Generalized Laws of Thermodynamics in the Presence of Correlations

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Abstract

The laws of thermodynamics, despite their wide range of applicability, are known to break down when systems are correlated with their environments. Here, we generalize thermodynamics to physical scenarios which allow presence of correlations, including those where strong correlations are present. We exploit the connection between information and physics, and introduce a consistent redefinition of heat dissipation by systematically accounting for the information flow from system to bath in terms of the conditional entropy. As a consequence, the formula for the Helmholtz free energy is accordingly modified. Such a remedy not only fixes the apparent violations of Landauer’s erasure principle and the second law due to anomalous heat flows, but also leads to a generally valid reformulation of the laws of thermodynamics. In this information-theoretic approach, correlations between system and environment store work potential. Thus, in this view, the apparent anomalous heat flows are the refrigeration processes driven by such potentials.

Introduction

Thermodynamics is one of the most successful physical theories ever formulated. Though it was initially developed to deal with steam engines and, in particular, the problem of conversion of heat into mechanical work, it has survived even after the scientific revolutions of relativity and quantum mechanics. Inspired by resource theories, recently developed in quantum information, a renewed effort has been made to understand the foundations of thermodynamics in the quantum domain [1–11], including its connections to statistical mechanics [12–14] and information theory [15–25]. However, all these approaches assume that the system is initially uncorrelated from the bath. In fact, in the presence of correlations, the laws of thermodynamics can be violated. In particular, when there are inter-system correlations, phenomena such as anomalous heat flows from cold to hot baths [26], and memory erasure accompanied by work extraction instead of heat dissipation [24] become possible. These two examples indicate a violation of the second law in its Clausius formulation, and the Landauer’s principle of information erasure [15] respectively. Due to the interrelration between the different laws of thermodynamics, the zeroth law and the first law can also be violated (see Supplementary Note 4 for simple and explicit examples of these violations).

The theory of thermodynamics can be summarized in its three main laws. The zeroth law introduces the notion of thermal equilibrium as an equivalence relation of states, where temperature is the parameter that labels the different equivalence classes. In particular, the transitive property of the equivalence relation implies that if a body A is in equilibrium with a body B, and B is with a third body C, then A and C are also in equilibrium. The first law assures energy conservation. It states that in a thermodynamic process not all of energy changes are of the same nature and distinguishes between work, the type of energy that allows for “useful” operations as raising a weight, and its complement heat, any energy change which is not work. Finally, the second law establishes an arrow of time. It has several formulations and perhaps the most common one is the Clausius statement, which reads: No process is possible whose sole result is the transfer of heat from a cooler to a hotter body. Such a restriction not only introduces the fundamental limit on how and to what extent various forms of energy can be converted to accessible mechanical work, but also implies the existence of an additional state function, the entropy, which has to increase. There is also the third law of thermodynamics; we shall, however, leave it out of the discussion, as it is beyond immediate context of the physical scenarios considered here.

Although the laws of thermodynamics were developed phenomenologically, they have profound implications in information theory. The paradigmatic example is the Landauer erasure principle, which states: “Any logically irreversible manipulation of information, such as the erasure of a bit or the merging of two computation paths, must be accompanied by a corresponding entropy increase in non-information-bearing degrees of freedom of the information-processing apparatus or its environment” [17]. Therefore, an erasing operation is bound to be associated with a heat flow to the environment.

An important feature in the microscopic regime is that the quantum particles can exhibit non-trivial correlations, such as entanglement [27] and other quantum correlations [28]. Thermodynamics in the presence of correlations has been considered only in limited physical situations. It is assumed, in nearly all cases of thermodynamical processes, that system and bath are initially uncorrelated, although correlations may appear in the course of the process. In fact, it has been noted that in the presence of such correlations, Landauer’s erasure principle could be violated [15]. Even more strikingly, with strong quantum correlation between two thermal baths of different temperatures, heat could flow from the colder bath to...
the hotter one [26, 29, 30].

The impact of inter-system correlations resulting from a strong system-bath coupling and its role in thermodynamics has been studied for some specific solvable models [31–33], and for general classical systems [34, 35]. It has been noted that presence of correlations requires certain adjustments of work and heat to fulfil the second law and the Landauer principle. Also, from an information theoretic perspective, both extractable work from correlations and work cost to create correlations have been studied [25, 36–38]. However, in all these works, there is no explanation of how to deal with general correlated scenarios irrespective of where the correlations come from and in systems away from thermal equilibrium.

Here we show that the violations of the laws of thermodynamics (see Supplementary Note 4) indicate that correlations between two systems, irrespective of the corresponding marginals being thermal states or not, manifest out-of-equilibrium phenomena. In order to re-establish the laws of thermodynamics, one not only has to look at the local marginal systems, but also the correlations between them. In particular, we start by redefining the notions of heat and work, then establish a generalized Landauer’s principle and introduce the generalized Helmholtz free-energy. The resulting laws are general in the sense that they rely on the least set of assumptions to formulate thermodynamics: a system, a considerably large thermal bath at well defined temperature, and separable initial and final Hamiltonians. The first two assumptions are obvious. The third assumption is basically required for system’s and bath’s energies to be well defined (see Supplementary Note 2 for details).

