No second law of entanglement manipulation after all

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Many fruitful analogies have emerged between the theories of quantum entanglement and thermodynamics, motivating the pursuit of an axiomatic description of entanglement akin to the laws of thermodynamics. A long-standing open problem has been to establish a true second law of entanglement, and in particular a unique function that governs all transformations between entangled systems, mirroring the role of entropy in thermodynamics. Contrary to previous promising evidence, here we show that this is impossible and that no direct counterpart to the second law of thermodynamics can be established. This is accomplished by demonstrating the irreversibility of entanglement theory from first principles. Assuming only the most general microscopic physical constraints of entanglement manipulation, we show that entanglement theory is irreversible under all non-entangling transformations. We furthermore rule out reversibility without significant entanglement expenditure, showing that reversible entanglement transformations require the generation of macroscopically large amounts of entanglement according to certain measures. Our results not only reveal fundamental differences between quantum entanglement transformations and thermodynamic processes, but also showcase a unique property of entanglement that distinguishes it from other known quantum resources.
With the advent of quantum information science, the phenomenon of quantum entanglement emerged as a physical resource in its own right, enabling remarkable advantages in tasks such as communication\(^{10-30}\), computation\(^1\) and cryptography\(^2\). The parallel with thermodynamics prompted a debate concerning the axiomatisation of entanglement theory\(^{13,16}\) and the possible emergence of a single entanglement measure, akin to entropy, which would govern all entanglement transformations and establish the reversibility of this resource\(^1,13,16-18\). Although later results suggested that entanglement may often be quite different from thermodynamics, even exhibiting irreversibility in some of the most practically relevant settings\(^{21}\), hope persisted for an axiomatic framework for entanglement manipulation that would exactly mirror thermodynamic properties. Notably, identifying a unique entropic measure of entanglement was long known to be possible for the special case of pure states\(^ {1,12}\), and several proposals for general reversible frameworks have been formulated\(^ {16,22}\). The seminal work of Brandão and Plenio\(^ {13,16}\) then provided further evidence in this direction by showing that reversible manipulation may\(^1\) be possible when the physical restrictions governing entanglement transformations are suitably relaxed. These findings strengthened the belief that a fully reversible and physically consistent theory of entanglement could be established.

Here, however, we prove a general no-go result showing that entanglement theory is fundamentally irreversible. Equivalently, we show from first principles that entanglement transformations cannot be governed by a single measure, and that an axiomatic second law of entanglement manipulation cannot be established.

Our sole assumption is that entanglement manipulation by separated parties should be accomplished by means of operations that make the theory fully consistent, that is, that never transform an unentangled system into an entangled one. This can be thought of as the analogue in the entanglement setting of the Kelvin–Planck statement of the second law, which in classical thermodynamics forbids the creation of resources (work) from objects that are not resourceful themselves (a single heat bath)\(^ {21}\). By imposing only this requirement, we dispense with the need to make any assumptions about the structure of the considered processes. For example, we do not even posit that all intermediate transformations obey the laws of standard quantum mechanics, as previous works implicitly did. Instead, we only look at the initial and final states of the system, and demand that no resource, in this case entanglement, is generated in the overall transformation. This philosophy, hereafter termed ‘axiomatic’, is analogous to that followed by the pioneers of thermodynamics (and more recently by Lieb and Yngvason)\(^ {5}\) to establish truly universal versions of the second law. Such a general approach allows us to preclude the reversibility of entanglement under all physically motivated manipulation protocols.

Importantly, however, our conclusions remain unaffected even when the above assumptions are substantially relaxed. It is intuitive to ask whether irreversibility could be avoided with just a small amount of generated entanglement, restoring the hope for reversible transformations in practice. We disprove such a possibility by strengthening our result to show that, with a suitable choice of an entanglement measure such as the entanglement negativity\(^7\), it is necessary to generate macroscopically large quantities of entanglement in the process—any smaller amount cannot break the fundamental irreversibility revealed in our work. In particular, as we argue below, macroscopic entanglement generation is the price one would have to pay in Brandão and Plenio’s framework\(^ {13,14}\) to restore reversibility.

The most surprising aspect of our findings is not only the stark contrast with thermodynamics, but rather the fact that several other quantum phenomena, including quantum coherence and purity, have been shown to be reversible in analogous axiomatic settings\(^ {21}\) and that no quantum resource has ever been found to be irreversible under similar assumptions. Our result is thus a first of its kind. It highlights a fundamental difference between entanglement on one side, and thermodynamics and all other quantum resource theories known to date on the other.

The generality of our approach allows for an extension of the results beyond the theory of entanglement of quantum states, to the manipulation of quantum operations\(^ {29}\). This corresponds to the setting of quantum communication, where the resource in consideration is the ability to reliably transmit quantum systems. Importantly, thermodynamics allows for the reversible manipulation of operations\(^ {31}\), as well, so an irreversibility of communication theory is, once again, in heavy contrast to thermodynamics.

**Entanglement manipulation**

The framework of entanglement theory features two separated parties, conventionally named Alice and Bob, who share a large number of identical copies of a bipartite quantum state and wish to transform them into as many copies as possible of some target state, all while achieving a vanishingly small error in the asymptotic limit. We introduce this setting in Fig. 1.

The figure of merit in transforming the input quantum state \(\rho\) into a target state \(\omega\) is the transformation rate \(R(\rho \rightarrow \omega)\), defined as the maximum ratio \(m/n\) that can be achieved in the limit \(n \rightarrow \infty\) under the condition that \(n\) copies of \(\rho\) are transformed into \(m\) copies of \(\omega\) with asymptotically vanishing error. Such a rate depends crucially on the set of allowed operations. In keeping with our axiomatic approach, we consider the largest physically consistent class of transformations, namely those that are incapable of generating entanglement and can only manipulate entanglement already present in the system.

