes U_{-\theta,\pi}\right)^\dagger \frac{\mathrm{d}^3\theta}{(2\pi)^3}\,, \\ U_{\theta,\pi} &:= \sum_{j=1}^3 e^{i\theta_j}\,|\pi(j)\rangle\!\langle j|\,. \end{aligned} \tag{S73}$$

with S_3 denoting the symmetric group over a set of 3 elements. Note that the last equality in (S72), which can be proved by inspection using the representation in (S73), amounts to the sought separable decomposition of the state $\frac{1}{2}(\omega_3 + \tau_3)$. This establishes that $E_{c,\,\mathrm{PPTP}}^\varepsilon(\omega_3) \leq E_{c,\,\mathrm{NE}}^\varepsilon(\omega_3) \leq 1$ and concludes the proof. □

Remark S11. One can wonder what other types of states cannot be reversibility manipulated. This is far from obvious, since the axiomatic classes of operations NE or PPTP are typically much more powerful than previously employed types of transformations; in particular, several types of states which have been used to show irreversibility in specific settings are actually *reversible* under NE or PPTP transformations.

The prime example of this is the antisymmetric state, defined on a bipartite system with Hilbert space $\mathbb{C}^d \otimes \mathbb{C}^d$ by

$$\alpha_d := \frac{\mathbb{1} - F}{d(d-1)}, \tag{S74}$$

where $F := \sum_{i,j=1}^d |ij\rangle\!\langle ji|$ is the flip operator. This state gained fame as the 'universal counterexample' which violates many properties obeyed by other types of quantum states [30]: for example, it is known that its manipulation is highly irreversible under LOCC — its distillable entanglement is of order $1/d$, while its entanglement cost is lower bounded by a d-independent non-zero constant. [31]. Curiously, however, α_d was also the first example of a mixed state whose manipulation is reversible under all PPT operations [32] — these transformations (hereafter simply denoted with PPT) are all maps Λ such that $\mathrm{id}_R \otimes \Lambda$ is PPT-preserving for all ancillary systems R [33], and are therefore a strict subset of the PPT-preserving operations considered herein. The reason why reversibility can be achieved in this setting is that the entanglement cost of α_d can be significantly lowered by considering PPT transformations instead of LOCC, allowing it to reach order $1/d$.

However, in Ref. [21], a related class of states supported on the asymmetric subspace was used to show the *ir*reversibility of entanglement manipulation under PPT operations. In particular, for the state $\rho_p := p\,|v_1\rangle\!\langle v_1| + (1-p)\,|v_2\rangle\!\langle v_2|$ with $|v_1\rangle = (|01\rangle - |10\rangle)/\sqrt{2}$ and $|v_2\rangle = (|02\rangle - |20\rangle)/\sqrt{2}$, it was shown that $E_{d,\mathrm{PPT}}(\rho_p) < 1 = E_{c,\mathrm{PPT}}(\rho_p)$. One might then wonder if these states could serve as a similar example of irreversibility for the larger class of PPT-preserving operations. However, this cannot be the case. To see this, we can use the fact that the quantity E_η considered in [21] constitutes a lower bound on the distillable entanglement $E_{d,\mathrm{PPTP}}$, but already in [21] it was shown that $E_\eta(\rho_p) = 1$, meaning that $E_{d,\mathrm{PPTP}}(\rho_p) = 1$ and this state is actually reversible under PPT-preserving maps.

In a way, this suggests that the state ω_3 is somewhat special, since its entanglement cost cannot be brought down low enough to match its distillable entanglement, even if we allow the extended classes of operations NE or PPTP. It would be interesting to study in more detail the special properties of ω_3 which induce this behaviour, and to understand exactly what types of states exhibit irreversibility in entanglement manipulation under NE and PPTP.

Remark S12. The proof of Theorem S9 actually allows us to compute also the zero-error costs of ω_3, namely

$$E_{c,\mathrm{NE}}^{\mathrm{exact}}(\omega_3) = E_{c,\mathrm{PPTP}}^{\mathrm{exact}}(\omega_3) = 1\,. \tag{S75}$$

As it turns out, the same entanglement cost of exact preparation of 1 can be achieved by means of a strict subset of PPT-preserving operations, namely, the aforementioned PPT operations. In fact, already the result by Audenaert et al. [32] guarantees that $E_{c,\mathrm{PPT}}^{\mathrm{exact}}(\omega_3) = E_N(\omega_3) = 1$, where E_N is the logarithmic negativity (S10), because ω_3 has vanishing 'binegativity',[2] as a straightforward check reveals. We can arrive at the same conclusion thanks to the complete characterisation of the exact PPT entanglement cost recently proposed by Wang and Wilde [34].

IV. HOW MUCH ENTANGLEMENT MUST BE GENERATED TO ACHIEVE REVERSIBILITY?

We first recall the framework and the claimed results of Brandão and Plenio [10] in detail. To begin, we need to fix some notation. For two bipartite quantum systems AB, $A'B'$, a given

[2] This just means that $\left|\omega_3^\Gamma\right|^\Gamma \geq 0$, where $|X| := \sqrt{X^\dagger X}$ is the operator absolute value.

non-negative function $M : \mathcal{D}(\mathcal{H}_{A'B'}) \to \mathbb{R}_+ \cup \{+\infty\}$ on the set of states on $A'B'$ that vanishes on $\mathcal{K}_{A'B'} \cap \mathcal{D}(\mathcal{H}_{A'B'})$, and some $\delta \geq 0$, we define the set of (M, δ)-approximately $\mathcal{K}$-preserving maps by

$$\mathrm{KP}_\delta^M (AB \to A'B') := \{\Lambda \in \mathrm{PTP}(AB \to A'B') : \ M(\Lambda(\sigma_{AB})) \leq \delta \ \ \forall \sigma_{AB} \in \mathcal{K}_{AB} \cap \mathcal{D}(\mathcal{H}_{AB})\} . \tag{S76}$$

Typically, M will be chosen to be an entanglement measure [16, 35]. In what follows, we will in fact assume that M is actually a family of functions defined on each bipartite quantum system. Given a sequence $(\delta_n)_{n\in\mathbb{N}}$ with $\delta_n \geq 0$ for all n, one can then define the distillable entanglement and entanglement cost under **$(M, (\delta_n)_n)$-approximately $\mathcal{K}$-preserving maps** by setting

$$E^{\varepsilon}_{d,\mathrm{KP}^M_{(\delta_n)}}(\rho_{AB}) := \sup \left\{ R > 0 : \ \limsup_{n\to\infty} \inf_{\Lambda_n \in \mathrm{KP}^M_{\delta_n}\left(A^nB^n \to A_0^{\lceil Rn \rceil} B_0^{\lceil Rn \rceil}\right)} \frac{1}{2} \left\| \Lambda_n\left(\rho_{AB}^{\otimes n}\right) - \Phi_2^{\otimes \lceil Rn \rceil} \right\|_1 \leq \varepsilon \right\} \tag{S77}$$

$$E^{\varepsilon}_{c,\mathrm{KP}^M_{(\delta_n)}}(\rho_{AB}) := \inf \left\{ R > 0 : \ \limsup_{n\to\infty} \inf_{\Lambda_n \in \mathrm{KP}^M_{\delta_n}\left(A_0^{\lfloor Rn \rfloor} B_0^{\lfloor Rn \rfloor} \to A^nB^n\right)} \frac{1}{2} \left\| \Lambda_n\left(\Phi_2^{\otimes \lfloor Rn \rfloor}\right) - \rho_{AB}^{\otimes n} \right\|_1 \leq \varepsilon \right\} . \tag{S78}$$

We also set $E_{d,\mathrm{KP}^M_{(\delta_n)}} := E^0_{d,\mathrm{KP}^M_{(\delta_n)}}$ and $E_{c,\mathrm{KP}^M_{(\delta_n)}} := E^0_{c,\mathrm{KP}^M_{(\delta_n)}}$. With this notation, we say that the theory of entanglement manipulation is reversible under $(M, (\delta_n)_n)$-approximately $\mathcal{K}$-preserving operations if it holds that $E_{d,\mathrm{KP}^M_{(\delta_n)}}(\rho_{AB}) = E_{c,\mathrm{KP}^M_{(\delta_n)}}(\rho_{AB})$ for all states ρ_{AB} on all bipartite quantum systems AB. We also say that the theory of entanglement manipulation is reversible under ***M*-asymptotically $\mathcal{K}$-preserving operations** if for all states ρ_{AB} there exists a sequence $(\delta_n)_{n\in\mathbb{N}}$ such that $\delta_n \xrightarrow[n\to\infty]{} 0$ and $E_{d,\mathrm{KP}^M_{(\delta_n)}}(\rho_{AB}) = E_{c,\mathrm{KP}^M_{(\delta_n)}}(\rho_{AB})$.

To state Brandão and Plenio's conjecture in this framework, we first need to introduce another entanglement monotone closely related to the standard robustness. Recalling first the definition (S11) of $R^s_{\mathcal{K}}$, namely, $R^s_{\mathcal{K}}(\rho) = \inf\{\mathrm{Tr}\,\delta : \delta \in \mathcal{K}, \ \rho + \delta \in \mathcal{K}\}$, the ***generalised robustness*** (or *global robustness*) [5, 36, 37] is defined similarly as

$$R^g_{\mathcal{K}}(\rho) = \inf\{\mathrm{Tr}\,\delta : \ \delta \geq 0, \ \rho + \delta \in \mathcal{K}\} . \tag{S79}$$

It is also an entanglement monotone, and many similarities between the two robustness measures have been found; for example, for any pure state Ψ it holds that $R^s_{\mathcal{K}}(\Psi) = R^g_{\mathcal{K}}(\Psi)$ [6, 7, 36, 37]. The two can, however, exhibit very different properties, as we will explicitly demonstrate below (see also Supplementary Note VI).

With this language, Brandão and Plenio's claim is that entanglement becomes reversible under $R^g_{\mathcal{S}}$-asymptotically $\mathcal{S}$-preserving maps.[3] Employing the simplified notation $\mathrm{KP}^g_{(\delta_n)} := \mathrm{KP}^{R^g_{\mathcal{S}}}_{(\delta_n)}$, we formalise their claim as follows.

Conjecture S13 (Reversibility under asymptotically non-entangling operations [10]). *For any state ρ_{AB} acting on a finite-dimensional Hilbert space, there exists a sequence $(\delta_n)_{n\in\mathbb{N}}$ such that $\lim_{n\to\infty} \delta_n = 0$ and*

$$E_{d,\mathrm{KP}^g_{(\delta_n)}}(\rho_{AB}) = E_{c,\mathrm{KP}^g_{(\delta_n)}}(\rho_{AB}) = E^\infty_{r,\mathcal{K}}(\rho_{AB}) , \tag{S80}$$

where $E^\infty_{r,\mathcal{K}}$ is the regularised relative entropy measure defined by (S57).

[3] We bring to the reader's attention the recently discovered technical issues underlying the proof of the main result of [10], as detailed in [38]. For this reason, we state the result here as a 'conjecture', and its validity is an open question. We nevertheless find it useful to discuss the result here in detail as we take a conceptual inspiration from the framework of [10]. Our findings are independent of whether this result is ultimately found to be correct or not.

Brandão and Plenio argue that the above means that entanglement can be reversibly interconverted without generating macroscopic amounts of it, since the supplemented entanglement is constrained by δ_n which vanishes in the asymptotic limit. This is certainly true if one quantifies entanglement with the generalised robustness. However, this is an a priori arbitrary choice: one could analogously choose the standard robustness $R^s_{\mathcal{K}}$ as a quantifier, and consider the (sequence of) sets of operations $\mathrm{KP}^s_{(\delta_n)} := \mathrm{KP}^{R^s_{\mathcal{S}}}_{(\delta_n)}$ defined by (S76) for the special case $M = R^s_{\mathcal{S}}$. Alternatively, when the negativity $N(\rho) := \frac{1}{2}\left(\left\|\rho^\Gamma\right\|_1 - 1\right)$ is used as the entanglement measure, we can instead look at the operations $\mathrm{KP}^N_{(\delta_n)}$. Recalling that $R^s_{\mathcal{K}}(\rho) \geq \frac{1}{2}\left(\left\|\rho^\Gamma\right\|_1 - 1\right) = N(\rho)$, this choice of definition ensures that $\mathrm{KP}^s_\delta \subseteq \mathrm{KP}^N_\delta$.