Results

Definition of heat

To reformulate thermodynamics, we start with redefining heat by properly accounting for the information flow and thereby restoring Landauer’s erasure principle. In general, heat is defined as the flow of energy from the environment, normally considered as a thermal bath at certain temperature, to a system, in some way different from work. Work, on the other hand, is quantified as the flow of energy, say to a bath or to an external agent, that could be extractable (or accessible). Consider a thermal bath with Hamiltonian $H_B$ and at temperature $T$ represented by the Gibbs state $\rho_B = \tau_B = \frac{1}{Z_B} \exp(-\frac{H_B}{kT})$, where $k$ is the Boltzmann constant, and $Z_B = \text{Tr}\{\exp(-\frac{H_B}{kT})\}$ is the partition function. The degrees of freedom in $B$ are quantified as the flow of energy, say to a bath or to an external agent, that could be extractable (or accessible). Consider a thermal bath with Hamiltonian $H_B$ and at temperature $T$ represented by the Gibbs state $\rho_B = \tau_B = \frac{1}{Z_B} \exp(-\frac{H_B}{kT})$, where $k$ is the Boltzmann constant, and $Z_B = \text{Tr}\{\exp(-\frac{H_B}{kT})\}$ is the partition function. The degrees of freedom in $B$ are considered as a large thermal bath at well defined temperature, and separable initial and final Hamiltonians. The first two assumptions are obvious. The third assumption is basically required for system’s and bath’s energies to be well defined (see Supplementary Note 2 for details).

\[
\Delta Q = -kT \Delta S_B, \tag{1}
\]

where $\Delta S_B = S(\rho'_B) - S(\rho_B)$ is the change in the bath’s von Neumann entropy, $S(\rho_B) = -\text{Tr}[\rho_B \log_2 \rho_B]$. Note that $\rho'_B$ is not in general thermal. In fact, the work stored in the bath is $\Delta F_B$, where $F(\rho_B) = E(\rho_B) - kT S(\rho_B)$ is the Helmholtz free energy, with $E(\rho_B) = \text{Tr}(H_B \rho_B)$. Heat expressed in Eq. (1) is the correct quantification of heat (for further discussion see Supplementary Note 1), which can be justified in two ways. First, it has a clear information-theoretic interpretation, which accounts for the information flow to the bath. Second, it is the flow of energy to the bath other than work and, with the condition of entropy preservation, any other form of energy flow to the bath will be stored as extractable work, and thus will not converted into heat. The process-dependent character of heat as defined here can be seen from the fact that it cannot be written as a difference of state functions of the system. In the Supplementary Note 1, this issue is discussed and the sources of irreversibility, i.e. the reasons for not saturating the Clausius inequality, are re-examined.

The transformations considered in our framework are entropy preserving operations. More explicitly, given a system-bath setting initially in a state $\rho_{SB}$, in which the reduced state of the system $\rho_S$ is arbitrary while $\rho_B$ is thermal, we consider transformations $\rho_{SB}' = \Lambda(\rho_{SB})$ such that the von Neumann entropy is unchanged i.e. $S(\rho_{SB}') = S(\rho_{SB})$. The Hamiltonians of the system and the bath are the same before and after the transformation $\Lambda(\cdot)$. Note that we do not demand energy conservation, rather assuming that a suitable battery takes care of that. In fact, the work cost of such an operation $\Lambda(\cdot)$ is quantified by the global internal energy change $\Delta W = \Delta E_S + \Delta E_B$. Another comment to make is that we implicitly assume a bath of unbounded size; namely, it consists of the part $\rho_B$ of which we explicitly track the correlations with $S$, but also of arbitrarily many independent degrees of freedom. Also, we are implicitly considering always the asymptotic scenario of $n \to \infty$ copies of the state in question (“thermodynamic limit”). These operations are general and include any process and situation in standard thermodynamics involving a single bath. It is the result of abstracting the essential elements of thermodynamic processes: existence of a thermal bath and global entropy preservation operations.

In extending thermodynamics in correlated scenarios and linking thermodynamics with information, we consider the quantum conditional entropy as the natural quantity to represent information content in the system as well as in the correlations. For a joint system-bath state $\rho_{SB}$, the information content in the system $S$, given all the information available in the bath $B$ at temperature $T$, is quantified by the conditional entropy $S(S|B) = S(\rho_{SB}) - S(\rho_B)$. It vanishes when the joint system-environment state is perfectly classically correlated and can even become negative in the presence of entanglement.

Generalized second law of information

With quantum conditional entropy, the generalized second law of information can be stated as follows. For an entropy preserving operation $\rho_{SB}' = \Lambda^{SB}(\rho_{SB})$, with the reduced states before (after) the evolution denoted $\rho_S(\rho'_S)$ and $\rho_B(\rho'_B)$, re-
spectively, we have
\[ \Delta S_B = -\Delta S(S|B), \]
where \( \Delta S_B = S(\rho_B') - S(\rho_B) \) is the change in (von Neumann) entropy of the bath, and \( \Delta S(S|B) = S(S'|B') - S(S|B) \) is the change in conditional entropy of the system. Note that in the presence of initial correlations, the informational second law could be violated if one considers only system entropy (see Supplementary Note 3).