To formalise this, we introduce the set of separable (or unentangled) states on a bipartite system \(AB\), composed of all those states \(\sigma_{AB}\) that admit a decomposition of the form\(^ {11,32}\).
In the asymptotic many-copy limit it is possible to obtain two copies of \( \omega \) from each three copies of \( \rho \), and vice versa.

\[
\sigma_{AB} = \int |{\psi}\rangle \langle {\psi}| \otimes |{\phi}\rangle \langle {\phi}| d\mu({\psi}, {\phi}). \tag{1}
\]

where \( \mu \) is an appropriate probability measure on the set of pairs of local pure states. Our assumption is that any allowed operation \( \Lambda \) should transform quantum states on \( AB \) into valid quantum states on some output system \( A' B' \), in such a way that \( \Lambda(\sigma_{AB}) \) is separable for all separable states \( \sigma_{AB} \). We refer to such operations as non-entangling (NE). They are also known as separability preserving. Hereafter, all transformation rates are understood to be with respect to this family of protocols.

We say that two states \( \rho, \omega \) can be interconverted reversibly if \( R(\rho \rightarrow \omega)R(\omega \rightarrow \rho) = 1 \) (Fig. 2). However, to demonstrate or disprove reversibility of entanglement theory as a whole, it is not necessary to check all possible pairs \( \rho, \omega \). Instead, we can fix one of the two states, say the second, to be the standard unit of entanglement, the two-qubit maximally entangled state ('entanglement bit') \( \Phi_2 := \frac{1}{\sqrt{2}} \sum_{i,j=1}^{2} |ii\rangle \langle jj| \).

The two quantities \( E_d(\rho) := R(\rho \rightarrow \Phi_2) \) and \( E_e(\rho) := R(\Phi_2 \rightarrow \rho)^{-1} \) are referred to as the distillable entanglement and the entanglement cost of \( \rho \), respectively. Entanglement theory is then reversible if \( E_e(\rho) = E_d(\rho) \) for all states \( \rho \).

**Irreversibility of entanglement manipulation**

By demonstrating an explicit example of a state that cannot be reversibly manipulated, we will show that reversibility of entanglement theory cannot be satisfied in general. We formalise this as follows.

**Theorem 1.** The theory of entanglement manipulation is irreversible under NE operations. More precisely, for the two-qutrit state \( \omega_3 = \frac{1}{8} \sum_{i,j=1}^{3} (|ii\rangle \langle ii| - |ii\rangle \langle jj|) \), it holds that

\[
E_e(\omega_3) = 1 > E_d(\omega_3) = \log_2(3/2). \tag{2}
\]

To show this result, we introduce a general lower bound on the entanglement cost \( E_e \) that can be efficiently computed as a semi-definite program. Our approach relies on a new entanglement monotone that we call the 'tempered negativity', defined through a suitable modification of a well-known entanglement measure called the negativity\(^7\). The situation described by Theorem 1 is depicted in Fig. 3. The full proof of the result is sketched in Methods section and described in detail in Supplementary Notes IV–VI as follows: (1) We show that irreversibility persists beyond NE transformations. The conclusion of Theorem 1 holds even when we allow for the generation of small amounts of entanglement (sub-exponential in the number of copies of the state), as quantified by several choices of entanglement measures such as the negativity or the standard robustness of entanglement\(^\text{19} \). What this means is that, to reversibly manipulate the state \( \omega \), one would need to generate macroscopic (exponential) amounts of entanglement. (2) We furthermore show that the irreversibility cannot be alleviated by allowing for a small, non-vanishing error in the asymptotic transformation—a property known as pretty strong converse\(^\text{19} \). (3) Finally, Theorem 1 can also be extended to the theory of point-to-point quantum communication, exploiting the connections between entanglement manipulation and communication schemes\(^\text{20,22} \). This is considered in detail in a follow-up work\(^\text{23} \). These extensions further solidify the fundamental character of the irreversibility uncovered in our work, showing that it affects both quantum states and channels, and that there are no ways to avoid it without incurring very large transformation errors or generating substantial amounts of entanglement.

**Why NE transformations?**

The intention behind our general, axiomatic framework is to prove irreversibility in as broad a setting as possible. The key strength of this approach is that irreversibility under the class of NE transformations enforces irreversibility under any smaller class of processes, which includes the vast majority of different types of operations studied in the manipulation of entanglement\(^\text{20,22} \). Furthermore, our result shows that even enlarging the previously considered classes of processes cannot enable reversibility, as long as the resulting transformations are NE.

To better understand the need for and the consequences of such a general approach, let us compare our framework with another commonly employed model, that where entanglement is manipulated by means of local operations and classical communication (LOCC). In this context, irreversibility was first found by Vidal and Cirac\(^\text{19} \). Albeit historically important, the LOCC model is built with a bottom-up mindset, and rests on the assumption that the two parties can only employ local quantum resources at all stages of the protocol. Already in the early days of quantum information, it was realised that relaxing such restrictions, for example by supplying some additional resources, can lead to improvements in the capability to manipulate entanglement\(^\text{21,22} \). Although attempts to construct a reversible theory of entanglement along these lines have been unsuccessful\(^\text{23} \), the assumptions imposed therein have left open the possibility of the existence of a larger class of operations that could remedy the irreversibility.

The limitations of such bottom-up approaches are best illustrated with a thermodynamical analogy: in this context, they would lead to operational statements of the second law concerning, say, the impossibility of realising certain transformations by means of mechanical
processes but would not tell us much about electrical or nuclear processes. Indeed, since we have no guarantee that the ultimate theory of Nature will be quantum mechanical, it is possible to envision a situation where, for instance, the exploitation of some exotic physical phenomena by one of the parties could enhance entanglement transformations. To construct a theory as powerful as thermodynamics, we followed instead a top-down, axiomatic approach, which – as discussed above – imposes only the weakest possible requirement on the allowed transformations, thereby ruling out reversibility under any physical processes.