An extension of our result is then as follows.

Theorem S14. *For any sequence $(\delta_n)_n$ such that $\delta_n = 2^{o(n)}$, the two-qutrit state ω_3 defined by (S60) satisfies that*

$$\begin{aligned} E_{d,\mathrm{NE}^s_{(\delta_n)}}(\omega_3) = E_{d,\mathrm{PPTP}^s_{(\delta_n)}}(\omega_3) = E_{d,\mathrm{NE}^N_{(\delta_n)}}(\omega_3) = E_{d,\mathrm{PPTP}^N_{(\delta_n)}}(\omega_3) \\ = \log_2 \frac{3}{2} \approx 0.585 \end{aligned} \tag{S81}$$

but

$$\begin{aligned} E^\varepsilon_{c,\mathrm{NE}^s_{(\delta_n)}}(\omega_3) = E^\varepsilon_{c,\mathrm{PPTP}^s_{(\delta_n)}}(\omega_3) = E^\varepsilon_{c,\mathrm{NE}^N_{(\delta_n)}}(\omega_3) = E^\varepsilon_{c,\mathrm{PPTP}^N_{(\delta_n)}}(\omega_3) \\ = 1 \end{aligned} \tag{S82}$$

for all $\varepsilon \in [0, 1/2)$. In particular, the resource theory of entanglement is irreversible under any class of operations which does not generate an amount of entanglement that grows exponentially in n, as quantified by either the standard robustness $R^s_{\mathcal{K}}$ or by the negativity N.

Here $\delta_n = 2^{o(n)}$ means that δ_n has a sub-exponential behaviour in n: for any $k > 0$, there exists $m \in \mathbb{N}$ such that $\delta_n < 2^{kn}$ for all $n \geq m$. Consequently, to have any hope of recovering reversibility, one needs δ_n (and hence the generated entanglement) to grow exponentially: there must exist a choice of $k > 0$ such that, for all $m \in \mathbb{N}$, $\delta_n \geq 2^{kn}$ for at least one $n \geq m$; in other words, δ_n is lower bounded by 2^{kn} infinitely often.[4]

Remark. One can wonder whether $R^g_{\mathcal{K}}$ can be considered as a more operationally meaningful measure of the supplemented entanglement, justifying its use over other measures such as $R^s_{\mathcal{K}}$ or N and thus substantiating the reversibility conjecture of [10] over the irreversibility result of Theorem S14. We do not believe that there is any compelling reason to do so: although $R^g_{\mathcal{K}}$ admits a very general operational interpretation as the quantifier of the advantage that a given state provides in channel discrimination tasks [7, 41, 42], $R^s_{\mathcal{K}}$ has an arguably even more relevant application, as it exactly quantifies the one-shot entanglement cost under non-entangling operations [43]. On the technical side, both of the quantities suffer from very similar issues in the many-copy limit, as they do not satisfy asymptotic continuity [25].

This is no coincidence, as Brandão and Plenio have shown that the choice of an asymptotically continuous monotone to quantify the supplemented entanglement leads to the trivialisation of the framework [10, Section V]. Note that almost all the most commonly used entanglement measures and all of those with the strongest operational meanings are in fact asymptotically continuous. Examples include the entanglement of formation [26, 44], the (LOCC) entanglement cost [26], the squashed entanglement [18, 45], and the (regularised) relative entropy of entanglement [25, 26]. In fact, among the most widely used entanglement measures, the only one that is *not* asymptotically

[4] Following the original notation introduced by Hardy and Littlewood [39], we could denote this behaviour as $\delta_n = 2^{\Omega(n)}$. However, the commonly used notation $\Omega(n)$ actually refers to a stronger property [40], so we do not use it here.

continuous is the logarithmic negativity [3, 4]. For this reason, we regard the failure of asymptotic continuity for the robustnesses as an issue of some conceptual importance, one that may cast some doubts on the status of approximately $\mathcal{K}$-preserving maps.

The choice of $R^g_{\mathcal{K}}$ in [10] is motivated *a posteriori* by the fact that it is claimed to lead to reversibility, rather than by *a prori* physical considerations. We are therefore inclined to believe that there is *no* unique and indisputable choice of a suitable entanglement measure, and we consider Theorem S14 to serve as evidence that the irreversibility of entanglement revealed in our work is very robust, and that avoiding it requires a very careful and deliberate choice of an entanglement monotone — according to other, equally reasonable choices, the generated entanglement must be exponentially large.

Remark. We should also note in passing that between the two sets of operations $\mathrm{NE}^s_{(\delta_n)}$ and $\mathrm{NE}^N_{(\delta_n)}$ that we considered in Theorem S14 above, the former is perhaps more adherent to our intuitive notion of approximately non-entangling maps. Indeed, since the standard robustness of entanglement $R^s_{\mathcal{S}}$ is a faithful measure, i.e. it is strictly positive on all entangled states, transformations in NE^s_0 are in fact non-entangling. Transformations in NE^N_0, on the contrary, map separable states to PPT states that can very well be entangled. However, since $\mathrm{NE}^s_\delta \subseteq \mathrm{NE}^N_\delta$, showing the irreversibility of entanglement under the operations NE^N_δ constitutes a strictly stronger result, and indeed shows also that generating PPT entangled states is not sufficient to recover reversibility — any reversible protocol must create highly non-PPT entanglement.

The first step in proving the Theorem is the following lemma, which establishes an approximate monotonicity of $R^s_{\mathcal{K}}$ under approximately $\mathcal{K}$-preserving maps, whether quantified by $R^s_{\mathcal{K}}$ itself or by the negativity.

Lemma S15. *For any $\Lambda \in \mathrm{KP}^s_\delta(AB \to A'B')$, it holds that*

$$R^s_{\mathcal{K}}(\Lambda(\rho_{AB})) + 1 \leq (1+2\delta)\,(R^s_{\mathcal{K}}(\rho_{AB}) + 1). \tag{S83}$$

Similarly, for any $\Lambda \in \mathrm{KP}^N_\delta(AB \to A'B')$, it holds that

$$\frac{1}{2}\left(\left\|\Lambda(\rho_{AB})^\Gamma\right\|_1 + 1\right) \leq (1+2\delta)\,(R^s_{\mathcal{K}}(\rho_{AB}) + 1). \tag{S84}$$

Proof. Let us take $\Lambda \in \mathrm{KP}^s_\delta(AB \to A'B')$ and consider any feasible decomposition for the standard robustness of ρ as $\rho = \sigma - \tau$ where $\sigma, \tau \in \mathcal{K}$ (noting that these are in general only unnormalised states). Since $R^s_{\mathcal{K}}\left(\frac{\Lambda(\sigma)}{\operatorname{Tr}\sigma}\right) \leq \delta$ and $R^s_{\mathcal{K}}\left(\frac{\Lambda(\tau)}{\operatorname{Tr}\tau}\right) \leq \delta$, for any $\varepsilon > 0$ there exist decompositions

$$\frac{\Lambda(\sigma)}{\operatorname{Tr}\sigma} = \sigma' - \tau', \qquad \frac{\Lambda(\tau)}{\operatorname{Tr}\tau} = \sigma'' - \tau'', \tag{S85}$$

for some $\sigma', \sigma'', \tau', \tau'' \in \mathcal{K}$ such that $\operatorname{Tr}\tau', \operatorname{Tr}\tau'' \leq \delta + \varepsilon$. Then

$$\Lambda(\rho) = [(\operatorname{Tr}\sigma)\sigma' + (\operatorname{Tr}\tau)\tau''] - [(\operatorname{Tr}\sigma)\tau' + (\operatorname{Tr}\tau)\sigma''] \,. \tag{S86}$$

This constitutes a valid feasible solution for the robustness of $\Lambda(\rho)$, giving

$$\begin{aligned} R^s_{\mathcal{K}}(\Lambda(\rho)) + 1 &\leq \operatorname{Tr}\sigma \operatorname{Tr}\sigma' + \operatorname{Tr}\tau \operatorname{Tr}\tau'' \\ &= (\operatorname{Tr}\tau + 1)(\operatorname{Tr}\tau' + 1) + \operatorname{Tr}\tau \operatorname{Tr}\tau'' \\ &\leq (\operatorname{Tr}\tau + 1)(\operatorname{Tr}\tau' + 1) + (\operatorname{Tr}\tau + 1)\operatorname{Tr}\tau'' \\ &\leq (\operatorname{Tr}\tau + 1)(\delta + \varepsilon + 1) + (\operatorname{Tr}\tau + 1)(\delta + \varepsilon). \end{aligned} \tag{S87}$$

Since this holds for any feasible value of $\operatorname{Tr}\tau$, it must also hold that $R^s_{\mathcal{K}}(\Lambda(\rho)) + 1 \leq (1 + 2\delta + 2\varepsilon)(R^s_{\mathcal{K}}(\rho) + 1)$, as $R^s_{\mathcal{K}}$ is defined precisely as the infimum of all feasible values of $\operatorname{Tr}\tau$. Since $\varepsilon > 0$ was arbitrary, the desired statement follows.

The case of $\Lambda \in \mathrm{KP}_\delta^N$ is similar. For any decomposition $\rho = \sigma - \tau$ with $\sigma, \tau \in \mathcal{K}$, the triangle inequality gives

$$\begin{aligned}\left\|\Lambda(\rho)^\Gamma\right\|_1 &\leq \operatorname{Tr}\sigma \left\|\frac{\Lambda(\sigma)^\Gamma}{\operatorname{Tr}\sigma}\right\|_1 + \operatorname{Tr}\tau \left\|\frac{\Lambda(\tau)^\Gamma}{\operatorname{Tr}\tau}\right\|_1 \\ &\leq (\operatorname{Tr}\sigma)(1+2\delta) + (\operatorname{Tr}\tau)(1+2\delta) \\ &= (1 + 2\operatorname{Tr}\tau)(1+2\delta)\end{aligned} \tag{S88}$$

where we used that $\frac{1}{2}\left(\left\|\Lambda(\sigma)^\Gamma\right\|_1 - 1\right) \leq \delta$ and analogously for τ. Rearranging, we get

$$\begin{aligned}\frac{1}{2}\left(\left\|\Lambda(\rho)^\Gamma\right\|_1 + 1\right) &\leq \delta + 2\delta\operatorname{Tr}\tau + \operatorname{Tr}\tau + 1 \\ &\leq (1+2\delta)(\operatorname{Tr}\tau + 1).\end{aligned} \tag{S89}$$

Minimising over all feasible values of $\operatorname{Tr}\tau$ gives $\frac{1}{2}\left(\left\|\Lambda(\rho)^\Gamma\right\|_1 + 1\right) \leq (1+2\delta)(R^s_{\mathcal{K}}(\rho)+1)$, as was to be shown. □

Theorem S14 then relies on the following extension of Theorem S7.

Theorem S16. *For $\mathcal{K} = \mathcal{S}$ or $\mathcal{K} = \mathcal{PPT}$, the entanglement cost under approximately $\mathcal{K}$-preserving operations satisfies that*

$$\inf_{\varepsilon \in [0,1/2)} E^\varepsilon_{c,\mathrm{KP}^s_{(\delta_n)}}(\rho) \geq L^\tau_{\mathcal{K}}(\rho) \geq E^\tau_N(\rho) \tag{S90}$$

and

$$\inf_{\varepsilon \in [0,1/2)} E^\varepsilon_{c,\mathrm{KP}^N_{(\delta_n)}}(\rho) \geq E^\tau_N(\rho) \tag{S91}$$

for any sequence $(\delta_n)_n$ such that $\delta_n = 2^{o(n)}$.