Let us point out that the conditional entropy of the system for a given bath is also used in [24] in the context of erasing. There, it is shown that the conditional entropy quantifies the amount of work necessary to erase quantum information. The formalism in [24] considers energy preserving but non-entropy preserving operations and that perfectly enables to quantify work. In contrast, in our formalism, as we attempt to quantify heat in connection with information flow, it is absolutely necessary to guarantee information conservation, thereby restrict ourselves to entropy preserving operations. This leads us to quantify heat in terms of conditional entropy. Both approaches are different and complement each other. In one, the conditional entropy quantifies work and on the other, it quantifies heat.

**Generalized Landauer’s principle**
The Landauer principle is required to be expressed in terms of conditional entropy of the system, rather than its local entropy. Therefore, the dissipated heat associated to information erasure of a system S connected to a bath B at temperature \( T \) by an entropy preserving operation \( \rho_{SB}' = \Lambda_{SB}(\rho_{SB}) \), is equal to
\[ \Delta Q = kT \Delta S(S|B). \]

Note that, in complete information erasure, the final conditional entropy vanishes, then \( \Delta Q = -kT S(S|B) \).

**Generalized Helmholtz free-energy**
We address extraction of work from a system S possibly correlated to a bath B at temperature \( T \). Without loss of generality, we assume that the system Hamiltonian \( H_S \) is unchanged in the process. Note that the extractable work has two contributions: one comes from system-bath correlations (cf. [25]) and the other from the local system alone, irrespective of its correlations with the bath. Here we consider these two contributions separately.

By extracting work from the correlation, we mean any process that returns the system and the bath in the original reduced states, \( \rho_S \) and \( \rho_B = \tau_B \). The maximum extractable work solely from the correlation, using entropy preserving operations, is given by
\[ W_C = kT I(S : B), \]
where \( I(S : B) = S_S + S_B - S_{SB} \) is the mutual information. This is illustrated in Fig. 1. The proof is given by the protocol described in Box 1.

Disregarding the correlations with a bath at temperature \( T \), the maximum extractable work from a state \( \rho_S \) is given by
\[ \Delta W_L = F(\rho_S) - F(\tau_S), \]
where \( \tau_S = \frac{1}{Z_S} \exp[-\frac{H_S}{kT}] \) is the corresponding thermal state of the system in equilibrium with the bath. Now, in addition to this “local work”, we have the work due to correlations, and so the total extractable work \( \Delta W_S \) is given by
\[ \Delta W_S = \Delta W_L + kT I(S : B)_{\rho_{SB}}, \]
where
\[ S(AS|B)_{\rho_{ASB}} = S(A'S|B')_{\rho_{ASB}}. \]
and thereby turning the additional state into a pure state \( \tau_A' = \langle \phi \rangle \langle \phi | \) of A, while leaving the marginal system and bath states unchanged. Clearly, the extractable work stored in the correlation is now transferred to the new additional system state \( \tau_A' \).

**Work extraction:** Work is extracted from \( \tau_A' \) at temperature \( T \), equal to \( W_C = I(S : B)_{\rho_{SB}} kT \).

Unlike the traditional free energy, the generalized free energy is not only a state function of the system S, but also of those degrees of freedom of the bath correlated with it. This is an unavoidable feature of the generalised formalism. Therefore, for a system-bath state \( \rho_{SB} \), maximum extractable work from the system can be given as
\[ \Delta W_S = \mathcal{F}(\rho_{SB}) - \mathcal{F}(\tau_S \otimes \tau_B), \]
where \( \mathcal{F}(\tau_S \otimes \tau_B) = F(\tau_S) \). Then, for a transformation, for which initial and final states are \( \rho_{SB} \) and \( \sigma_{SB} \), respectively, the maximum extractable work from the system, is \( \Delta W_S = -\Delta \mathcal{F} = \mathcal{F}(\rho_{SB}) - \mathcal{F}(\sigma_{SB}) \).

We observe that all this is of course consistent with what we know from situations with an uncorrelated bath. Indeed, we can simply make the conceptual step of calling SB “the system”, allowing for arbitrary correlations between S and B, with a suitable infinite bath B’ that is uncorrelated from SB. Then, the free energy as
we know it is \( F(\rho_{SB}) = E_S - kT S(S|B) + E_B - kT S(T_B) \), where the first term is the modified free energy in Eq. (6), and the second term is the free energy of the bath in its thermal state. As the latter cannot become smaller in any entropy-preserving operation, the maximum extractable work is \(-\Delta F^\ast\).

**Generalized laws of thermodynamics**

Now, equipped with the proper definition of heat (as in Eq. (3)) and work (based on generalized free energy in Eq. (6)) in the presence of correlations, we put forward the generalized laws of thermodynamics.

We start with generalized first law, which states: given an entropy preserving operation \( \rho_{SB} \to \rho_{SB}' \), the distribution of the change in the system’s internal energy into work and heat satisfies

\[
\Delta E_S = -(\Delta W_S + \Delta F_B) + (\Delta Q + \Delta F_B),
\]

where the heat dissipated to the bath is given by \( \Delta Q = -kT \Delta S(S|B) \), the maximum extractable work from the system is \( \Delta W_S = -(\Delta E_S - kT \Delta S(S)) \), and the work performed on the bath is \( \Delta F_B = \Delta E_B - kT \Delta S_B \geq 0 \).