The NE operations considered here are examples of ‘resource non-generating operations’, commonly employed in the study of many other quantum resource theories. In all of these other contexts, such operations have always been shown to lead to the reversibility of the given theory. For instance, Gibbs-preserving maps in quantum thermodynamics are a broad, axiomatic formulation of the constraints governing thermodynamic transformations of quantum systems analogous to NE operations. Under such operations, the theory of thermodynamics is known to be reversible. An equivalent result has also been shown in the resource theory of quantum coherence, suggesting that reversibility could be a generic feature in the manipulation of different resources under all resource non-generating transformations. Our result, however, shows entanglement theory to be fundamentally different from thermodynamics and from all other known quantum resources: not even the vast class of all NE maps can enable reversible entanglement manipulation. What this means is that, under the exact same assumptions that suffice to facilitate the reversibility of other quantum resources, entanglement remains irreversible.

Macroscopic entanglement generation is necessary for reversibility

A similar axiomatic mindset to the one employed in our work has already proved to be useful. Notably, it led Brandão and Plenio to construct a theory of entanglement that was claimed to be fully reversible. Recently, an issue that casts some doubts on the validity of their mathematical proof has transpired. In spite of this, it remains a possibility that the theory of entanglement proposed by Brandão and Plenio may actually be reversible, so let us discuss it here in detail. This theory features so-called asymptotically NE operations, defined as those that may generate some limited amounts of entanglement, provided that any such supplied resources are vanishingly small in the asymptotic limit. This, on the surface, appears consistent with how fluctuations are treated in the theory of thermodynamics. However, the key question to ask here is: according to what measure should one enforce the generated entanglement to be small? Brandão and Plenio choose to quantify entanglement with the standard robustness, so-called asymptotically operationally justified reason to prefer the generalised robustness over the other monotones. If anything, the most operationally meaningful monotones to select here would be those defined directly in terms of practical tasks, such as the entanglement cost $E$. Indeed, the choice of $M$ turns out to be pivotal. Brandão and Plenio’s main result claims that, with the specific choice of $M$ being the generalised robustness of entanglement, entanglement can be manipulated reversibly even if we take $\delta_n > 0$ as $n \to \infty$. In stark contrast, we now show that a completely opposite conclusion is reached when $M$ is taken to be either the standard robustness or the entanglement negativity.

Theorem 2. The theory of entanglement manipulation is irreversible under operations that generate sub-exponential amounts of entanglement according to the negativity $N$ or the standard robustness $R$. Specifically, if $M = N$ or $M = R$, then for any sequence $(\delta_n)_n$ such that $\delta_n \to 0$ as $n \to \infty$, it holds that

$$E_{c, \mathrm{NE}}(\rho) = 1 > E_{c, \mathrm{NE}}(\rho) = \log_2(3/2).$$

Comparing this with Brandão and Plenio’s conclusion, we can observe that the operations employed there may only hope to achieve reversibility by generating exponential amounts of entanglement, as measured by either the negativity or the standard robustness.

We stress that there is no a priori operationally justified reason to prefer the generalised robustness over the other monotones. If anything, the most operationally meaningful monotones to select here would be those defined directly in terms of practical tasks, such as the entanglement cost $E$. Indeed, following this route actually trivialises the theory, entailing that different choices of monotones need to be employed to give meaningful results. Even between the generalised robustness (as employed by Brandão and Plenio) and the standard robustness $R$, it is actually the latter that admits a clearer operational interpretation in this context. $R$ quantifies exactly the entanglement cost of a state in the one-shot setting, when asymptotic transformations are not allowed. These ambiguities in the choice of a ‘good’ measure, and the vastly disparate physical consequences of the different choices, put the physicality of the reversibility result claimed by Brandão and Plenio into question: why should one such framework be considered more physical than the other, irreversible ones?

Importantly, since the core concept of separability is independent of the particular choice of a measure, our axiomatic assumption of strict no entanglement generation bypasses the above problems completely. It removes the dependence on any entanglement measure
and ensures that the physical constraints are enforced at all scales, therefore yielding an unambiguously physical model of general entanglement transformations. However, should such a requirement be considered too strict, our Theorem 2 shows that irreversibility of entanglement is robust to fluctuations in the generated resources. Let us also point out that the assumptions of Brandão and Plenio (and of Theorem 2) are in fact more permissive than those typically employed in quantum thermodynamic frameworks [35,41,42], where one usually allows fluctuations in the sense of the consumption of small ancillary resources, but not fluctuations in the very physical laws governing the process. In a thermodynamic sense, entanglement transformations under approximately NE maps could be compared to the manipulation of systems under transformations that do not conserve the overall energy—a relaxation that would go against standard axiomatic assumptions [36,37] or other known quantum resources [38].

Discussion

Our results close a major open problem in the characterisation of entanglement [39] by showing that a reversible theory of this resource cannot be established under any set of ‘free’ transformations that do not generate entanglement. Indeed, from our characterisation, we can conclude not only that entanglement generation is necessary for reversibility, but also that macroscopically large amounts of it must be supplemented. This shows that the framework proposed by Brandão and Plenio [32,34] is effectively the smallest possible one that could allow reversibility, although only at the cost of substantial entanglement expenditure. That the seemingly small revision of the underlying technical assumptions we advocated by enforcing strict entanglement non-generation can have such far-reaching consequences, namely precluding reversibility, is truly unexpected. In fact, as remarked above, the opposite of this phenomenon has been observed in a number of fundamentally important quantum resource theories, where the set of all resource non-generating operations suffices to enable reversibility. It is precisely the necessity to generate entanglement in order to reversibly manipulate it that distinguishes the theory of entanglement from thermodynamics and other quantum resources. This fundamental difference contrasts not only with the previously established information-theoretic parallels, but also with the many links that have emerged between entanglement and thermodynamics in broader contexts such as many-body and relativistic physics [40–42]. It then becomes an enthralling foundational problem to understand what makes entanglement theory special in this respect, and where its fundamental irreversibility may come from. Additionally, the axiomatic theory of entanglement manipulation delineated here leaves several outstanding follow-up questions. For instance, it would be very interesting to understand whether a closed expression for the associated entanglement cost can be established, and whether the phenomenon of entanglement catalysis [43–45] can play a role in this setting.