Proof. In complete analogy with the proof of Theorem S7, for any feasible sequence $(\Lambda_n)_{n\in\mathbb{N}}$ of maps such that $\Lambda_n \in \mathrm{KP}^s_{\delta_n}$ and $\left\|\Lambda_n(\Phi_2^{\otimes\lfloor Rn\rfloor}) - \rho^{\otimes n}\right\|_1 =: \varepsilon_n \xrightarrow[n\to\infty]{} \varepsilon < 1/2$, we can write

$$\begin{aligned}2^{\lfloor Rn\rfloor} &= 1 + R^s_{\mathcal{K}}\left(\Phi_2^{\otimes\lfloor Rn\rfloor}\right) \\ &\geq (1+2\delta_n)^{-1}\left(1 + R^s_{\mathcal{K}}\left(\Lambda_n\left(\Phi_2^{\otimes\lfloor Rn\rfloor}\right)\right)\right) \\ &\geq (1+2\delta_n)^{-1}(1-2\varepsilon_n)\left(1 + R^\tau_{\mathcal{K}}\left(\rho^{\otimes n}\right)\right)\end{aligned} \tag{S92}$$

where now we used Lemma S15 to incorporate the approximate entanglement non-generation. Then

$$\begin{aligned}R &= \lim_{n\to\infty}\frac{\lfloor Rn\rfloor}{n} \\ &\geq \limsup_{n\to\infty}\frac{1}{n}\log_2\left(1 + R^\tau_{\mathcal{K}}\left(\rho^{\otimes n}\right)\right) + \liminf_{n\to\infty}\frac{1}{n}\log_2(1-2\varepsilon_n) - \limsup_{n\to\infty}\frac{1}{n}\log_2(1+2\delta_n) \\ &= L^\tau_{\mathcal{K}}(\rho) - \limsup_{n\to\infty}\frac{1}{n}\log_2(1+2\delta_n) \\ &= L^\tau_{\mathcal{K}}(\rho)\,,\end{aligned} \tag{S93}$$

where in the last line we used the fact that

$$\lim_{n\to\infty}\frac{\log_2\delta_n}{n} = 0 \tag{S94}$$

by hypothesis. The rest of the proof of the first part of the Theorem is then exactly the same as in Theorem S7.

The second part of the proof is very similar in that it follows Theorem S7, but it goes directly to the tempered negativity N_τ rather than the intermediate quantity $L^\tau_{\mathcal{K}}$. Taking now any feasible sequence $(\Lambda_n)_{n\in\mathbb{N}}$ with $\Lambda_n \in \mathrm{KP}^N_{\delta_n}$, we have

$$\begin{aligned} 2^{\lfloor Rn \rfloor} &= 1 + R^s_{\mathcal{K}}\left(\Phi_2^{\otimes \lfloor Rn \rfloor}\right) \\ &\overset{\text{(i)}}{\geq} (1+2\delta_n)^{-1} \frac{1 + \left\| \Lambda_n\left(\Phi_2^{\otimes \lfloor Rn \rfloor}\right)^\Gamma \right\|_1}{2} \\ &\overset{\text{(ii)}}{\geq} (1+2\delta_n)^{-1} \frac{1 + N_\tau\left(\Lambda_n\left(\Phi_2^{\otimes \lfloor Rn \rfloor}\right) \middle| \rho^{\otimes n}\right)}{2} \\ &\overset{\text{(iii)}}{\geq} (1+2\delta_n)^{-1}(1-2\varepsilon_n)\frac{1 + N_\tau\left(\rho^{\otimes n}\right)}{2}, \end{aligned} \tag{S95}$$

where in (i) we used Lemma S15, in (ii) Proposition S5(b), and in (iii) the ε-lemma (Lemma S6). This gives

$$\begin{aligned} R &= \lim_{n\to\infty} \frac{\lfloor Rn \rfloor}{n} \\ &\geq \limsup_{n\to\infty} \frac{1}{n}\log_2 \frac{1+N_\tau\left(\rho^{\otimes n}\right)}{2} + \liminf_{n\to\infty} \frac{1}{n}\log_2(1-2\varepsilon_n) - \limsup_{n\to\infty}\frac{1}{n}\log_2(1+2\delta_n) \\ &\geq \limsup_{n\to\infty} \left(\frac{1}{n}\log_2 N_\tau(\rho^{\otimes n}) - \frac{1}{n}\right) - \limsup_{n\to\infty}\frac{1}{n}\log_2 \delta_n \\ &\geq \limsup_{n\to\infty} \left(\frac{1}{n}\log_2 N_\tau(\rho)^n - \frac{1}{n}\right) \\ &= E^\tau_N(\rho) \end{aligned} \tag{S96}$$

using the super-multiplicativity of the tempered negativity (Proposition S5(e)) and the assumption that $\delta_n = 2^{o(n)}$. □

As the final ingredient that will be required in the proof of Theorem S14, we need to show that the distillable entanglement cannot increase even if we allow sub-exponential entanglement generation.

Lemma S17. *Consider any state ρ_{AB} and let $\mathcal{K} = \mathcal{S}$ or $\mathcal{K} = \mathcal{PPT}$. For any $\varepsilon \in [0,1)$ and any non-negative sequence $(\delta_n)_n$ it holds that*

$$E^\varepsilon_{d,\mathrm{KP}^s_{(\delta_n)}}(\rho) = E^\varepsilon_{d,\mathrm{KP}^N_{(\delta_n)}}(\rho) = E^\varepsilon_{d,\mathrm{KP}^g_{(\delta_n)}}(\rho)\,. \tag{S97}$$

Moreover, if $\delta_n = 2^{o(n)}$ then also

$$E^\varepsilon_{d,\mathrm{KP}}(\rho) = E^\varepsilon_{d,\mathrm{KP}^s_{(\delta_n)}}(\rho) = E^\varepsilon_{d,\mathrm{KP}^N_{(\delta_n)}}(\rho) = E^\varepsilon_{d,\mathrm{KP}^g_{(\delta_n)}}(\rho)\,. \tag{S98}$$

Proof. The proof will proceed in two steps. First, we establish expressions for the minimal error achievable in distillation with (M,δ)-approximately $\mathcal{K}$-preserving operations. Then, we argue that, for sub-exponential δ_n, the contributions from the parameter δ_n to the performance of the distillation task can be absorbed into the transformation rates, effectively preventing any improvement in the asymptotic distillation error.

Consider first any operation $\Lambda \in \mathrm{KP}^g_\delta\left(A^nB^n \to A_0^m B_0^m\right)$ for a fixed δ, where m is a generic positive integer. We would then like to understand exactly the error in the transformation from

$\rho^{\otimes n}$ to the maximally entangled state at some rate R, which we will for now quantify using the fidelity $F(\omega,\tau) := \|\sqrt{\omega}\sqrt{\tau}\|_1^2$. We have

$$\begin{aligned} F\left(\Lambda(\rho^{\otimes n}), \Phi_2^{\otimes m}\right) &= \operatorname{Tr} \Lambda\left(\rho^{\otimes n}\right) \Phi_2^{\otimes m} \\ &= \operatorname{Tr} \rho^{\otimes n} \Lambda^\dagger\left(\Phi_2^{\otimes m}\right) \end{aligned} \tag{S99}$$

using that Φ_2 is a pure state. Notice now that, by definition of the generalised robustness, for any $\sigma \in \mathcal{K} \cap \mathcal{D}(\mathcal{H})$ we have $\Lambda(\sigma) \leq (1+\delta)\sigma'$ for some $\sigma' \in \mathcal{K} \cap \mathcal{D}(\mathcal{H})$. Thus, for any $\sigma \in \mathcal{K} \cap \mathcal{D}(\mathcal{H})$,

$$\begin{aligned} \operatorname{Tr} \Lambda^\dagger\left(\Phi_2^{\otimes m}\right)\sigma &\leq \operatorname{Tr} \Phi_2^{\otimes m}(1+\delta)\sigma' \\ &\leq (1+\delta)\frac{1}{2^m} \end{aligned} \tag{S100}$$

where in the first line we used the positivity of Φ_2, and in the second that the maximal overlap of $\Phi_2^{\otimes m}$ with a separable state is $\frac{1}{2^m}$ [46]. Noting also that $0 \leq \Lambda^\dagger\left(\Phi_2^{\otimes m}\right) \leq \mathbb{1}$ due to the fact that Λ is positive and trace preserving, we get

$$\begin{aligned} \sup_{\Lambda \in \mathrm{KP}_\delta^g} F\left(\Lambda(\rho^{\otimes n}), \Phi_2^{\otimes m}\right) &= \sup_{\Lambda \in \mathrm{KP}_\delta^g} \operatorname{Tr} \rho \Lambda^\dagger\left(\Phi_2^{\otimes m}\right) \\ &\leq \sup \left\{ \operatorname{Tr} \rho^{\otimes n} W \;:\; 0 \leq W \leq \mathbb{1},\ \operatorname{Tr} W\sigma \leq (1+\delta)\frac{1}{2^m}\ \ \forall \sigma \in \mathcal{K} \cap \mathcal{D}(\mathcal{H}) \right\} \\ &=: \varphi_{n,m}(\delta)\,. \end{aligned} \tag{S101}$$

The argument for operations KP_δ^N, where now negativity is the figure of merit, proceeds analogously. We have, for any $\sigma \in \mathcal{K} \cap \mathcal{D}(\mathcal{H})$ and any $\Lambda \in \mathrm{KP}_\delta^N$, that

$$\begin{aligned} \operatorname{Tr} \Lambda^\dagger\left(\Phi_2^{\otimes m}\right)\sigma &= \operatorname{Tr} \left(\Phi_2^{\otimes m}\right)^\Gamma \Lambda(\sigma)^\Gamma \\ &\leq \left\| \left(\Phi_2^{\otimes m}\right)^\Gamma \right\|_\infty \left\|\Lambda(\sigma)^\Gamma\right\|_1 \\ &\leq \frac{1}{2^m}(1+2\delta) \end{aligned} \tag{S102}$$

where in the second line we used the Cauchy–Schwarz inequality, and in the third we used that $\frac{1}{2}\left(\left\|\Lambda(\sigma)^\Gamma\right\| - 1\right) \leq \delta$ and that $\left\|\Phi_2^\Gamma\right\|_\infty = \frac{1}{2}$. This gives

$$\begin{aligned} \sup_{\Lambda \in \mathrm{KP}_\delta^N} F\left(\Lambda(\rho^{\otimes n}), \Phi_2^{\otimes m}\right) &\leq \sup \left\{ \operatorname{Tr} \rho^{\otimes n} W \;:\; 0 \leq W \leq \mathbb{1},\ \operatorname{Tr} W\sigma \leq (1+2\delta)\frac{1}{2^m}\ \ \forall \sigma \in \mathcal{K} \cap \mathcal{D}(\mathcal{H}) \right\} \\ &= \varphi_{n,m}(2\delta)\,, \end{aligned} \tag{S103}$$

exactly the same as in (S101) up to the substitution $\delta \mapsto 2\delta$, which we will see to be immaterial.

For the other direction, define the separable [9] state $\tau_m := (\mathbb{1} - \Phi_2^{\otimes m})/(4^m - 1) \in \mathcal{K} \cap \mathcal{D}(\mathcal{H})$, which satisfies also that [9] $\Phi_2^{\otimes m} + (2^m - 1)\tau_m \in \mathcal{K}$. Note that $R^s_{\mathcal{K}}\left(\Phi_2^{\otimes m}\right) = 2^m - 1$ by (S16), and therefore τ_m is the state that achieves the minimum in the definition of $R^s_{\mathcal{K}}$ (S11) for $\Phi_2^{\otimes m}$. Take any operator W such that $0 \leq W \leq \mathbb{1}$ and $\operatorname{Tr} W\sigma \leq (1+\delta)\frac{1}{2^m}$ for all $\sigma \in \mathcal{K} \cap \mathcal{D}(\mathcal{H})$, where we assume that $(1+\delta)\frac{1}{2^m} \leq 1$ without loss of generality. Define the map Γ_W by

$$\Gamma_W(X) = (\operatorname{Tr} WX)\, \Phi_2^{\otimes m} + (\operatorname{Tr}(\mathbb{1} - W)X)\, \tau_m\,. \tag{S104}$$

This map is explicitly completely positive and trace preserving, and we can furthermore verify that, since $0 \leq \operatorname{Tr} W\sigma \leq (1+\delta)\frac{1}{2^m}$, for any $\sigma \in \mathcal{K} \cap \mathcal{D}(\mathcal{H})$ we have

$$\Gamma_W(\sigma) \in \operatorname{conv} \left\{ \tau_m,\ (1+\delta)\frac{1}{2^m}\, \Phi_2^{\otimes m} + \left(1 - (1+\delta)\frac{1}{2^m}\right)\tau_m \right\}, \tag{S105}$$

where conv denotes the convex hull. Since $\tau_m \in \mathcal{K}$ and $\Phi_2^{\otimes m} + (2^m - 1)\tau_m \in \mathcal{K}$, also

$$(1+\delta)\frac{1}{2^m}\,\Phi_2^{\otimes m} + \left(1 - (1+\delta)\frac{1}{2^m}\right)\tau_m + \delta\tau_m = \frac{1+\delta}{2^m}\left(\Phi_2^{\otimes m} + (2^m-1)\,\tau_m\right) \in \mathcal{K}\,. \tag{S106}$$