The quantity \( \Delta W_S = -(\Delta E_S - kT \Delta S(S|B)) \) was shown to be the maximum extractable work, as it is equal to \(-\Delta F_S\). The maximum work \( \Delta W_S \) is extracted by thermodynamically reversible processes. Irreversible processes require that some work is performed on the bath \( \Delta F_B > 0 \) followed by an equilibration process, which happens due to spontaneous relaxation of the bath. Such amount of work is transformed into heat and hence cannot be accessed any more. Note that such an equilibration process is not entropy preserving [12] which is not allowed in our setup. The entropy production of such relaxation is precisely \( \Delta F_B/T \), and in that case heat flow from the bath is exactly equal to the decrease of its internal energy.

![Figure 2. Anomalous heat flows.](image)

In the presence of correlations, spontaneous heat flows from cold to hot baths are possible [26]. This is an apparent violation of second law, if one ignores the work potential stored in correlation. Otherwise, it is a refrigeration process.

In this new approach, the second law is also modified. The Clausius statement of the generalized second law states that no process is possible whose sole result is the transfer of heat from a cooler to a hotter body, where the work potential stored in the correlations, as defined in Eq. (4), does not increase. To prove it, consider a state transformation \( \rho_{AB}' \to \rho_{AB}^\ast \) where \( \Lambda_{AB}^\ast \) is an entropy preserving and and energy non-increasing operation. As the thermal state minimizes the free energy, the final reduced states \( \rho'_S \) and \( \rho'_B \) have increased their free energy, i.e., \( \Delta E_A - kT_A \Delta S_A \geq 0 \) and \( \Delta E_B - kT_B \Delta S_B \geq 0 \), where \( T_{A/B} \), \( \Delta E_{A/B} \) and \( \Delta S_{A/B} \) are the initial temperatures, changes in internal energy and entropy of the baths respectively. By adding the former inequalities and considering energy non-increasing, we get \( T_A \Delta S_A + T_B \Delta S_B \leq 0 \). Due to the conservation of total entropy, the change in mutual information is simply \( \Delta I(A : B) = \Delta S_A + \Delta S_B \), with \( I(A : B) = S_A + S_B - S_{AB} \). This allows us to conclude

\[
-\Delta Q_A(T_B - T_A) > kT_A T_B \Delta I(A : B),
\]

which implies Clausius statement of the generalized second law.

Note that if the initial state \( \rho_{AB} \) is correlated, then the change in mutual information could be negative, \( \Delta I(A : B) \leq 0 \), and \( -\Delta Q_A(T_B - T_A) \leq 0 \). Note that for \( T_A \leq T_B \) and \( \Delta I(A : B) \leq 0 \), there could be a heat flow from the cold to the hot bath \( \Delta Q_A \geq 0 \), i.e., an apparent anomalous heat flow. From our new perspective, we interpret the anomalous heat flow as a refrigeration driven by the work potential stored in correlations. In this case, it is interesting to determine its coefficient of performance \( \eta_{ CPC} \), from that Eq. (8) leads, with the work performed on the hot bath \( \Delta W_C(T_B) = -kT_B \Delta I(A : B) \), to

\[
\eta_{ CPC} \doteq \frac{\Delta Q_A}{\Delta W_C(T_B)} \leq \frac{T_A}{T_B - T_A}
\]

which is nothing else than the Carnot coefficient of performance (see Fig. 2). Note that we have taken the work value of the correlations \( W_C \) with respect to the hot bath \( T_B \). This is due to the fact that for this refrigeration process the hot bath is the one acting as a reservoir.

Equation (9) is a nice reconciliation with traditional thermodynamics. The Carnot coefficient of performance is a consequence of the fact that reversible processes are optimal, otherwise the perpetual mobile could be built by concatenating a “better” process and a reversed reversible one. Hence, it is natural that the refrigeration process driven by the work stored in the correlations preserves Carnot statement of second law.

![Figure 3. Violation of the zeroth law.](image)

In the presence of correlations, the notion of equilibrium is not an equivalence relation. Consider 3-party state \( \rho_{AB} \otimes \rho_{C} \) with all marginals thermal states. The thermal equilibria \( A \equiv B \) and \( B \equiv C \) implies that \( A \), \( B \) and \( C \) share the same temperature. But, in the presence of correlations between \( A \) and \( C \), that does not assure the equilibrium \( A \equiv C \). Therefore, the transitive property of the equivalence relation is violated. This is justified, on the right, as \( F(\rho_{AB} \otimes \rho_{C}) > F(\rho_A \otimes \rho_{C}) \). Thus, the generalized zeroth law has to overcome these limitations.

Now, we reconstruct the zeroth law which can be violated in the presence of correlations as shown in Fig. 3. To do this,
we redefine the notion of equilibrium beyond an equivalence relation when correlations between systems are present. Thus, the Universal zeroth law states that, a collection \( \{ \rho_X \} \) of states is said to be in mutual thermal equilibrium with each other if and only if no work can be extracted from any of their combinations under entropy preserving operations. This is the case if and only if all the parties \( X \) are uncorrelated and each of them is in a thermal state with the same temperature.