We remark that the recently identified gap in Brandão and Plenio’s proof [46], which came to light after this work was completed, does not affect our results or conclusions in any way, since the methods that we use are independent of those in refs. [23,24]. Our main finding—that of entanglement irreversibility under NE operations—is complementary to the result of Brandão and Plenio [32,34], as discussed above and in Supplementary Notes IV and V. This recent development does, however, rekindle the question of whether entanglement can be reversibly manipulated whatsoever [33], even in a more permissive framework such as Brandão and Plenio’s.

In conclusion, we have highlighted a fundamental difference between the theory of entanglement manipulation and thermodynamics, proving that no microscopically consistent second law can be established for the former. At its heart, our work reveals an inescapable restriction precipitated by the laws of quantum physics—one that has no analogue in classical theories and that was previously unknown even within the realm of quantum theory.

Online content

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-022-01873-9.

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Methods
In the following, we sketch the main ideas needed to arrive at a proof of our main result in Theorem 1 and extensions thereof.

Asymptotic transformation rates under NE operations
We start by defining rigorously the fundamental quantities we are dealing with. Given two separable Hilbert spaces \(\mathcal{H}\) and \(\mathcal{H}'\) and the associated spaces of trace class operators \(\mathcal{T}(\mathcal{H})\) and \(\mathcal{T}(\mathcal{H}')\), a linear map \(A : \mathcal{T}(\mathcal{H}) \to \mathcal{T}(\mathcal{H}')\) is said to be positive and trace preserving if it transforms density operators on \(\mathcal{H}\) into density operators on \(\mathcal{H}'\). As is well known, physically realisable quantum operations need to be completely positive and not merely positive. While we could enforce this additional assumption without affecting any of our results, we will only need to assume the positivity of the transformations, establishing limitations also for processes more general than quantum channels.

Since we are dealing with entanglement, we need to make both \(\mathcal{H}\) and \(\mathcal{H}'\) bipartite systems. We shall therefore assume that \(\mathcal{H} = \mathcal{H}_A \otimes \mathcal{H}_B\) and \(\mathcal{H}' = \mathcal{H}_A' \otimes \mathcal{H}_B'\) have a tensor product structure. Separable states on \(AB\) are defined as those that admit a decomposition as in equation (1). A positive trace-preserving operation \(A : \mathcal{T}(\mathcal{H}_A \otimes \mathcal{H}_B) \to \mathcal{T}(\mathcal{H}_A' \otimes \mathcal{H}_B')\), which we shall denote compactly as \(A_{AB \to A'B'}\), is said to be NE or separability preserving if it transforms separable states on \(AB\) into separable states on \(A'B'\). We will denote the set of NE operations from \(AB\) to \(A'B'\) as \(NE_{AB \	o A'B'}\).

The central questions in the theory of entanglement manipulation are the following. Given a bipartite state \(\rho_{AB}\) and a set of quantum operations, how much entanglement can be extracted from \(\rho_{AB}\)? How much entanglement does it cost to generate \(\rho_{AB}\) in the first place? The ultimate limitations to these two processes, called entanglement distillation and entanglement dilution, respectively, are well captured by looking at the asymptotic limit of many copies. As remarked above, this procedure is analogous to the thermodynamic limit. The resulting quantities are called the distillable entanglement and the entanglement cost, respectively. We already discussed their intuitive operational definitions, so we now give their mathematical forms:

\[
E_d(\rho_{AB}) := \sup \left\{ R > 0 : \lim_{n \to \infty} \inf_{\Lambda_n \in \mathcal{NE}_{AB \to A'B'}} \left\| \left( \rho_{AB}^{\otimes n} \right)^{\otimes n} - \Phi_2^{\otimes n} \right\|_1 = 0 \right\},
\]

(4)

\[
E_c(\rho_{AB}) := \inf \left\{ R > 0 : \lim_{n \to \infty} \inf_{\Lambda_n \in \mathcal{NE}_{AB \to A'B'}} \left\| \left( \Phi_2^{\otimes n} \right)^{\otimes n} - \rho_{AB}^{\otimes n} \right\|_1 = 0 \right\}.
\]

(5)

Here, \(A'B'\) is the system composed by \(n\) copies of \(AB\), \(A_nB_n\) denotes a fixed two-qubit quantum system, and \(\Phi_2 = \{\Phi_2, \Phi_3\}\), with \(\Phi_2 = \frac{1}{2} (|00\rangle + |11\rangle)\), is the maximally entangled state of \(A_nB_n\), also called the ‘entanglement bit’.

One question that could be raised at this point is: is our definition of transformation rates not too restrictive? Such a reservation could be motivated by the fact that, for example, in the resource theory of quantum thermodynamics, employing only energy-conserving unitary transformations is known to be insufficient to achieve general transformations. To avoid this issue, additional resources are provided in the form of ancillary systems composed of a sublinear number of qubits, allowing one to circumvent the restrictions of energy conservation without affecting the underlying physics. Such an approach can be adapted to more general resources. In our setting, however, this is already implicitly included in the definition of \(E_d\) and \(E_c\), since such ancillary systems can be absorbed into the asymptotic transformation rates. That is, we could have equivalently defined

\[
E_d(\rho_{AB}) := \sup \left\{ R > 0 : \lim_{n \to \infty} \inf_{\Lambda_n \in \mathcal{NE}_{AB \to A'B'}} \left\| \left( \rho_{AB}^{\otimes n} \otimes \tau_n \right)^{\otimes n} - \Phi_2^{\otimes n} \right\|_1 = 0 \right\},
\]

(6)

\[
E_c(\rho_{AB}) := \inf \left\{ R > 0 : \lim_{n \to \infty} \inf_{\Lambda_n \in \mathcal{NE}_{AB \to A'B'}} \left\| \left( \Phi_2^{\otimes n} \otimes \tau_n \right)^{\otimes n} - \rho_{AB}^{\otimes n} \right\|_1 = 0 \right\}.
\]

(7)

where \(\tau_n\) are arbitrary (possibly entangled) systems such that \(\dim \tau_n = 2^{o(n)}\). The rates are not affected by the addition of such an ancilla, since its sub-exponential size means that any contributions to the rate due to \(\tau_n\) will vanish asymptotically. This is addressed in more detail in Supplementary Note V.