The convexity of $R^s_{\mathcal{K}}$ (which follows directly from the convexity of $\mathcal{K}$ itself) then implies that $R^s_{\mathcal{K}}(\Gamma_W(\sigma)) \le \delta$, and hence $\Gamma_W \in \mathrm{KP}^s_\delta$. Noting that $F\big(\Gamma_W(\rho^{\otimes n}), \Phi_2^{\otimes m}\big) = \operatorname{Tr}\rho^{\otimes n} W$, optimising over all feasible W yields

$$\begin{aligned}\sup_{\Lambda\in\mathrm{KP}^s_\delta} F\left(\Lambda(\rho^{\otimes n}), \Phi_2^{\otimes m}\right) &\ge \sup\left\{\operatorname{Tr}\rho^{\otimes n} W \,:\, 0 \le W \le \mathbb{1},\ \operatorname{Tr} W\sigma \le (1+\delta)\frac{1}{2^m}\ \ \forall \sigma \in \mathcal{K}\cap\mathcal{D}(\mathcal{H})\right\}\\ &= \varphi_{n,m}(\delta)\,,\end{aligned} \tag{S107}$$

where the function $\varphi_{n,m}(\delta)$ is defined in (S101). Using the inclusion between the sets of operations $\mathrm{KP}^s_\delta \subseteq \mathrm{KP}^g_\delta$ and $\mathrm{KP}^s_\delta \subseteq \mathrm{KP}^N_\delta$, we therefore obtain that for all n, m, δ such that $(1+\delta)\frac{1}{2^m} \le 1$

$$\begin{aligned}\sup_{\Lambda\in\mathrm{KP}^s_\delta} F\left(\Lambda(\rho^{\otimes n}), \Phi_2^{\otimes m}\right) = \sup_{\Lambda\in\mathrm{KP}^g_\delta} F\left(\Lambda(\rho^{\otimes n}), \Phi_2^{\otimes m}\right) &= \varphi_{n,m}(\delta)\,,\\ \varphi_{n,m}(\delta) \le \sup_{\Lambda\in\mathrm{KP}^N_\delta} F\left(\Lambda(\rho^{\otimes n}), \Phi_2^{\otimes m}\right) &\le \varphi_{n,m}(2\delta)\,.\end{aligned} \tag{S108}$$

We now pass from the fidelity to the trace distance. We claim that for $M \in \{R^s_{\mathcal{K}}, R^g_{\mathcal{K}}, N\}$ it holds that

$$\inf_{\Lambda\in\mathrm{KP}^M_\delta} \frac{1}{2}\left\|\Lambda(\rho^{\otimes n}) - \Phi_2^{\otimes m}\right\|_1 = 1 - \sup_{\Lambda\in\mathrm{KP}^M_\delta} F\left(\Lambda(\rho^{\otimes n}), \Phi_2^{\otimes m}\right). \tag{S109}$$

To see why this is the case, it suffices to observe that we can always twirl the output of Λ without loss of generality, i.e. we can substitute $\Lambda \mapsto \mathcal{T}_m \circ \Lambda$, where $\mathcal{T}_m$ is defined as in (S28). Doing so does not change the fact that $\Lambda \in \mathrm{KP}^M_\delta$, simply because M is convex and invariant under local unitaries; furthermore, twirling leaves $F\left(\Lambda(\rho^{\otimes n}), \Phi_2^{\otimes m}\right) = \operatorname{Tr}\Lambda(\rho^{\otimes n})\Phi_2^{\otimes m}$ invariant and never increases $\frac{1}{2}\left\|\Lambda(\rho^{\otimes n}) - \Phi_2^{\otimes m}\right\|_1$ (see (S29)). At the same time, since the output state of Λ is now twirled, and hence it is a convex combination of the orthogonal states $\Phi_2^{\otimes m}$ and τ_m, we have that

$$\frac{1}{2}\left\|\Lambda(\rho^{\otimes n}) - \Phi_2^{\otimes m}\right\|_1 = 1 - \operatorname{Tr}\Lambda(\rho^{\otimes n})\Phi_2^{\otimes m} = 1 - F\left(\Lambda(\rho^{\otimes n}), \Phi_2^{\otimes m}\right), \tag{S110}$$

completing the proof of (S109). Combining (S108) with (S109) we arrive at

$$\begin{aligned}\inf_{\Lambda\in\mathrm{KP}^s_\delta} \frac{1}{2}\left\|\Lambda(\rho^{\otimes n}) - \Phi_2^{\otimes m}\right\|_1 = \inf_{\Lambda\in\mathrm{KP}^g_\delta} \frac{1}{2}\left\|\Lambda(\rho^{\otimes n}) - \Phi_2^{\otimes m}\right\|_1 &= 1 - \varphi_{n,m}(\delta)\,,\\ 1 - \varphi_{n,m}(2\delta) \le \inf_{\Lambda\in\mathrm{KP}^N_\delta} \frac{1}{2}\left\|\Lambda(\rho^{\otimes n}) - \Phi_2^{\otimes m}\right\|_1 &\le 1 - \varphi_{n,m}(\delta)\,.\end{aligned} \tag{S111}$$

The above relations imply immediately that

$$E^\varepsilon_{d,\mathrm{KP}^s_{(\delta_n)}}(\rho) = E^\varepsilon_{d,\mathrm{KP}^g_{(\delta_n)}}(\rho) \le E^\varepsilon_{d,\mathrm{KP}^N_{(\delta_n)}}(\rho) \tag{S112}$$

for all sequences $(\delta_n)_n$. We now proceed to show that in fact also $E^\varepsilon_{d,\mathrm{KP}^N_{(\delta_n)}}(\rho) \le E^\varepsilon_{d,\mathrm{KP}^g_{(\delta_n)}}(\rho)$, which will complete the proof of the claim (S97). To this end, fix $\zeta > 0$, and let $R \ge E^\varepsilon_{d,\mathrm{KP}^N_{(\delta_n)}}(\rho) - \zeta$ be an achievable rate for entanglement distillation from ρ at error ε with operations from $\mathrm{KP}^N_{(\delta_n)}$; by definition, this implies that

$$\limsup_{n\to\infty} \inf_{\Lambda\in\mathrm{KP}^N_\delta} \frac{1}{2}\left\|\Lambda(\rho^{\otimes n}) - \Phi_2^{\otimes\lceil Rn\rceil}\right\|_1 \le \varepsilon\,, \tag{S113}$$

and thus, by (S111), that $\liminf_{n\to\infty} \varphi_{n,\lceil Rn\rceil}(2\delta_n) \geq 1-\varepsilon$. Now, since $\frac{1+\delta_n}{2^{\lceil (R-\zeta)n\rceil}} \geq \frac{1+2\delta_n}{2^{\lceil Rn\rceil}}$ for all sufficiently large n, we see by direct inspection of the definition of $\varphi_{n,m}(\delta)$ in (S101) that

$$\varphi_{n,\,\lceil (R-\zeta)n\rceil}(\delta_n) \geq \varphi_{n,\lceil Rn\rceil}(2\delta_n) \tag{S114}$$

for all sufficiently large n, thus implying that also

$$\liminf_{n\to\infty} \varphi_{n,\,\lceil (R-\zeta)n\rceil}(\delta_n) \geq 1-\varepsilon\,. \tag{S115}$$

Again by (S111), this is equivalent to stating that $R-\zeta$ is an achievable rate for $E^{\varepsilon}_{d,\,\mathrm{KP}^g_{(\delta_n)}}(\rho)$. Putting all together, we see that

$$E^{\varepsilon}_{d,\,\mathrm{KP}^g_{(\delta_n)}}(\rho) \geq R-\zeta \geq E^{\varepsilon}_{d,\,\mathrm{KP}^N_{(\delta_n)}}(\rho) - 2\zeta\,. \tag{S116}$$

Since $\zeta>0$ was arbitrary, we conclude that indeed $E^{\varepsilon}_{d,\,\mathrm{KP}^g_{(\delta_n)}}(\rho) \geq E^{\varepsilon}_{d,\,\mathrm{KP}^N_{(\delta_n)}}(\rho)$, as desired. This establishes (S97).

The reasoning for (S98) is analogous. If $\delta_n = 2^{o(n)}$ then $\frac{1+\delta_n}{2^{\lceil Rn\rceil}} \leq \frac{1}{2^{\lceil (R-\zeta)n\rceil}}$ for all $\zeta>0$ and all sufficiently large n. Therefore, if R is an achievable rate for $E^{\varepsilon}_{d,\,\mathrm{KP}^g_{(\delta_n)}}(\rho)$ then $R-\zeta$ must be achievable for $E^{\varepsilon}_{d,\,\mathrm{KP}}(\rho)$; since $\zeta>0$ is arbitrary, this ensures that $E^{\varepsilon}_{d,\,\mathrm{KP}^g_{(\delta_n)}}(\rho) \leq E^{\varepsilon}_{d,\,\mathrm{KP}}(\rho)$. The other inequality follows trivially from the inclusion $\mathrm{KP} \subseteq \mathrm{KP}^g_{\delta_n}$ for any δ_n. This proves also (S98). □

Proof of Theorem S14. For the entangement cost, it holds that

$$1 \overset{\text{(i)}}{=} E^{\tau}_N(\omega_3) \overset{\text{(ii)}}{\leq} E^{\varepsilon}_{c,\,\mathrm{KP}^N_{(\delta_n)}}(\omega_3) \overset{\text{(iii)}}{\leq} E^{\varepsilon}_{c,\,\mathrm{KP}^s_{(\delta_n)}}(\omega_3) \overset{\text{(iii)}}{\leq} E^{\varepsilon}_{c,\,\mathrm{KP}}(\omega_3) \overset{\text{(i)}}{=} 1 \tag{S117}$$

and so all inequalities must be equalities. Here, we used (i) our results in Theorem S9, (ii) Theorem S16, and (iii) the inclusion $\mathrm{KP} \subseteq \mathrm{KP}^s_{\delta_n} \subseteq \mathrm{KP}^N_{\delta_n}$ for any δ_n.

For the distillable entanglement, Lemma S17 gives

$$E_{d,\,\mathrm{KP}^s_{(\delta_n)}}(\omega_3) = E_{d,\,\mathrm{KP}^g_{(\delta_n)}}(\omega_3) = E_{d,\,\mathrm{KP}^N_{(\delta_n)}}(\omega_3) = E_{d,\,\mathrm{KP}}(\omega_3) = \log_2 \frac{3}{2}, \tag{S118}$$

where the last inequality was already shown in Theorem S9. □

V. FLUCTUATIONS IN ENTANGLEMENT MANIPULATION PROTOCOLS AND COMPARISON WITH THERMODYNAMICS

When studying the asymptotic behaviour of quantum systems, allowing for some type of microscopic fluctuation is arguably very natural from a thermodynamical perspective. Indeed, thermodynamics is a theory of macroscopic states and quantities, which should be unaffected by microscopic degrees of freedom and by the fluctuations associated with them. Although in Supplementary Note IV we justified our framework for entanglement manipulation by demonstrating the issues that come with enforcing only asymptotic (rather than exact) entanglement non-generation, it is natural to wonder: Is it not more physically meaningful to allow for fluctuations at the level of microscopic systems, rather than completely forbid entanglement generation? Let us discuss the types of fluctuations that are allowed in our entanglement manipulation framework, to illustrate why our setting is in fact very similar to others considered previously in the literature, and to pinpoint the key difference with Brandão and Plenio's approach [10, 47].

Although fluctuations are a natural component of our theory, not all fluctuations are equal: in the context of state-to-state transformations in entanglement theory (cf. the discussion in Section I B), and more generally in that of quantum resource theories [48], we find it useful to introduce the following classification.

(I) *Fluctuations at the level of microscopic transformation error.* Specifically, one typically requires that transformations of the form $\rho \to \omega$ are only realised approximately as $\Lambda(\rho^{\otimes n}) \approx \omega^{\otimes m}$ with some non-zero error (usually measured by the trace norm), and only in the limit $n \to \infty$ does the error vanish. Such fluctuations are common to most approaches to quantum resource theories, including quantum thermodynamics as well as entanglement theory [48]. Crucially, without such fluctuations, even quantum thermodynamics is no longer a reversible theory, and many state transformations become impossible [49–51].