**Discussion**

Landauer exorcised Maxwell’s demon and saved the second law of thermodynamics by taking into account the work potential of information. In this work, we extend this idea to include also the information about the system that is stored in its correlations with the environment. With this approach, we easily resolve the apparent violations of thermodynamics in correlated scenarios, and generalize it by reformulating its zero-th, first, and second laws.

An important remark is that, our generalized thermodynamics is formulated in the asymptotic limit of many copies. A relevant question is how the laws of thermodynamics are expressed for a single system. In our forthcoming paper, we will address these questions by discussing consistent notions of one-shot heat, one-shot Landauer erasure, and of one-shot work extraction from correlations.

**References**

[1] Fernando G. S. L. Brandão, Michał Horodecki, Jonathan Oppenheim, Joseph M. Renes, and Robert W. Spekkens, “Resource theory of quantum states out of thermal equilibrium,” Physical Review Letters 111, 250404 (2013).

[2] Michał Horodecki and Jonathan Oppenheim, “Fundamental limitations for quantum and nanoscale thermodynamics,” Nature Communications 4, 2059 (2013).

[3] Johan Aberg, “Truly work-like work extraction via a single-shot analysis,” Nature Communications 4, 1925 (2013).

[4] Philippe Faist, Frééric Dupuis, Jonathan Oppenheim, and Renato Renner, “The minimal work cost of information processing,” Nature Communications 6, 7669 (2015).

[5] Paul Skrzypczyk, Anthony J. Short, and Sandu Popescu, “Work extraction and thermodynamics for individual quantum systems,” Nature Communications 5, 4185 (2014).

[6] Fernando G. S. L. Brandao, Michał Horodecki, Nelly Ng, Jonathan Oppenheim, and Stephanie Wehner, “The second laws of quantum thermodynamics,” Proceedings of the National Academy of Sciences 112, 3275–3279 (2015).

[7] Piotr Ćwikliński, Michał Studziński, Michał Horodecki, and Jonathan Oppenheim, “Limitations on the evolution of quantum coherences: Towards fully quantum second laws of thermodynamics,” Physical Review Letters 115, 210403 (2015).

[8] Matteo Lostaglio, Kamil Korzekwa, David Jennings, and Terry Rudolph, “Quantum coherence, time-translation symmetry, and thermodynamics,” Physical Review X 5, 021001 (2015).

[9] Matteo Lostaglio, David Jennings, and Terry Rudolph, “Description of quantum coherence in thermodynamic processes requires constraints beyond free energy,” Nature Communications 6, 6383 (2015).

[10] Oscar C. O. Dahlsten, Renato Renner, Elisabeth Rieper, and Vlatko Vedral, “Inadequacy of von neumann entropy for characterizing extractable work,” New Journal of Physics 13, 053015 (2011).

[11] D Eglolf, O C O Dahlsten, R Renner, and V Vedral, “A measure of majorization emerging from single-shot statistical mechanics,” New Journal of Physics 17, 073001 (2015).

[12] Christian Gogolin and Jens Eisert, “Equilibration, thermalisation, and the emergence of statistical mechanics in closed quantum systems,” Reports on Progress in Physics 79, 056001 (2016).

[13] Anthony J Short, “Equilibration of quantum systems and subsystems,” New Journal of Physics 13, 053009 (2011).

[14] Sandu Popescu, Anthony J. Short, and Andreas Winter, “Entanglement and the foundations of statistical mechanics,” Nature Physics 2, 745–758 (2006).

[15] David Reeb and Michael M Wolf, “An improved landauer principle with finite-size corrections,” New Journal of Physics 16, 103011 (2014).

[16] Koji Maruyama, Franco Nori, and Vlatko Vedral, “Colloquium: The physics of maxwell’s demon and information,” Reviews in Modern Physics 81, 1–23 (2009).

[17] Charles H. Bennett, “The thermodynamics of computation—a review,” International Journal of Theoretical Physics 21, 905–940 (1982).

[18] Juan M. R. Parrondo, Jordan M. Horowitz, and Takahiro Sagawa, “Thermodynamics of information,” Nature Physics 11, 131–139 (2015).

[19] H. S. Leff and A. F. Rex, Maxwell’s Demon: Entropy, Information, Computing (London: Taylor and Francis, 1990).

[20] H. S. Leff and A. F. Rex, Maxwell’s Demon 2: Entropy, Classical and Quantum Information, Computing (London: Taylor and Francis, 2002).

[21] R. Landauer, “Irreversibility and heat generation in the computing process,” IBM Journal of Research and Development 5, 183–191 (1961).

[22] L. Szilard, “Über die Entropieverminderung in einem thermodynamischen System bei Eingriffen intelligenter Wesen,” Zeitschrift für Physik 53, 840–856 (1929).

[23] John Goold, Marcus Huber, Arnau Riera, Lidia del Rio, and Paul Skrzypczyk, “The role of quantum information in thermodynamics—a topical review,” Journal of Physics A: Mathematical and Theoretical 49, 143001 (2016).