The main idea: tempered negativity
Let us commence by looking at a well-known entanglement measure called the logarithmic negativity. For a bipartite state \(\rho_{AB}\), this is formally defined by

\[
E_N(\rho_{AB}) := \log_2 \left\| \rho_{AB}^\Gamma \right\|_1,
\]

(8)

where \(\Gamma\) denotes the partial transpose, that is, the linear map \(\Gamma : \mathcal{T}(\mathcal{H}_A' \otimes \mathcal{H}_B) \to \mathcal{T}(\mathcal{H}_A \otimes \mathcal{H}_B')\), where \(\mathcal{B}(\mathcal{H}_A' \otimes \mathcal{H}_B)\) is the space of bounded operators on \(\mathcal{H}_A' \otimes \mathcal{H}_B\), that acts as \(\Gamma(\chi_{i} \otimes \rho_{j}) = \chi_{i} \otimes \rho_{j}^\Gamma\) with superscript ‘\(\Gamma\)’ denoting transpose with respect to a fixed basis, and is extended by linearity and continuity to the whole \(\mathcal{B}(\mathcal{H}_A' \otimes \mathcal{H}_B)\) (ref. 1). It is understood that \(E_N(\rho_{AB}) = 0\) if \(\rho_{AB}^\Gamma\) is not of trace class. Remarkably, the logarithmic negativity does not depend on the basis chosen for the transposition. Also, since \(\rho_{AB}^\Gamma\) is a valid state for any separable \(\sigma_{AB}\) (ref. 5), this measure vanishes on separable states, that is,

\[
\sigma_{AB} \text{ is separable} \Rightarrow \left\| \sigma_{AB}^\Gamma \right\|_1 = 1 \Rightarrow E_N(\sigma_{AB}) = 0.
\]

(9)

Given a non-negative real-valued function on bipartite states \(E\) that we think of as an ‘entanglement measure’, when can it be used to give bounds on the operationally relevant quantities \(E_d\) and \(E_c\)? It is often claimed that, for this to be the case, \(E\) should obey, among other things, a particular technical condition known as asymptotic continuity. Since a precise technical definition of this term is not crucial for this discussion, it suffices to say that it amounts to a strong form of uniform continuity, in which the approximation error does not grow too large in the dimension of the underlying space. While asymptotic continuity is certainly a critical requirement in general, it is not always indispensable. The starting point of our approach is the elementary observation that the logarithmic negativity \(E_N\) for instance, is not asymptotically continuous, yet it yields an upper bound on the distillable entanglement. The former claim can be easily understood by casting equation (8) into the equivalent form

\[
E_N(\rho_{AB}^\Gamma) = \log_2 \sup \left\{ \text{Tr} \chi : \left\| \chi^\Gamma \right\|_1 \leq 1 \right\}.
\]

(10)

where \(\| Z \|_1 := \sup_{\| \psi \| = 1} \| Z \| \langle \psi \rangle\) is the operator norm of \(Z\), and the supremum is taken over all normalised state vectors \(\psi\). Since the trace norm and the operator norm are dual to each other, the continuity of \(E_N\) with respect to the trace norm is governed by the operator norm of \(X\) in the optimisation in equation (10). However, while the operator norm of \(X^\Gamma\) is at most 1, that of \(X\) can only be bounded as \(\| X \|_{\infty} \leq d \| X^\Gamma \|_1 \leq d\), where \(d := \min \{ \dim(\mathcal{H}_A), \dim(\mathcal{H}_B)\}\) is the minimum of the local dimensions. This bound is generally tight. Since \(d\) grows exponentially in the
number of copies, it implies that $E_N$ is not asymptotically continuous.

But then why is it that $E_3$ still gives an upper bound on the distillable entanglement? A careful examination of the proof by Vidal and Werner\(^{21}\) (see the discussion surrounding equation (46) therein) reveals that this is only possible because the exponentially large number $d$ actually matches the value taken by the supremum in equation (10) on the maximally entangled state, that is, on the target state of the distillation protocol. Let us try to adapt this capital observation to our needs. Since we want to employ a negativity-like measure to lower bound the entanglement cost instead of upper bounding the distillable entanglement, we need a substantial modification.

The above discussion inspired our main idea. Let us tweak the variational program in equation (10) by imposing that the operator norm of $X$ be controlled by the final value of the program itself. The logic of this reasoning may seem circular at first sight, but we will see that it is not so. For two bipartite states $\rho_{AB}$, $\omega_{AB}$, we define the tempered negativity by

$$N_1(\rho,\omega) := \sup \{ \text{Tr} X \varphi : \| X \|_\infty \leq 1, \| X \|_\infty = \text{Tr} X \omega \},$$

and the corresponding tempered logarithmic negativity by

$$E_1(\rho) := \log_2 N_1(\rho).$$

This definition encapsulates the above idea of tying together the value of the function and its continuity properties, and indeed will turn out to yield the desired lower bound on the entanglement cost. Note the critical fact that, in the definition of $N_1(\rho)$, the operator norm of $X$ is given precisely by the value of $N_1(\rho)$ itself.