We do note, however, that in some contexts one may in fact be interested in exact, i.e. zero-error, entanglement distillation [52, 53] or dilution [32, 34, 54]. In the language we are using here, such settings would explicitly rule out microscopic transformation errors (as in (S22)–(S23)), or would require them to vanish exceedingly — i.e. super-exponentially — fast [55]. For these reasons, these approaches are generally regarded as less physically motivated than the one we take here, which explicitly allows for vanishingly small transformation errors.

(II) *Fluctuations in the resources consumed by the process.* For example, in quantum thermodynamics, one often includes a small ancillary system to activate a desired transformation [56, 57]. This is arguably necessary to circumvent the energy super-selection rule that prohibits the creation of coherence between different energy levels under an energy-preserving unitary: without the ancilla, an energy-incoherent state would remain energy-incoherent under thermal operations, thus preventing dilution of resources altogether [56]. Including a small ancillary system that carries some coherence in the energy eigenbasis solves the problem, but its small size means that its contributions to the rate will asymptotically vanish, leaving the underlying physics unaffected.

More detailed insights on sensible physical requirements to impose on the ancillary system can be deduced from [57]. There the authors look at transformations of the form

$$\rho^{\otimes n} \longrightarrow \rho^{\otimes n} \otimes \eta_n \longrightarrow \mathrm{Tr}_2 \left[U \left(\rho^{\otimes n} \otimes \eta_n \right) U^\dagger \right], \tag{S119}$$

where U is required to be an energy-preserving unitary, η_n is a 'small' and 'not too energetic' ancilla, and Tr_2 denotes partial trace over the ancillary modes. Although the setting of that work is slightly different than the one considered here or in [56], the conclusions are nevertheless insightful: in [57] it is found that, although fluctuations (II) are indeed necessary to enable the reversibility of the theory, ancillae with subexponential dimension $\log \dim \eta_n = O\left(\sqrt{n \log n}\right)$ and sublinear Hamiltonian operator norm $\|H\|_\infty = O\left(n^{2/3}\right)$ suffice to established the desired result.

Since they play such a key role in thermodynamics, it is certainly meaningful to try to take into account fluctuations of this type in entanglement theory as well. However, although it may not be apparent at first sight, these fluctuations are already implicitly included in our framework — and in fact, in most of the commonly encountered formulations of entanglement theory. They take the form of sublinear fluctuations in the number of entanglement bits consumed by either the distillation or the dilution process. But in the definitions of E_d (S19) and E_c (S20) we only care about asymptotic rates, so sublinear fluctuations are suppressed in the limit.

In other words: suppose that in our original definitions in Section I B we chose to consider a larger class of transformations than $\mathcal{K}$-preserving ones; a general transformation of this new class would be obtained by: (a) attaching an ancilla of $o(n)$ many qubits per party initialised in any (possibly entangled) state, and (b) performing a $\mathcal{K}$-preserving operation on the joint system. In this way, the allowed transformations are of the form

$$\rho^{\otimes n} \longrightarrow \rho^{\otimes n} \otimes \tau_n \longrightarrow \Lambda_n \left(\rho^{\otimes n} \otimes \tau_n \right), \tag{S120}$$

where $\Lambda_n \in \mathrm{KP}$ is an arbitrary $\mathcal{K}$-preserving operation, and $\tau_n = \tau_n^{A'_n B'_n}$ is an ancillary state over a system $A'_n B'_n$. Our assumptions on this ancilla are as follows:

(i) $A'_nB'_n$ can be an arbitrary bipartite system, possibly dependent on n, but it has to have subexponential dimension, i.e. $\dim(A'_nB'_n) = \dim\tau_n = 2^{o(n)}$; that is, $\log\dim(A'_nB'_n) = o(n)$ is required to be sublinear, or in other words $A'_nB'_n$ should be made of sublinearly many qubits;

(ii) Apart from this, τ_n is completely arbitrary. It could be for instance a maximally entangled state composed of sublinearly many ebits.

Formally, the new distillable entanglement and entanglement cost now look like this:

$$\widetilde{E}_{d,\mathrm{KP}}(\rho_{AB}) := \sup\left\{R>0:\ \lim_{n\to\infty}\inf_{\Lambda_n\in\mathrm{KP}}\frac{1}{2}\left\|\Lambda_n\left(\rho_{AB}^{\otimes n}\otimes\tau_n\right)-\Phi_2^{\otimes\lceil Rn\rceil}\right\|_1=0,\ \lim_{n\to\infty}\frac{\log\dim\tau_n}{n}=0\right\}, \tag{S121}$$

$$\widetilde{E}_{c,\mathrm{KP}}(\rho_{AB}) := \inf\left\{R>0:\ \lim_{n\to\infty}\inf_{\Lambda_n\in\mathrm{KP}}\frac{1}{2}\left\|\Lambda_n\left(\Phi_2^{\otimes\lfloor Rn\rfloor}\otimes\tau_n\right)-\rho_{AB}^{\otimes n}\right\|_1=0,\ \lim_{n\to\infty}\frac{\log\dim\tau_n}{n}=0\right\}. \tag{S122}$$

Here $\dim\tau_n$ denotes the dimension of the $A'_nB'_n$ system of the ancilla.

We can now ask ourselves: can it be that $\widetilde{E}_{d,\mathrm{KP}}(\rho_{AB}) > E_{d,\mathrm{KP}}(\rho_{AB})$ or $\widetilde{E}_{c,\mathrm{KP}}(\rho_{AB}) < E_{c,\mathrm{KP}}(\rho_{AB})$? In other words, can it happen that granting a sublinear number of ancillary qubits allows for better rates in either entanglement distillation or entanglement dilution? The answer turns out to be negative, and the fundamental reason is that ancillae made of a sublinear number of qubits cannot change the rate. In full detail, a proof can be constructed as follows.

Lemma S18. *For any state ρ_{AB}, it holds that*

$$\widetilde{E}_{d,\mathrm{KP}}(\rho_{AB}) = E_{d,\mathrm{KP}}(\rho_{AB}) \qquad \textit{and} \qquad \widetilde{E}_{c,\mathrm{KP}}(\rho_{AB}) = E_{c,\mathrm{KP}}(\rho_{AB})\,. \tag{S123}$$

Proof. It is clear that $\widetilde{E}_{d,\mathrm{KP}}(\rho_{AB}) \geq E_{d,\mathrm{KP}}(\rho_{AB})$ and $\widetilde{E}_{c,\mathrm{KP}}(\rho_{AB}) \leq E_{c,\mathrm{KP}}(\rho_{AB})$, so we only need to prove the converse inequalities. To do so, assume that R is an achievable rate for $\widetilde{E}_{c,\mathrm{KP}}(\rho_{AB})$, so that we can find a sequence of ancillae τ_n and $\mathcal{K}$-preserving operations $\Lambda_n \in \mathrm{KP}$ with the property that

$$\frac{1}{2}\left\|\Lambda_n\left(\Phi_2^{\otimes\lfloor Rn\rfloor}\otimes\tau_n\right)-\rho_{AB}^{\otimes n}\right\|_1 \xrightarrow[n\to\infty]{} 0\,. \tag{S124}$$

Fix $\delta>0$, and take n large enough so that $2^{n\delta}\geq\dim\tau_n$. Since any state can be prepared from the maximally entangled state of the same dimension via LOCC, we can prepare τ_n starting from $\Phi_2^{\otimes\lceil n\delta\rceil}$ and using an LOCC Ξ_n, i.e. consuming $\lceil n\delta\rceil$ many ebits. In formula,

$$\Xi_n\left(\Phi_2^{\otimes\lceil\delta n\rceil}\right)=\tau_n\,. \tag{S125}$$

Now, consider the operation $\Lambda'_n := \Lambda_n\circ\left(I^{\otimes\lfloor Rn\rfloor}\otimes\Xi_n\right)$, where $I^{\otimes\lfloor Rn\rfloor}$ denotes the identity channel on the first $\lfloor Rn\rfloor$ qubits. Note that Λ'_n is still $\mathcal{K}$-preserving, because the set of $\mathcal{K}$-preserving operations is closed under composition, and LOCCs are $\mathcal{K}$-preserving. Moreover, it satisfies that

$$\Lambda'_n\left(\Phi_2^{\otimes(\lfloor Rn\rfloor+\lceil\delta n\rceil)}\right)=\Lambda_n\left(\Phi_2^{\otimes\lfloor Rn\rfloor}\otimes\Xi_n\left(\Phi_2^{\otimes\lceil\delta n\rceil}\right)\right)=\Lambda_n\left(\Phi_2^{\otimes\lfloor Rn\rfloor}\otimes\tau_n\right). \tag{S126}$$

Thus, thanks to (S124)

$$\frac{1}{2}\left\|\Lambda'_n\left(\Phi_2^{\otimes(\lfloor Rn\rfloor+\lceil\delta n\rceil)}\right)-\rho_{AB}^{\otimes n}\right\|_1 \xrightarrow[n\to\infty]{} 0\,. \tag{S127}$$

Since now there is no ancilla, we obtain that the rate

$$\lim_{n\to\infty} \frac{\lfloor Rn \rfloor + \lceil \delta n \rceil}{n} = R + \delta \tag{S128}$$

is actually achievable for $E_{c,\mathrm{KP}}(\rho_{AB})$. Therefore,

$$E_{c,\mathrm{KP}}(\rho_{AB}) \leq R + \delta \,. \tag{S129}$$

Taking the infimum over R yields that $E_{c,\mathrm{KP}}(\rho_{AB}) \leq \widetilde{E}_{c,\mathrm{KP}}(\rho_{AB}) + \delta$. Since $\delta > 0$ was arbitrary, we obtain that in fact

$$E_{c,\mathrm{KP}}(\rho_{AB}) \leq \widetilde{E}_{c,\mathrm{KP}}(\rho_{AB}) \,, \tag{S130}$$

which is what we wanted to show.

To show that $\widetilde{E}_{d,\mathrm{KP}}(\rho_{AB}) \leq E_{d,\mathrm{KP}}(\rho_{AB})$, assume that R is an achievable rate for $\widetilde{E}_{d,\mathrm{KP}}(\rho_{AB})$, i.e. that there exists a valid choice of $(\tau_n)_n$ and $\mathcal{K}$-preserving operations $\Lambda_n \in \mathrm{KP}$ such that

$$\frac{1}{2} \left\| \Lambda_n \left(\rho^{\otimes n} \otimes \tau_n \right) - \Phi_2^{\otimes \lceil Rn \rceil} \right\|_1 \xrightarrow[n\to\infty]{} 0 \,. \tag{S131}$$

Noting that τ_n is a finite-dimensional system, the optimal decomposition for the standard robustness in (S11) exists, and we can write

$$\tau_n = \left(1 + R^s_{\mathcal{K}}(\tau_n)\right) \sigma_n^+ - R^s_{\mathcal{K}}(\tau_n)\, \sigma_n^- \tag{S132}$$

for some normalised states $\sigma_n^\pm \in \mathcal{K}$. Observe then that for any state $\sigma \in \mathcal{K}_{A^n B^n}$, it holds that

$$\begin{aligned} \Lambda_n(\sigma \otimes \tau_n) &= \left(1 + R^s_{\mathcal{K}}(\tau_n)\right) \Lambda_n\left(\sigma \otimes \sigma_n^+\right) - R^s_{\mathcal{K}}(\tau_n)\, \Lambda_n\left(\sigma \otimes \sigma_n^-\right) \\ &= \left(1 + R^s_{\mathcal{K}}(\tau_n)\right) \sigma' - R^s_{\mathcal{K}}(\tau_n)\, \sigma'' \end{aligned} \tag{S133}$$

for some states $\sigma', \sigma'' \in \mathcal{K}$ due to the fact that $\mathcal{K}$ is closed under tensor product. This implies that

$$R^s_{\mathcal{K}}\left(\Lambda_n(\sigma \otimes \tau_n)\right) \leq R^s_{\mathcal{K}}(\tau_n) \leq \dim \tau_n - 1, \tag{S134}$$

where the last inequality is due to the fact that the value of $R^s_{\mathcal{K}}(\Phi_{\dim \tau_n}) = \dim \tau_n - 1$ is maximal among states of the same dimension. As $\dim \tau_n = 2^{o(n)}$ by assumption, this entails that each $\Lambda_n(\cdot \otimes \tau_n)$ is an $(R^s_{\mathcal{K}}, 2^{o(n)})$-approximately $\mathcal{K}$-preserving operation (cf. Supplementary Note IV). The proof is thus concluded by recalling from Lemma S17 that such operations cannot increase the rate of distillation compared to exactly $\mathcal{K}$-preserving operations, that is, $R \leq E_{d,\mathrm{KP}}(\rho)$. □

In the context of quantum thermodynamics, we can compare this to works which studied the manipulation of quantum systems under so-called Gibbs-preserving operations (e.g. [58]), defined to be all channels Λ such that $\Lambda(\gamma) = \gamma'$ where γ, γ' are the equilibrium states of the input and output system, respectively. Although no explicit resource fluctuations are allowed in such a definition, the Gibbs-preserving framework recovers the exact same asymptotic rates as frameworks that do consider fluctuations in the consumed resources [56, 57] — this is precisely because the type-II fluctuations are implicit there, as one only looks at the rates.