[24] Lidia del Rio, Johan Aberg, Renato Renner, Oscar C. O. Dahlsten, and Vlatko Vedral, “The thermodynamic meaning of negative entropy,” Nature 474, 61–63 (2011).

[25] Martí Perarnau-Llobet, Karen V. Hovhannisyan, Marcus Huber, Paul Skrzypczyk, Nicolas Brunner, and Antonio Acín, “Extractable work from correlations,” Phys. Rev. X 5, 041011 (2015).

[26] David Jennings and Terry Rudolph, “Entanglement and the thermodynamic arrow of time,” Physical Review E 81, 061130 (2010).

[27] Ryszard Horodecki, Paweł Horodecki, Michał Horodecki, and Karol Horodecki, “Quantum entanglement,” Reviews of Modern Physics 81, 865–942 (2009).

[28] Kavan Modi, Aharon Brodutch, Hugo Cable, Tomasz Paterek, and Vlatko Vedral, “The classical-quantum boundary for correlations: Discord and related measures,” Reviews of Modern Physics 84, 1655–1707 (2012).

[29] Sania Jevtic, David Jennings, and Terry Rudolph, “Maximally and minimally correlated states attainable within a closed
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Author contributions

M. N. B. and A. R. have equally contributed to this work.
SUPPLEMENTARY INFORMATION

SUPPLEMENTARY NOTE 1: DEFINITIONS OF HEAT

In the main text, the heat dissipated in a process involving a system and a bath $B$ has been defined as $\Delta Q = kT\Delta S_B$, such as the common description: “flow of energy to a bath some way other than through work” suggests. Note, however, that this is not the most extended definition of heat that one finds in many works, e.g., [? ? ], where heat is defined as the change in the internal energy of the bath, i. e.

$$\Delta \tilde{Q} := -\Delta E_B,$$

and no different types of energy are distinguished in this increase of energy. In this section, we compare these two definitions and argue why the approach taken here, though less extended, seems the most appropriate.

The ambiguity in defining heat comes from the different ways in which the change in the internal energy of the system $E_S$ can be decomposed. More explicitly, let us consider a unitary process $U_{SB}$ acting on a system-bath state $\rho_{SB}$ with $\rho_B = \text{Tr}_S \rho_{SB} = e^{-H_B/kT}$ and global Hamiltonian $H = H_S \otimes I + I \otimes H_B$. The change in the total internal energy $\Delta E_{SB}$ is the sum of system and bath internal energies $\Delta E_{SB} = \Delta E_S + \Delta E_B$, or equivalently

$$\Delta E_S = \Delta E_{SB} - \Delta E_B.$$  

Many text-books identify in this decomposition $\Delta W := -\Delta E_{SB}$ as work and $\Delta \tilde{Q} = -\Delta E_B$ as heat. Nevertheless, note that it also assigns to heat increases of the internal energy that are not irreversibly lost and can be recovered when having a bath at our disposal.

To highlight the incompleteness of the above definition, let us consider a reversible process $U_{SB} = I \otimes U_B$ that acts trivially on the system. Then, even though the state of the system is untouched in such a process, the amount of heat dissipated is $\Delta \tilde{Q} = -\Delta E_B = \text{Tr}[H_B(\rho_B - U_B\rho_B U_B^\dagger)]$.

In order to avoid this kind of paradoxes and in the spirit of the definition given above, we subtract from $\Delta E_B$ its component of energy that can still be extracted (accessed). Then for a transformation $\rho_B \rightarrow \rho_B'$, the heat transferred is given as

$$\Delta Q = - (\Delta E_B - \Delta F_B),$$

where $\Delta F_B = F(\rho_B') - F(\rho_B)$ is the work stored on the bath and can be extracted. Here, $F(\rho_X) = E_X - kT S(\rho_X)$ is the Helmholtz free energy, $E_X$ is the internal energy and $\Delta S_B = S(\rho_B') - S(\rho_B)$ is the change in the bath’s von Neumann entropy, $S(\rho_B) = -\text{Tr}[\rho_B \log_2 \rho_B]$. Throughout this work, we consider $\log_2$ as the unit of entropy.

Let us remark that in practical situations, in the limit of large baths, both definitions coincide. To see it, take Supplementary Eq. (12) and note that both definitions only differ in the free energy difference term, which together with the fact that the free energy is minimized by the thermal state, implies that the difference is very small when the bath is slightly perturbed. However, when studying thermodynamics at the quantum regime with small machines approaching the nanoscale such conceptual differences are crucial to extend, for instance, the domain of standard thermodynamics to situations where the correlations become relevant.