Properties of the tempered negativity

The tempered negativity $N_1(\rho,\omega)$ given by equation (11) can be computed as a semi-definite program for any given pair of states $\rho$ and $\omega$, which means that it can be evaluated efficiently (in time polynomial in the dimension\(^{19}\)). Moreover, it obeys three fundamental properties, the proofs of which can be found in Supplementary Note II. In what follows, the states $\rho_{AB}$, $\omega_{AB}$ are entirely arbitrary.

(a) Lower bound on negativity: $\| \rho X \|_1 \geq N_1(\rho,\omega)$, and in fact $\| \rho X \|_1 = \sup_{\omega_{AB}} N_1(\rho,\omega)$.

(b) Super-additivity:

$$N_1(\rho^{\otimes n}) \geq n N_1(\rho), \quad E_1(\rho^{\otimes n}) \geq n E_1(\rho).$$

(c) The ‘$e$-lemma’:

$$\frac{1}{2} \| \rho - \omega \|_1 \leq e \Rightarrow N_1(\rho,\omega) \geq (1 - 2e) N_1(\omega).$$

The tempered negativity, just like the standard (logarithmic) negativity, is monotonic under several sets of quantum operations commonly employed in entanglement theory, such as LOCC or positive partial transpose operations\(^{20}\), but not under NE operations. Quite remarkably, it still plays a key role in our approach.

Sketch of the proof of Theorem 1

To prove Theorem 1, we start by establishing the general lower bound

$$E_3(\rho_{AB}) \geq E_1(\rho_{AB})$$

on the entanglement cost of any state $\rho_{AB}$ under NE operations. To show the inequality (16), let $R > 0$ be any number belonging to the set in the definition of $E_1$ in equation (5). In quantum information, this is known as an achievable rate for entanglement dilution. By definition, there exists a sequence of NE operations $A_\nu \in \text{NE}(X_{\rho_{AB}} \rightarrow A^\nu B^\nu)$ such that $\epsilon_n := \frac{1}{d} \sum_{\nu=1}^d (X_{\rho_{AB}})_{ii} (X_{\rho_{AB}})_{jj} \rightarrow 0$, where we used the notation $\Phi_A := \frac{1}{d} \sum_{ij} (|ii\rangle \langle jj|)_{ii}$ for a two-qudit maximally entangled state, and observed that $\Phi_A \neq \Phi_2$.

A key step in our derivation is to write $\Phi_A$—which is, naturally, a highly entangled state—as the difference of two multiples of separable states. (In fact, this procedure leads to the construction of a related entanglement monotone called the standard robustness of entanglement\(^{39}\). We consider it in detail in the Supplementary Information.) It has long been known that this can be done by setting

$$\sigma_+ := \frac{1}{d} \Phi_A, \quad \sigma_- := \frac{1}{d-1} \Phi_A, \quad \Phi_A = d \sigma_+ - (d-1) \sigma_-.$$  

(17)

where $\sigma_\pm$ stands for the identity on the two-qudit, $d^2$-dimensional Hilbert space. Crucially, both $\sigma_\pm$ are separable\(^{40}\). Applying a NE operation $A$ acting on a two-qudit system yields $A(\Phi_A) = d (\sigma_+) - (d-1) (A(\sigma_-))$. Since $A(\sigma_-)$ are again separable, we can then employ the observation that $\| A(\sigma_-) \|_1 = 1$ for separable states (recall equation (9)) together with the triangle inequality for the trace norm, and conclude that

$$\| A(\Phi_A) \|_1 \leq 2d - 1.$$  

(18)

We are now ready to present our main argument, expressed by the chain of inequalities

$$2^{R_{0,1}+1} \geq \| A(\Phi_A) \|_1 \geq \| N_1(\Phi_A) \|_1 \geq (1 - 2e) \| \rho^{\otimes n} \|_1 \geq \| N_1(\rho^{\otimes n}) \|_1 \geq n E_1(\rho).$$

(19)

derived using the inequality (18) together with the above properties (a)–(c) of the tempered negativity. Evaluating the logarithm of both sides, diving by $n$ and taking the limit $n \to \infty$ gives $R \geq E_1(\rho)$. A minimisation over the achievable rates $R > 0$ then yields inequality (16), according to the definition of $E_1$ in equation (5).

We now apply inequality (16) to the two-qudit state

$$\rho_3 = \frac{1}{2} P_3 - \frac{1}{2} \Phi_3 = \frac{1}{6} \sum_{i,j=1}^3 (|ii\rangle \langle jj| - |ii\rangle \langle jj|),$$

(19)

where $P_3 := \sum_{i=1}^3 |ii\rangle \langle ii|$. To compute its tempered logarithmic negativity, we construct an ansatz for the optimisation in the definition in equation (11) of $N_1$ by setting $X_\nu := 2 P_3 - 3 \Phi_3$. Since it is straightforward to verify that $\| X_\nu \|_\infty = 1$ and $\| X_\nu \|_1 = 2$, this yields

$$E_1(\rho_3) \geq E_3(\rho_3) \geq 1.$$  

(20)

In Supplementary Note III, we show that the above inequalities are in fact all equalities.

It remains to upper bound the distillable entanglement of $\omega_3$. This can be done by estimating its relative entropy of entanglement\(^{22}\), which quantifies its distance from the set of separable states as measured by the quantum relative entropy\(^{43}\). Simply taking the separable state $P_3/3$ as an ansatz shows that

$$E_3(\omega_3) \leq \log_2 \frac{3}{2},$$  

(21)

and once again this estimate turns out to be tight. Combining the inequalities (20) and (21) demonstrates a gap between $E_3$ and $E_1$, thus
proving Theorem 1 on the irreversibility of entanglement theory under NE operations.