(III) *Genuine fluctuations in the fundamental physical laws governing the process.* These fluctuations are excluded in most of the literature on quantum thermodynamics: for example, in Refs. [56, 57] the unitary operators acting on the joint system (including the ancilla) are required to be *exactly* and not only approximately energy-preserving. This is well justified from a physical perspective: analogously, we would not claim that energy is only approximately preserved in a piece of uranium because we lost track of some neutrinos; include those back into the picture, and you will restore exact energy preservation. It should be noted at this point that although the law of energy conservation is, to the best of our knowledge, exactly obeyed, this fact alone does not play a decisive role for the validity of thermodynamics, which is a macroscopic rather than microscopic theory. Still, the fact that it is conceivable that Nature preserves energy exactly shows that a setting excluding genuine type-III fluctuations — yet encompassing type-I and type-II fluctuations — is somewhat reasonable, if not completely satisfactory.

Importantly, in most of the known physical theories including thermodynamics, disallowing type-III fluctuations does not change the underlying physics whatsoever. Although they *can* in principle be considered, as we elaborate on below, genuine fluctuations of the physical laws governing thermodynamical processes — such as global unitarity and energy conservation [56] — are not necessary to construct a reversible theory of quantum thermodynamics [56, 58–60].

A possible objection to shis claim could be that the maps considered in [56, 57] are in fact not exactly but only approximately Gibbs-preserving. While this could seem *prima facie* an example of a type-III fluctuation, it is only a spurious one, essentially because of the aforementioned fact that these maps can be thought of as Gibbs-preserving operations acting on a larger quantum system with an ancilla of sublinear size. These apparent type-III fluctuations are thus in fact type-II fluctuations in disguise, and as such they are included also in our original framework.

What our results show in this context is that Brandão and Plenio's asymptotically non-entangling operations [10, 47] admit fluctuations that are *genuinely* of type III (see Section IV). Specifically, they cannot be re-absorbed into the type-II category — if this were the case, then the entanglement cost $E_{c,\,\mathrm{ANE}}$ under asymptotically non-entangling operations [10, Definition III.2] would necessarily be equal to that under non-entangling operations, which we denoted by $E_{c,\,\mathrm{NE}}$. Since the former is given by the regularised relative entropy of entanglement[5] [10, 61], in formula $E_{c,\,\mathrm{ANE}} = E_{r,\mathcal{S}}^{\infty}$, we would deduce that the entanglement cost under non-entangling operations must also equal $E_{c,\,\mathrm{NE}} = E_{r,\mathcal{S}}^{\infty}$. But that this is not the case in general is precisely the content of our main result (Theorem S9), whose proof reveals that $E_{c,\,\mathrm{NE}}(\omega_3) = 1 > \log_2 \frac{3}{2} = E_{r,\mathcal{S}}^{\infty}(\omega_3)$.

Nevertheless, one can argue that allowing type-III fluctuations is actually the sensible thing to do in entanglement manipulation, as it allows one to achieve an even greater level of generality and a stricter adherence to the spirit of thermodynamics. This is especially important when discussing no-go results such as asymptotic irreversibility: if reversibility could be restored with just an unequivocally vanishing amount of fluctuations — even ones of type III — then, arguably, such irreversibility would not be robust. This is precisely the motivation for our Theorem S14, where we clarify that it is impossible to have any reversible framework of entanglement that generates only small amounts of it: using as an entanglement quantifier either the standard robustness of entanglement or the negativity, we show that these 'fluctuations' must in fact be macroscopically large, putting into question the physicality of any conceivable reversible theory of entanglement.

What is pivotal to understanding the consequences of our results is that thermodynamics is a reversible theory even when fluctuations of type III are not allowed. This is addressed in works such as Ref. [56, 59, 60], where the manipulation of quantum states under thermal operations was considered, and extended also to quantum channels in [58] under the Gibbs-preserving

[5] Here we note that, despite the issue with the proof of [10] uncovered in [38], this part of the argument by Brandão and Plenio is not affected. The proof can also be obtained by combining the framework of [10] with the asymptotic equipartition property of $R_{\mathcal{S}}^{g}$, for which an alternative proof is given in [61].

framework. That is, fluctuations of types I and II fully suffice to recover the entropy as the unique quantity governing the thermodynamical transformations of macroscopic systems, and the physical constraints of energy conservation can be enforced at all scales, including microscopic ones. In fact, we stress that in the Gibbs-preserving framework — which is completely analogous to the non-entangling operations used in our work — even fluctuations of type I are sufficient to establish the reversibility of thermodynamics [58], but one can equivalently consider type-I and type-II fluctuations.

Therefore, our entanglement manipulation framework under non-entangling operations — which does allow type-I and type-II but forbids type-III fluctuations — follows exactly the same reasoning as the quantum thermodynamics frameworks of Refs. [56–60, 62–65], and yet it leads to a completely opposite conclusion, as per Theorem S9. It strikes us as surprising the stark contrast between thermodynamics, which is asymptotically reversible, and entanglement theory, which turns out to be fundamentally irreversible under the same assumptions, no matter how much one strives to avoid it.

That is not to say that there is something inherently wrong with the framework proposed by Brandão and Plenio [10] — quite the contrary, there is no intrinsic reason why fluctuations of type III *cannot* be allowed, even if they are not needed in thermodynamics. However, due to the ambiguity in defining 'small' amounts of entanglement in the asymptotic limit, even such permissive fluctuations are not enough in light of our Theorem S14, unless they are made non-vanishingly large. Our work therefore provides insight into how any potential reversible entanglement framework would function and the size of 'fluctuations' it would need to allow.

In conclusion, although connections between entanglement theory and thermodynamics such as the one conjectured in [10] can possibly be established, our work conclusively shows that reversibility of entanglement, if at all possible, cannot play out in the same way as it does in the theory of thermodynamics (with type-I and type-II fluctuations, but without type-III fluctuations), or indeed not even in a similar way (with universally small type-III fluctuations). A fundamental and inexorable difference thus exists between the two theories.

VI. FURTHER CONSIDERATIONS

A. Confessions of ω_3

Here we explain the intuition behind our choice of the state ω_3 as the counterexample to the reversibility of entanglement manipulation (Theorems S9 and S14). It will rely on the relation between two entanglement monotones that we have encountered in the course of this work — the standard robustness $R^s_{\mathcal{K}}$ and the generalised robustness $R^g_{\mathcal{K}}$. Crucially, the work of Brandão and Plenio [10] connected each of these quantities with the operational tasks of entanglement distillation and dilution. In particular, Ref. [10] (cf. [43]) showed that, when considering only a single copy of a state rather than an asymptotic rate, the entanglement cost under $\mathcal{K}$-preserving operations is given exactly by the logarithm of $1 + R^s_{\mathcal{K}}(\rho)$. This then implies that the asymptotic exact cost of entanglement (Eq. (S23)) is given by

$$E^{\text{exact}}_{c,\,\text{KP}}(\rho) = \lim_{n\to\infty} \frac{1}{n} \log_2 \left(1 + R^s_{\mathcal{K}}\left(\rho^{\otimes n}\right)\right). \tag{S135}$$

The entanglement cost $E_{c,\,\text{KP}}$ also needs to account for an asymptotically vanishing transformation error, and it can thus be expressed by suitably 'smoothing' the robustness over the ε-ball around a given state, i.e.

$$E_{c,\,\text{KP}}(\rho) = \lim_{\varepsilon\to 0^+} \lim_{n\to\infty} \frac{1}{n} \inf_{\substack{\rho'\in\mathcal{D}(\mathcal{H}_{AB}^{\otimes n}),\\ \frac12\left\|\rho'-\rho^{\otimes n}\right\|_1\le\varepsilon}} \log_2\left(1 + R^s_{\mathcal{K}}(\rho')\right). \tag{S136}$$

On the other hand, employing a connection established between the generalised robustness $R^g_{\mathcal{K}}$ and the regularised relative entropy $E^\infty_{r,\mathcal{K}}$ [10, 61], the distillable entanglement can be tightly upper bounded as [10, 28]

$$E_{d,\mathrm{KP}}(\rho) \leq E^\infty_{r,\mathcal{K}}(\rho_{AB}) = \lim_{\varepsilon\to 0^+} \lim_{n\to\infty} \frac{1}{n} \inf_{\substack{\rho'\in\mathcal{D}(\mathcal{H}_{AB}^{\otimes n}),\\ \frac12\left\|\rho'-\rho^{\otimes n}\right\|_1\leq\varepsilon}} \log_2\left(1+R^g_{\mathcal{K}}(\rho')\right). \tag{S137}$$

These expressions lead to a very curious fact: showing the irreversibility of entanglement manipulation under the class of $\mathcal{K}$-preserving maps can be done by exhibiting a gap between the smoothed regularisations of $R^s_{\mathcal{K}}$ and $R^g_{\mathcal{K}}$ in (S136) and (S137).

The daunting expressions for the regularised robustness measures do not immediately make studying reversibility any easier. Let us then start by asking a more basic question: is there a gap between the standard and the generalised entanglement robustness, $R^s_{\mathcal{S}}$ and $R^g_{\mathcal{S}}$? We stumbled upon the state ω_3 precisely when looking for a way to demonstrate such a gap. Indeed, we have already seen in the course of proving Theorem S9 that

$$R^s_{\mathcal{S}}(\omega_3) = \frac{3}{4}. \tag{S138}$$

It is also not difficult to show that the decomposition $\omega_3 + \frac12\Phi_3 = \frac12 P_3$ is optimal for the generalised robustness of this state, giving

$$R^g_{\mathcal{S}}(\omega_3) = \frac{1}{2}. \tag{S139}$$

We thus see an explicit gap between the two robustness measures. The direct connections between $R^s_{\mathcal{S}}$ and $R^g_{\mathcal{S}}$ on one side and, respectively, the entanglement cost and distillable entanglement on the other then motivated us to look into ω_3 as a possible candidate for a state whose cost could be strictly larger than the entanglement that can be distilled from it. This leads directly to the considerations expounded in the main text of the paper and to the main results of this work.

B. Strong converses and error-rate trade-offs

According to Definition S3, the theory of entanglement manipulation is considered reversible under some set of operations if the distillable entanglement and the entanglement cost coincide for all states. As we have seen, these latter two quantities are defined in terms of asymptotic transformations in which the allowed error is required to vanish as the number of copies of the state grows. In information theory, classical as well as quantum, one can study also weaker notions of transformation rates, dubbed *strong converse rates*. In this setting, one requires instead that the error, as measured by half of the trace norm distance between the output and the target state, is bounded away from its maximum value of 1. Intuitively, what this means is that attempting to distill or dilute entanglement at a rate larger than the strong converse one necessarily incurs an error that grows to 1, making the protocol useless. Formally, we can define the strong converse rates of distillation and dilution, respectively, by

$$E^\dagger_{d,\mathrm{KP}}(\rho_{AB}) := \sup_{\varepsilon\in[0,1)} E^\varepsilon_{d,\mathrm{KP}}(\rho_{AB}), \qquad E^\dagger_{c,\mathrm{KP}}(\rho_{AB}) := \inf_{\varepsilon\in[0,1)} E^\varepsilon_{c,\mathrm{KP}}(\rho_{AB}). \tag{S140}$$

From these definitions and from the discussion that inspired them it is clear that $E^\dagger_{d,\mathrm{KP}}(\rho) \geq E_{d,\mathrm{KP}}(\rho)$ and $E^\dagger_{c,\mathrm{KP}}(\rho) \leq E_{c,\mathrm{KP}}(\rho)$ for all states ρ.