Note finally that both definitions express a path dependent quantity of the system like heat in terms of a difference of state functions of the bath. The path dependence character comes from the fact that there are several processes that leave the system in the same state but the bath in a different one. This connects with Clausius inequality, which is usually stated as

$$\oint \frac{dQ}{T} \leq 0$$

where the integral is taken over a cyclic path and the equality is only saturated by quasiestatic processes. In our framework and for the case of defining heat by means of information (entropy), the Clausius inequality is a consequence of the positivity of the mutual information. That is, by assuming global entropy preservation we have

$$\Delta S_S = -\Delta S_B + \Delta I(S : B) = \frac{\Delta Q}{T} + \Delta I(S : B)$$

where $I(S : B) = S_S + S_B - S_{SB}$ is the mutual information. For an initially uncorrelated system-bath, the mutual information can only increase $\Delta I(S : B) \geq 0$, and

$$\frac{\Delta Q}{T} \leq \Delta S_S.$$  

For the definition of heat as an increase of the internal energy, we have

$$\Delta \tilde{Q} = \Delta Q - \Delta F_B \leq \Delta Q,$$

where we have used Supplementary Eq. (12) and the positivity of the free energy change. In sum, for the case of initially uncorrelated states, we recover the Clausius inequality,

$$\Delta \tilde{Q} \leq \Delta Q \leq T \Delta S.$$  

The deficit for the first inequality to be saturated is $\Delta F_B$, that is, the energy that can still extracted from the bath. If one has a limited access to the bath, an apparent relaxation process will follow and the bath will thermalize keeping its energy constant. This will imply an entropy increase of the bath $\Delta F_B/T$ which will make $\Delta \tilde{Q}$ and $\Delta Q$ coincide.

The deficit to saturate the second inequality in Supplementary Eq. (17) is $\Delta I(S : B)$, that is, the amount of enabled correlations during the process. One of the main ideas of this work is to show that these correlations capture a free energy that can be extracted.
SUPPLEMENTARY NOTE 2: SET OF OPERATIONS

The set of operations that we consider in this manuscript is the so called entropy preserving operations. Given a system initially in a state \( \rho \), the set of entropy preserving operations are all the operations that change arbitrarily the state but keep its entropy constant

\[
\rho \rightarrow \sigma : S(\rho) = S(\sigma),
\]

where \( S(\rho) := -\text{Tr}(\rho \log \rho) \) is the Von Neumann entropy. It is important to note that an operation that acting on \( \rho \) produces a state with the same entropy does not mean that will also preserve entropy when acting on other states. In other words, such entropy preserving operations are in general not linear, since they have to be constraint to some input state. In fact, in [39], it is shown that a quantum channel \( \Lambda(\cdot) \) that preserves entropy and respects linearity, i.e. \( \Lambda(\rho \rho_1) + (1-p)\rho_2 = p\Lambda(\rho_1) + (1-p)\Lambda(\rho_2) \), has to be necessarily unitary.

One could think then that the extension of the unitaries to a set of entropy preserving operations is rather unphysical since they are not linear. However, they can be microscopically described by global unitaries in the limit of many copies [40]. That is, given any two states \( \rho \) and \( \sigma \) with equal entropies \( S(\rho) = S(\sigma) \), then there exists a unitary \( U \) and an additional system \( O(\sqrt{n} \log n) \) ancillary qubits such that

\[
\lim_{n \to \infty} \| \text{Tr}_{\text{anc}} (U \rho^\text{anc} \otimes \eta U^\dagger) - \sigma^\text{anc} \| = 0,
\]

where \( \| \cdot \| \) is the one-norm and the partial trace is performed on the ancillary qubits. The reverse statement is also true, i.e. if two states can be related as in Supplementary Eq. (19) then they have equal entropies. This is proven in Theorem 4 of Ref. [40].

Sometimes it can be interesting to restrict entropy preserving operations to also be energy preserving. The set of energy and entropy preserving channels can also be described as a global energy preserving unitary in the many copy limit. More explicitly, in Theorem 1 of Ref. [40], it is proven that two states \( \rho \) and \( \sigma \) having equal entropies and energies \( S(\rho) = S(\sigma) \) and \( E(\rho) = E(\sigma) \) is equivalent to the existence of some \( U \) and an additional system \( O(\sqrt{n} \log n) \) ancillary qubits with Hamiltonian \( \| H_\chi \| \leq O(n^{2/3}) \) in some state \( \eta \) for which Supplementary Eq. (19) is fulfilled. Note that the amount of energy and entropy of the ancillary system per copy vanishes in the large \( n \) limit.

In sum, considering the set of entropy preserving operations means implicitly taking the limit of many copies and global unitaries. In addition, as that the set of entropy preserving operations contains the set of unitaries, any constraint that appears as a consequence of entropy preservation will be also respected by individual quantum systems.

The Hamiltonians of the system and the bath are the same before and after the transformation \( \Lambda(\cdot) \). This can be done without loss of generality since, when this is not the case and the final Hamiltonian is different from the initial one, the two situations are related by a simple quench (instantaneous change of the Hamiltonian). More explicitly, let us consider a process (a) with equal initial and final Hamiltonian, and an identical process (b) with different,

(a) \( (H, \rho_i) \rightarrow (H, \rho_f) \)
(b) \( (H, \rho_i) \rightarrow (H', \rho_f) \)

where \( \rho_{if} \) is the initial/final state, \( H \) the initial Hamiltonian and \( H' \) the final Hamiltonian of the process with different Hamiltonians. Then, it is trivial to relate the work and heat involved in both processes

\[
W' = W + \text{Tr} ((H - H')\rho_i)
\]
\[
Q' = Q,
\]

where \( W' \) and \( Q' \) are the work and heat associated to the process (b) and we have only used that the process (a) is the composition of the process (a) followed by a quantum quench.