**Consequences and further considerations**

Our results explicitly show that there cannot exist a single quantity that governs asymptotic entanglement transformations, thus ruling out a second law of entanglement theory under NE operations. Specifically, it is already known that, were such a quantity to exist, it would have to equal the regularised relative entropy of entanglement \( E_{\text{rel}}(\rho) \) (refs. 14-16). But then consider the fact that \( E_{\text{rel}}(\rho^3) = 2 \) while, as we show in Supplementary Note III, \( E_{\text{rel}}(\rho^3) = 3 \log_2 \frac{3}{2} \approx 1.75 \). Thus, if the second law held, then from two copies of \( \rho \), one should be able to obtain three copies of \( \rho \). But Theorem 1 explicitly shows that only two copies of \( \rho \) can be obtained from two copies of \( \Phi \).

An interesting aspect of our lower bound on the entanglement cost in the inequality (20) is that it can therefore be strictly better than the (regularised) relative entropy bound. Previously known lower bounds on entanglement cost that can be computed in practice are actually worse than the relative entropy13,15, which means that our methods provide a bound that both is computable and can improve on previous approaches.

As a final remark, we note that, instead of the class of NE (separability-preserving) operations, we could have instead considered all positive partial transpose-preserving maps, which are defined as those that leave invariant the set of states whose partial transpose is positive. Within this latter approach, we are able to establish an analogous irreversibility result for the theory of entanglement manipulation, recovering and strengthening the findings of Wang and Duan15. Explicit details are provided in the Supplementary Information.

**Necessity of macroscopic entanglement generation**

In Theorem 2, we strengthen the result of Theorem 1 further by considering operations that are not required to be NE but only approximately so, allowing for the possibility of microscopic fluctuations in the form of small amounts of entanglement being generated. As discussed in the main text, this mirrors the approach taken by Brandão and Plenio12-15, where reversibility of entanglement was claimed under similar constraints. The reason we call that framework into question is that the entanglement generated by the ‘asymptotically NE maps’ based on NE operations, with \( \delta \rightarrow 0 \), coincides with its regularised relative entropy of entanglement. In the case of \( \omega \), this equals \( \log_2(3/2) \), which matches its distillable entanglement. Therefore, while we still lack a general proof of reversibility that holds for all states, at least \( \omega \) is a reversible state under Brandão and Plenio’s asymptotically NE operations, provided that one makes the choice \( M = \rho^d \).

However, modifying this choice ever so slightly by picking the standard instead of the generalised robustness shatters reversibility altogether. The choice of the measure in (22) is, for all intents and purposes, a free parameter, and—as we just showed—a crucial one, on which the conclusion hinges. This ambiguity is precisely why no one framework of this type can be deemed more physical than another. There does not appear to be a reason to consider \( M = \rho^d \) a better motivated choice than \( M = \rho^3 \). Due to the inability to unambiguously define a sensible notion of ‘small’ entanglement, especially when the macroscopic limit is involved, we thus posit that the only way to enforce fully physically consistent manipulation of entanglement is to forbid any entanglement generation whatsoever, as we have done in our approach based on NE operations.

**Extension to quantum communication**

The setting of quantum communication is a strictly more general framework in which the manipulated objects are quantum channels themselves. Specifically, consider the situation where the separated parties Alice and Bob are attempting to communicate through a noisy quantum channel \( \Lambda : \mathcal{H}_A \rightarrow \mathcal{H}_B \). To every such channel we associate its Choi–Jamiołkowski state, defined through the application of the channel \( \Lambda \) to one half of a maximally entangled state: \( \rho_m := |\psi^+\rangle \langle \psi^+| \). This state encodes all the information about a given channel20,21. The parallel with entanglement manipulation is then made clear by noticing that communicating one qubit of information is equivalent to Alice and Bob realising a noiseless qubit identity channel, \( I_d \). However, the Choi–Jamiołkowski operator \( I_d \) is just the maximally entangled state \( |\psi^+\rangle \). so the process of quantum communication can be understood as Alice and Bob trying to establish a ‘maximally entangled state’ in the form of a noiseless communication channel. The distillable entanglement in this setting is the (two-way assisted) quantum capacity of the channel20-22, corresponding to the rate at which maximally entangled states can be extracted by the separated parties, and therefore the rate at which quantum information can be sent through the channel with asymptotically vanishing error. In a similar way, we can consider the entanglement cost of the channel20, that is, the rate of pure entanglement that needs to be used to simulate the channel \( \Lambda \). Theorem 2 then tells us that, as long as the generated entanglement stays sub-exponential according to \( M = \rho^m \), reversibility persists. The key step in proving this result is an approximate monotonicity of the two measures under all \( (M, \delta_n) \)-approximately NE operations. Specifically, we can show that, under the application of any map \( A_n \) satisfying (22), the corresponding measure cannot increase by more than a factor \( O(1 + \delta_n) \). But if \( \delta_n \approx 2^{-m} \), then any such additional term will vanish in the limit \( n \rightarrow \infty \), meaning that the basic idea of our proof of Theorem 1 can be applied almost unchanged, as the asymptotic bounds will not be affected by \( (M, \delta_n) \) entanglement generation. A full discussion of the proof and the requirements on entanglement creation required to achieve reversibility can be found in Supplementary Note IV.

This contrasts with the result claimed by Brandão and Plenio12-15, where choosing as \( M \) the generalised robustness \( R^2 \) is conjectured to yield full reversibility of the theory. In support of this conjecture, note that, due to Brandão and Plenio’s result concerned with entanglement dilution whose proof is not affected by the aforementioned issue12-15—the entanglement cost of an arbitrary state under \( (R^2, \delta_n) \) approximately NE operations, with \( \delta_n \rightarrow 0 \), coincides with its regularised relative entropy of entanglement. In the case of \( \omega \), this equals \( \log_2(3/2) \), which matches its distillable entanglement. Therefore, while we still lack a general proof of reversibility that holds for all states, at least \( \omega \) is a reversible state under Brandão and Plenio’s asymptotically NE operations, provided that one makes the choice \( M = \rho^d \).