We could thus wonder whether there is a possibility of achieving a larger distillable entanglement or a lower entanglement cost, perhaps even restoring a (substantially weaker) form of reversibility, by passing to the corresponding strong converse rates. In the case of entanglement distillation from the state ω_3 defined in (S60), such possibility can be ruled out by exploiting a result of Hayashi [28, Theorem 8.7] (see also [10]), who showed that the regularised relative entropy distance from the set of states in $\mathcal{K}$ defined in (S57) is also an upper bound on the strong converse distillable entanglement, i.e. $E_{d,\mathrm{KP}}(\rho_{AB}) \leq E^{\dagger}_{d,\mathrm{KP}}(\rho_{AB}) \leq E^{\infty}_{r,\mathcal{K}}(\rho_{AB})$ (cf. (S58)). Our result in Theorem S9 then implies that $E_{d,\mathrm{KP}}(\omega_3) = E^{\dagger}_{d,\mathrm{KP}}(\omega_3) = \frac{3}{2}$. Indeed, coupled with Lemma S17, the same strong converse bound holds for distillation with operations that generate sub-exponential amounts of entanglement.

As for the entanglement cost, we have not yet been able to prove that no improvement can be obtained by considering the corresponding strong converse rate. However, we deem such a possibility quite unlikely, and we present strong evidence against it by showing that any such error would have to be very large, which effectively rules out this approach as a practically viable way of improving entanglement dilution. We leave the complete solution of this problem, which we formalise below, as an open question left for future work.

Conjecture S19. *For $\mathcal{K} = \mathcal{S}, \mathcal{PPT}$, the two-qutrit state ω_3 defined by (S60) satisfies that*

$$\inf_{\varepsilon\in[0,1)} E^{\varepsilon}_{c,\mathrm{KP}}(\omega_3) = E^{\dagger}_{c,\mathrm{KP}}(\omega_3) = 1\,. \tag{S141}$$

If the above conjecture is true, then we see that

$$E_{d,\mathrm{KP}}(\omega_3) = E^{\dagger}_{d,\mathrm{KP}}(\omega_3) = \log_2 \frac{3}{2} < 1 = E^{\dagger}_{c,\mathrm{KP}}(\omega_3) = E_{c,\mathrm{KP}}(\omega_3)\,, \tag{S142}$$

i.e. the gap between distillable entanglement and entanglement cost remains even upon passing to the strong converse exponents.

Theorem S9 already guarantees that

$$\inf_{\varepsilon\in[0,\,\varepsilon_{\max})} E^{\varepsilon}_{c,\mathrm{KP}}(\omega_3) = 1 \tag{S143}$$

holds for $\varepsilon_{\max} = 1/2$. The problem we are faced with now is how to increase the value of $\varepsilon_{\max}$ in (S143). In general, we would like to study more in depth the behaviour of the function $E^{\varepsilon}_{c,\mathrm{KP}}(\omega_3)$ for values of ε close to 1. A partial result is as follows:

Lemma S20. *For all $\delta \in (0, 1]$, it holds that*

$$\inf_{\varepsilon\in[0,\,1-\delta)} E^{\varepsilon}_{c,\mathrm{NE}}(\omega_3) \geq \inf_{\varepsilon\in[0,\,1-\delta)} E^{\varepsilon}_{c,\mathrm{PPTP}}(\omega_3) \geq \begin{cases} 1 & \textit{if } 1/3 \leq \delta \leq 1, \\ \log_2 \frac{3(1-\delta)}{2-3\delta} & \textit{if } 0 < \delta < 1/3. \end{cases} \tag{S144}$$

Let us discuss the consequences of this result. First, we see that performing entanglement dilution at the rate 1 — which we know is optimal for $E_{c,\mathrm{NE}}$ — must incur an error of at least $\frac{2}{3}$ asymptotically. This means that we can in fact take $\varepsilon_{\max} = 2/3$ in (S143), which already improves upon the error $\varepsilon_{\max} = 1/2$ that we obtained in Theorem S9. Furthermore, attempting to perform dilution of ω_3 at a rate below 1 must yield an even larger error; the smaller the rate, the greater the error. One commonly refers to such behaviour as an *error-rate trade-off*.

Proof of Lemma S20. Let R be an achievable rate for the entanglement cost of ω_3 at error threshold $\varepsilon < 1-\delta$. That is, let there exist a sequence of non-entangling operations $\Lambda_n \in \mathrm{NE}_{A_0^{\lfloor Rn\rfloor} B_0^{\lfloor Rn\rfloor} \to A^n B^n}$, with A_0, B_0 being single-qubit systems, such that

$$\Omega_n := \Lambda_n\left(\Phi_2^{\otimes \lfloor Rn\rfloor}\right), \qquad \varepsilon_n := \frac{1}{2}\left\|\Omega_n - \omega_3^{\otimes n}\right\|_1, \qquad \limsup_{n\to\infty} \varepsilon_n < 1-\delta\,. \tag{S145}$$

For $\mathcal{K} = \mathcal{S}, \mathcal{PPT}$, we have that

$$
\begin{aligned}
2^{\lfloor Rn \rfloor} &\overset{\text{(i)}}{\geq} 1 + R_{\mathcal{K}}^{s}(\Omega_n) \\
&\overset{\text{(ii)}}{=} 1 + \sup_{\Omega'} R_{\mathcal{K}}^{\tau}(\Omega_n \,|\, \Omega') \\
&\overset{\text{(iii)}}{\geq} \frac{1}{2} \sup_{\Omega'} \left\{ N_\tau(\Omega_n \,|\, \Omega') + 1 \right\} \\
&\geq \frac{1}{2} N_\tau\left(\Omega_n \,\middle|\, \omega_3^{\otimes n}\right).
\end{aligned}
\tag{S146}
$$

Here, the inequality in (i) is just a rephrasing of that in step (ii) of (S51). In the subsequent lines of (S146), the identity in (ii) and the inequality in (iii) follow from Proposition S5, items (a) and (d), respectively.

To continue our argument, we need to lower bound $N_\tau\left(\Omega_n \,\middle|\, \omega_3^{\otimes n}\right)$, as usual. While in the case of vanishing error the operator X_3 defined by (S64) was the right ansatz for the optimisation (S32) defining the tempered negativity, this time we will make a more sophisticated choice. Using the notation defined in (S59), set

$$
X_3(\delta) := \begin{cases} 2P_3 - 3\Phi_3 & \text{if } 1/3 \leq \delta \leq 1, \\ \frac{3}{2-3\delta}\left((1-\delta)P_3 - \Phi_3\right) & \text{if } 0 < \delta < 1/3. \end{cases}
\tag{S147}
$$

We now verify that (a) $\left\|X_3(\delta)^\Gamma\right\|_\infty = 1$ and (b) $\|X_3(\delta)\|_\infty = \operatorname{Tr} X_3(\delta)\omega_3$. Thanks to (S67)–(S68), we can limit ourselves to the case where $0 < \delta < 1/3$. As for (a), we have that

$$
\left\|X_3(\delta)^\Gamma\right\|_\infty = \frac{3}{2-3\delta}\left\|(1-\delta)P_3 - \frac{1}{3}F_3\right\|_\infty = \frac{3}{2-3\delta}\max\left\{\frac{2}{3} - \delta,\, \frac{1}{3}\right\} = 1\,,
\tag{S148}
$$

where $F_3 = \sum_{j,k=1}^{3} |jk\rangle\!\langle kj|$ is as usual the flip operator, and the second equality is elementary because P_3 and F_3 commute. Concerning (b), it suffices to observe that

$$
\|X_3(\delta)\|_\infty = \frac{3}{2-3\delta}\max\{\delta,\, 1-\delta\} = \frac{3(1-\delta)}{2-3\delta} = \operatorname{Tr} X_3(\delta)\omega_3\,,
\tag{S149}
$$

where again the computation of the operator norm is elementary because P_3 and Φ_3 commute. From the above claims (a) and (b), we deduce immediately that also (a') $\left\|\left(X_3(\delta)^{\otimes n}\right)^\Gamma\right\|_\infty = 1$ and (b') $\|X_3(\delta)^{\otimes n}\|_\infty = \operatorname{Tr}\left[X_3(\delta)^{\otimes n}\omega_3^{\otimes n}\right]$. This in turn ensures that $X_3(\delta)^{\otimes n}$ is a feasible ansatz for the optimisation that defines $N_\tau\left(\Omega_n \,\middle|\, \omega_3^{\otimes n}\right)$ (cf. (S32)). Hence,

$$
N_\tau\left(\Omega_n \,\middle|\, \omega_3^{\otimes n}\right) \geq \operatorname{Tr}\left[X_3(\delta)^{\otimes n}\Omega_n\right].
\tag{S150}
$$

To further lower bound the right-hand side, we need to have a closer look at the spectral structure of $X_3(\delta)^{\otimes n}$. Let us distinguish the two cases $0 < \delta < 1/3$ and $1/3 \leq \delta \leq 1$. In the former case, according to (S149) the largest eigenvalue of this operator, $\frac{3^n(1-\delta)^n}{(2-3\delta)^n}$, corresponds to the eigenspace with projector $(P_3 - \Phi_3)^{\otimes n}$. The smallest eigenvalue, instead, is easily seen to be $-\frac{3^n(1-\delta)^{n-1}\delta}{(2-3\delta)^n}$. We deduce that

$$
\begin{aligned}
X_3(\delta)^{\otimes n} &\geq \frac{3^n(1-\delta)^n}{(2-3\delta)^n}(P_3 - \Phi_3)^{\otimes n} - \frac{3^n(1-\delta)^{n-1}\delta}{(2-3\delta)^n}\left(\mathbb{1} - (P_3 - \Phi_3)^{\otimes n}\right) \\
&= \frac{3^n(1-\delta)^{n-1}}{(2-3\delta)^n}\left((P_3 - \Phi_3)^{\otimes n} - \delta\mathbb{1}\right).
\end{aligned}
\tag{S151}
$$

The above operator inequality is valid for $0 < \delta < 1/3$. When $1/3 \leq \delta \leq 1$, instead, an analogous reasoning yields more simply

$$
X_3(\delta)^{\otimes n} \geq 2^n(P_3 - \Phi_3)^{\otimes n} - 2^{n-1}\left(\mathbb{1} - (P_3 - \Phi_3)^{\otimes n}\right) = 2^{n-1}\left(3(P_3 - \Phi_3)^{\otimes n} - \mathbb{1}\right).
\tag{S152}
$$

Before completing the proof, let us observe that by virtue of (S145) we have

$$\operatorname{Tr}\left[(P_3-\Phi_3)^{\otimes n}\Omega_n\right] = 1 + \operatorname{Tr}\left[(P_3-\Phi_3)^{\otimes n}\left(\Omega_n - \omega_3^{\otimes n}\right)\right] \geq 1-\varepsilon_n\,. \tag{S153}$$

Continuing from (S150), when $0<\delta<1/3$ we infer

$$\begin{aligned} N_\tau\left(\Omega_n\,\middle|\,\omega_3^{\otimes n}\right) &\overset{\text{(iv)}}{\geq} \frac{3^n(1-\delta)^{n-1}}{(2-3\delta)^n}\operatorname{Tr}\left[\left((P_3-\Phi_3)^{\otimes n}-\delta\mathbb{1}\right)\Omega_n\right] \\ &\overset{\text{(v)}}{\geq} \frac{3^n(1-\delta)^{n-1}}{(2-3\delta)^n}\left(1-\delta-\varepsilon_n\right) \\ &= \left(\frac{3(1-\delta)}{2-3\delta}\right)^n\left(1-\frac{\varepsilon_n}{1-\delta}\right). \end{aligned} \tag{S154}$$

Here, the inequality in (iv) is obtained by plugging (S151) into (S150), while that in (v) comes from (S153). Remembering (S146), we obtain that

$$2^{\lfloor Rn\rfloor} \geq \frac12\left(\frac{3(1-\delta)}{2-3\delta}\right)^n\left(1-\frac{\varepsilon_n}{1-\delta}\right). \tag{S155}$$

Computing the logarithm of both sides, dividing by n, and using (S145) to take the limit for $n\to\infty$ yields $R\geq\log_2\frac{3(1-\delta)}{2-3\delta}$. The reasoning for the case where $1/3\leq\delta\leq1$ is entirely analogous, leading to $R\geq1$. Since the rate R was completely arbitrary, the proof is complete. □

C. Implications for coherence theory

The resource theory of quantum coherence [66, 67] is concerned with the study of the operational manipulation of the resource represented by superposition in a fixed basis $\{|i\rangle\}_i$ of a Hilbert space — very much analogously to the way that entanglement has been studied in this work. Indeed, one can define the asymptotic quantities such as coherence cost C_c and distillable coherence C_d in a manner completely equivalent to the entanglement-based definitions of our work [68]. It is, in particular, known that the manipulation of coherence is asymptotically reversible under the class of maximally incoherent operations (MIO) [68, 69], which are the coherence theory equivalent of non-entangling transformations: they map any incoherent (diagonal) state into another incoherent state. For such maps, it then holds that $C_{d,\mathrm{MIO}}(\omega)=C_{c,\mathrm{MIO}}(\omega)$ for any state.

A strong parallel between the theories of coherence and entanglement was noticed when the latter is restricted to the so-called maximally correlated states [68], of the form

$$\rho=\sum_{i,j}\rho_{ij}\,|ii\rangle\!\langle jj| \tag{S156}$$

for some bases of $\mathcal{H}_A$ and $\mathcal{H}_B$ and coefficients ρ_{ij}. Such states are separable if and only if they are diagonal in the given basis, and their operational properties — including the values of entanglement cost and distillable entanglement under LOCC — exactly match the values of coherence cost and distillable coherence of the corresponding single-party state

$$\widetilde{\rho}=\sum_{i,j}\rho_{ij}\,|i\rangle\!\langle j| \tag{S157}$$

when coherence manipulation is considered under the class of incoherent operations (IO) [66, 68].

Our result, however, shows a major difference between the theories of entanglement for maximally correlated states and quantum coherence, and breaks the quantitative equivalence when the transformations under non-entangling operations NE and maximally incoherent operations MIO are considered. Crucially, our counterexample ω_3 is a maximally correlated state. This implies that

$$E_{c,\mathrm{NE}}(\omega_3) > E_{d,\mathrm{NE}}(\omega_3) = C_{d,\mathrm{MIO}}(\widetilde{\omega}_3) = C_{c,\mathrm{MIO}}(\widetilde{\omega}_3), \tag{S158}$$

where the first equality is due to the fact that both $E_{d,\mathrm{NE}}(\rho)$ and $C_{d,\mathrm{MIO}}(\widetilde{\rho})$ are given by the relative entropy of coherence of $\widetilde{\rho}$ [33, 68, 69] for any maximally correlated state. Intuitively, any MIO transformation can be seen to give rise to a transformation which is non-entangling, but only when restricted to the maximally correlated subspace; our result shows that such maps cannot always be extended to a transformation which is non-entangling for any input state, and so NE operations are weaker at manipulating maximally correlated states than MIO operations are at manipulating their corresponding single-party systems.

D. More open questions

Our results motivate a number of extensions and follow-up results that would strengthen the understanding of general entanglement manipulation. We have already remarked several of them throughout this Supplementary Information; let us collect them here and discuss other open questions.

First, although our bound $E_{c,\,\mathrm{NE}}(\rho) \geq E_N^\tau(\rho)$ is good enough to establish the irreversibility of entanglement manipulation, one could ask whether a tighter computable bound can be obtained. For instance, does the regularised quantity $L_{\mathcal{K}}^\tau(\rho)$ in Theorem S7 admit a single-letter expression? Even more ambitiously, one could ask whether an exact expression for the entanglement cost $E_{c,\,\mathrm{NE}}$ itself can be established. Such questions are interesting not only from an axiomatic perspective, but also because any such result would provide improved bounds on $E_{c,\,\mathrm{LOCC}}$.

Additionally, as mentioned in Supplementary Note III, several quantum states which have previously been used as examples of irreversibility under smaller sets of operations are actually reversible under NE and PPTP. An understanding of what exactly makes a state such as ω_3 irreversibile under *all* non-entangling transformations, and — more generally — a complete characterisation of all irreversible states could help shed light on the stronger type of irreversibility that we have revealed in this work.

Another result uncovered in Supplementary Note IV of our work is that, although Brandão and Plenio's framework [10, 47] suggests that entanglement can be reversibly manipulated while generating only small (asymptotically vanishing) amounts of the generalised robustness $R_{\mathcal{S}}^g$, requiring that the standard robustness $R_{\mathcal{S}}^s$ also be small completely breaks reversibility. It would be very interesting to understand exactly why such a 'phase transition' in the task of entanglement dilution occurs, and whether one can tighten our and Brandão and Plenio's results with other choices of monotones.

Also, the phenomenon of catalysis [70] is a remarkable feature that can significantly enhance feasible entanglement transformations: the fact that a transformation $\rho \longrightarrow \omega$ is impossible does not necessarily mean that $\rho\otimes\tau \longrightarrow \omega\otimes\tau$ cannot be accomplished with the same set of allowed processes. One could then ask about transformation rates where, instead of requiring that $\Lambda(\rho^{\otimes n}) \longrightarrow \omega^{\otimes m}$, one asks that $\Lambda(\rho^{\otimes n} \otimes \tau) \longrightarrow \omega^{\otimes m} \otimes \tau$ for some state τ, and define the distillable entanglement E_d and entanglement cost E_c correspondingly. Additionally, the operations NE and PPTP are rather curious in that they are not closed under tensor product, in the sense that $\Lambda, \Lambda' \in \mathrm{NE}$ does not mean that $\Lambda \otimes \Lambda' \in \mathrm{NE}$ when acting on a larger system. Such properties leave open the possibility of significantly different behaviour when catalysis is employed, something that was already remarked in [10, 47].

Finally, a fundamentally important question is: what is it about entanglement that makes it irreversible? As discussed in the main text, several examples of quantum resource theories have been shown to be reversible when all resource–non-generating operations (counterparts to non-entangling or PPT-preserving maps) are allowed [48]. Although there is no a priori reason to expect reversibility to be a generic property of quantum resources, one can note that the framework and main results of Brandão and Plenio may be adapted to more general convex resource theories [71], suggesting [38] that reversibility can be guaranteed when *asymptotically* resource–non-generating transformations are allowed and the generated resource is quantified with a generalised robustness–type measure $R^g_{\mathcal{K}}$. Our question then reduces to: in what types of resources can we enforce strict resource non-generation and still maintain reversibility? Is entanglement truly unique in its general irreversibility?

VII. SUBTLETIES RELATED TO INFINITE DIMENSION

Throughout this section we will elaborate on some issues that arise specifically in dealing with infinite-dimensional spaces.

A. On the definition of partial transpose

The definition of the partial transposition [2] requires some further care when one deals with infinite-dimensional spaces, the main reason being that this operation does not preserve the space of trace class operators. We first define it on the dense subspace $\mathcal{T}(\mathcal{H}_A)\otimes\mathcal{T}(\mathcal{H}_B)\subseteq\mathcal{T}(\mathcal{H}_{AB})$ of finite linear combinations of simple tensors by the expression $\Gamma(X_A\otimes Y_B)=(X_A\otimes Y_B)^\Gamma := X_A\otimes Y_B^\intercal$, already encountered in (S8), extended by linearity.

Now, one observes that the linear map $\Gamma:\mathcal{T}(\mathcal{H}_A)\otimes\mathcal{T}(\mathcal{H}_B)\to\mathcal{B}(\mathcal{H}_{AB})$ we just constructed has operator norm

$$\|\Gamma\|_{1\to\infty} := \sup_{0\neq Z\in\mathcal{T}(\mathcal{H}_A)\otimes\mathcal{T}(\mathcal{H}_B)} \frac{\left\|Z^\Gamma\right\|_\infty}{\|Z\|_1} = 1\,,$$

where $\|Z\|_\infty := \sup_{|\psi\rangle}\|Z\,|\psi\rangle\|$. To see this, it suffices to write the singular value decomposition of any $Z\in\mathcal{T}(\mathcal{H}_A)\otimes\mathcal{T}(\mathcal{H}_B)\subseteq\mathcal{T}(\mathcal{H}_{AB})$ as $Z=\sum_{i=0}^\infty \lambda_i\,|\Psi_i\rangle\!\langle\Phi_i|$, where $\|Z\|_1=\sum_{i=0}^\infty|\lambda_i|$, and then notice that $\left\||\Psi\rangle\!\langle\Phi|^\Gamma\right\|_\infty\leq 1$ for all pairs of pure states $|\Psi\rangle\,,|\Phi\rangle\in\mathcal{H}_{AB}$, so that

$$\left\|Z^\Gamma\right\|_\infty \leq \sum_{i=0}^\infty |\lambda_i| \left\||\Psi\rangle\!\langle\Phi|^\Gamma\right\|_\infty \leq 1\,.$$

Finally, one may use the continuous extension theorem to lift Γ to a continuous map $\Gamma:\mathcal{T}(\mathcal{H}_{AB})\to\mathcal{B}(\mathcal{H}_{AB})$. It still holds that $\|\Gamma\|_{1\to\infty}=1$.

B. Topological properties of the cone of PPT operators

An important property of the two cones of separable and of PPT operators is that they are closed with respect to the topology induced on $\mathcal{T}(\mathcal{H}_{AB})$ by its pre-dual, the Banach space of compact operators on $\mathcal{H}_{AB}$. We will refer to this topology as the *weak*-topology*. The fact that $\mathcal{S}_{AB}$ is weak*-closed has been established in Ref. [6, Lemma 25]. An analogous statement for $\mathcal{PPT}_{AB}$ can be proved even more directly (cf. [72, Lemma 13]).

Lemma S21. *The cone $\mathcal{PPT}_{AB}\subseteq\mathcal{T}(\mathcal{H}_{AB})$ defined by (S7) is weak*-closed, i.e. closed with respect to the topology induced on $\mathcal{T}(\mathcal{H}_{AB})$ by its pre-dual, the Banach space of compact operators on $\mathcal{H}_{AB}$.*

Proof. Note that $\mathcal{PPT}_{AB}=\mathcal{T}_+(\mathcal{H}_{AB})\cap(\Gamma(\mathcal{T}_+(\mathcal{H}_{AB}))\cap\mathcal{T}(\mathcal{H}_{AB}))$, where the intersection with $\mathcal{T}(\mathcal{H}_{AB})$ in the second factor reminds us of the fact that $\Gamma(\mathcal{T}_+(\mathcal{H}_{AB}))\subseteq\mathcal{B}(\mathcal{H}_{AB})$ contains operators that are not of trace class. Since $\mathcal{T}_+(\mathcal{H}_{AB})$ is well known to be weak*-closed, it suffices to show that so is

$\Gamma(\mathcal{T}_+(\mathcal{H}_{AB})) \cap \mathcal{T}(\mathcal{H}_{AB})$ as well. Pick local orthonormal bases $\{|n\rangle_A\}_{n\in\mathbb{N}}$ and $\{|m\rangle_B\}_{m\in\mathbb{N}}$. Let us say that an operator $Y \in \mathcal{T}(\mathcal{H}_{AB})$ has a finite expansion if $\langle n, m|Y|n', m'\rangle \neq 0$ only for a finite number of quadruples $(n, m, n', m') \in \mathbb{N}^4$. If this is the case, it is simple to verify that also the partial transpose Y^Γ has a finite expansion; in particular, both Y and Y^Γ are compact operators.

We now claim that $X \in \mathcal{T}(\mathcal{H}_{AB})$ satisfies that $X \in \Gamma(\mathcal{T}_+(\mathcal{H}_{AB}))$ if and only if $\mathrm{Tr}\left[XY^\Gamma\right] \geq 0$ for all $Y \geq 0$ with a finite expansion. To see why, note that a necessary and sufficient condition for $X^\Gamma \geq 0$ is that all (finite) principal minors of X^Γ are positive. In particular, $X^\Gamma \geq 0$ if and only if $\mathrm{Tr}\left[X^\Gamma Y\right] = \mathrm{Tr}\left[XY^\Gamma\right] \geq 0$ for all $Y \geq 0$ with a finite expansion. Since any such Y^Γ is compact, the functionals $X \mapsto \mathrm{Tr}\left[XY^\Gamma\right]$ are weak*-continuous; we deduce immediately that $\Gamma(\mathcal{T}_+(\mathcal{H}_{AB})) \cap \mathcal{T}(\mathcal{H}_{AB})$ is weak*-closed. This concludes the proof. □

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