As a final remark, we note that, instead of the class of NE (separability-preserving) operations, we could have instead considered all positive partial transpose-preserving maps, which are defined as those that leave invariant the set of states whose partial transpose is positive. Within this latter approach, we are able to establish an analogous irreversibility result for the theory of entanglement manipulation, recovering and strengthening the findings of Wang and Duan15. Explicit details are provided in the Supplementary Information.
We sketch the basic idea here, as it is very similar to the approach we took for quantum states above. The complete details of the proof in the channel setting will be published elsewhere.29

The major difference between quantum communication and the manipulation of static entanglement arises in the way that Alice and Bob can implement the processing of their channels. Having access to \( n \) copies of a quantum state \( \rho_{AB} \) is fully equivalent to having the tensor product \( \rho_{AB}^n \) at one’s disposal, but the situation is more complex when \( n \) uses of a quantum channel \( \Lambda \) are available, as they can be exploited in many different ways: in parallel as \( \Lambda^n \), or sequentially, where the output of one use of the channel can be used to influence the input to the subsequent uses, or even in more general ways that do not need to obey a fixed causal order between channel uses, and can exploit phenomena such as superposition of causal orders.27,28 This motivates us, once again, to consider a general, axiomatic approach that covers all physically consistent ways to manipulate quantum channels, as long as they do not generate entanglement between Alice and Bob if it was not present in the first place. Specifically, we will consider the following: Given \( n \) channels \( \Lambda_1, \ldots, \Lambda_n \), we define an \( n \)-channel quantum process to be any \( n \)-linear map \( \mathcal{Y} \) such that \( \mathcal{Y}(\Lambda_1, \ldots, \Lambda_n) \) is also a valid quantum channel. Now, channels \( \Gamma_1, \ldots, \Gamma_n \) such that \( \Gamma_j \) is separable are known as entanglement-breaking channels.21 We define a NE process to be one such that \( \mathcal{Y}(\Gamma_1, \ldots, \Gamma_n) \) is entanglement breaking whenever \( \Gamma_1, \ldots, \Gamma_n \) are all entanglement breaking.

The quantum capacity \( Q(\Lambda) \) is then defined as the maximum rate \( R \) at which NE \( n \)-channel processes can establish the noiseless communication channel \( \mathcal{I}_d^\otimes n \) when the channel \( \Lambda \) is used \( n \) times. As in the case of quantum state manipulation, the transformation error here is only required to vanish asymptotically. Analogously, the (parallel) entanglement cost \( E(\Lambda) \) is given by the rate at which noiseless identity channels \( I_d \) are required in order to simulate parallel copies of the given communication channel \( \Lambda \).

The first step of the extension of our results to the channel setting is then conceptually simple: we define the tempered negativity of a channel as

\[
E_n^t(\Lambda) := \sup_{\rho \in \mathcal{S}} E_n^t(I_d \otimes \Lambda(\rho)),
\]

where the supremum is over all bipartite quantum states \( \rho \in \mathcal{S}(\mathcal{H}_A \otimes \mathcal{H}_B) \) on two copies of the Hilbert space of Alice’s system. A careful extension of the arguments we made for states—accounting in particular for the more complicated topological structure of quantum channels—can be shown to give

\[
E(\Lambda) \geq E_n^t(\Lambda)
\]

for any \( \Lambda : A \rightarrow B \), whether finite or infinite dimensional. For our example of an irreversible channel, we will use the qudit-to-qudit channel \( \Omega_3 \) whose Choi–Jamiołkowski state is \( \omega_3 \), namely

\[
\Omega_3 := \frac{3}{2} \Delta - \frac{1}{2} I_d
\]

where \( \Delta(\cdot) = \sum_{i=0}^{n-1} \langle \cdot | i \rangle \langle i | \cdot \rangle \) is the completely dephasing channel. Our lower bound (24) on the entanglement cost then gives \( E(\Omega_3) \geq E_n^t(\Omega_3) \geq E_n^t(\omega_3) \geq 1 \). To upper bound the quantum capacity of \( \Omega_3 \) several approaches are known. If the manipulation protocols we consider were restricted to adaptive quantum circuits, we could follow established techniques76,95 and use the relative entropy to obtain a bound very similar to the one we employed in the state case (equation (21)). However, to maintain full generality, we will instead employ a recent result53 showing that an upper bound on \( Q \) under the action of arbitrary NE protocols—not restricted to quantum circuits, and not required to have a definite causal order—is given by the max-relative entropy77 between a channel and all entanglement-breaking channels.

Using the completely dephasing channel \( \Delta \) as an ansatz, we get

\[
Q(\Omega_3) \leq \log_2 \frac{3}{2} < 1 \leq E(\Omega_3),
\]

establishing the irreversibility in the manipulation of quantum channels under the most general transformation protocols.

Data availability
No data sets were generated during this study.

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Acknowledgements
We are grateful to P. Faist, M. B. Plenio, M. M. Wilde and A. Winter for discussions as well as for helpful comments and suggestions on the manuscript. We also thank S.H. Lie for notifying us of a typo in a preliminary version of the paper. L.L. was supported by the Alexander von Humboldt Foundation. B.R. was supported by the Japan Society for the Promotion of Science (JSPS) KAKENHI grant no. 21F21015, a JSPS Postdoctoral Fellowship for Research in Japan, and a Presidential Postdoctoral Fellowship from Nanyang Technological University, Singapore.

Author contributions
Both authors contributed to all aspects of this manuscript. B.R. identified and formalised the problem studied here. L.L. conceived the notion of tempered negativity and used it to prove the irreversibility of entanglement theory under NE operations. B.R. and L.L. generalised the argument to approximately NE operations and to quantum channels. Both authors contributed equally to the writing of the paper.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41567-022-01873-9.

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Peer review information Nature Physics thanks the anonymous reviewer(s) for their contribution to the peer review of this work.

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