Let us finally point out that initially and finally the Hamiltonians of system and bath are not interacting, or in other words, the system is decoupled from the bath

\[
H = H_S \otimes 1 + 1 \otimes H_B,
\]

with \( H_{S/B} \) the Hamiltonian of the system/bath. This is a necessary condition to be able to consider system and bath as independent systems each with a well defined notion of energy. Otherwise, assigning an energy to the system and to the bath would not be possible beyond the weak coupling limit. Note that the system and the bath interact (arbitrarily strongly) during the process, in which for instance a non-product unitary could be performed.

SUPPLEMENTARY NOTE 3: THE LANDAUER PRINCIPLE

The information theory and statistical mechanics have longstanding and intricate relation. In particular, to exorcise Maxwell's demon in the context of statistical thermodynamics, Landauer first indicated that information is physical and any manipulation of that has thermodynamic cost. As put forward by Bennett [? ], the Landauer information erasure principle (LEP) implies that "any logically irreversible manipulation of information, such as the erasure of a bit or the merging of two computation paths, must be accompanied by a corresponding entropy increase in non-information-bearing degrees of freedom of the information-processing apparatus or its environment."

Following the definition of heat, it indicates that, in such processes, entropy increase in non-information-bearing degrees of freedom of a bath is essentially associated with flow of heat to the bath. The major contribution of this work is to exclusively quantify heat in terms of flow of information, instead of counting it with the flow of non-extractable energy.
the work. To establish this we start with the case of information erasure of a memory. Consider a physical process where an event, denoted with $i$, happens with the probability $p_i$. Then storing (classical) information memorizing the process means constructing a $d$-dimensional system (a memory-dit) in a state $\rho_S = \sum_i p_i |i\rangle\langle i|$, where $|i\rangle$ are the orthonormal basis correspond to the event $i$. In other words, memorizing the physical process is nothing but constructing a memory state $\rho_S = \sum_i p_i |i\rangle\langle i|$ from a memoryless state $|\phi\rangle\langle \phi|$ where $i$ could assume any values $1 \leq i \leq d$. On the contrary, process of erasing requires the transformation of a memory state $\rho_S = \sum_i p_i |i\rangle\langle i|$ to a memoryless state $|\phi\rangle\langle \phi|$ for any $i$. Landauer’s erasure principle (LEP) implies that erasing information, a process involving a global evolution of the memory-dit system and its environment, is inevitably associated with an increase in entropy in the environment.

In establishing the connection between information erasing and heat dissipation, we make two assumptions to start with. First, the memory-system ($S$) and bath ($B$) are both described by the Hilbert space $\mathcal{H}_S \otimes \mathcal{H}_B$. Secondly, the erasing process involves entropy preserving operation $\Lambda_{SB}$, i.e., $\rho'_{SB} = \Lambda_{SB} (\rho_{SB})$. The latter assumption is most natural and important, as it preserves information content in the joint memory-environment system. Without loss of generality, one can further assume that the system and bath Hamiltonians remain unchanged throughout the erasing process, to ease the derivations.

Now we consider the simplest information erasing scenario, which leads to LEP in its traditional form. In this scenario, a system $\rho_S$ is brought in contact with a bath $\rho_B$ and the system is transformed to a information erased state, say $|0\rangle\langle 0|_S$, by performing a global entropy-preserving operation $\Lambda_{SB}$, i.e.,

$$\rho_S \otimes \rho_B \xrightarrow{\Lambda_{SB}} |0\rangle\langle 0|_S \otimes \rho'_B,$$

(25)

where initial and final joint system-bath states are uncorrelated. The joint operation guarantees that the decrease in system’s entropy is exactly equal to the increase in bath entropy and heat dissipated to the bath is $\Delta Q = -kT \Delta S_B$. It clearly indicates that an erasure process is expected to heat up the bath. This in turn also says that $\Delta Q = kT \Delta S_S$, where $\Delta S_S = S(\rho'_S) - S(\rho_S)$. In the case where the $d$-dimensional system memorizes maximum information, or in other words it is maximally mixed and contains $\log_2 d$ bits of information, the process dissipates an amount $kT \log_2 d$ of heat to completely erase the information. In other words, to erase one bit of information system requires the dissipation of $kT$ of heat and we denote it as one heat-bit or $\ell$-bit (in honour of Landauer).

In the case where the final state may be correlated, the dissipated heat in general is lower bounded by the entropy reduction in the system, i.e.,

$$|\Delta Q| \geq kT |\Delta S_S|.$$  

(26)

This is what is generally known as the Landauer’s erasure principle (LEP), in terms of heat.

The above formulation of LEP crucially relies on the fact that any change in system entropy leads to a larger change in the bath entropy, which is also traditionally known as the second law for the change in the information, i.e.,

$$\Delta S_B \geq -\Delta S_S.$$  

(27)

However, it is limited by the assumptions made above and can be violated with initial correlations. Consider the examples in section of the Supplementary Information. In both the examples, $\Delta S_B \neq -\Delta S_S$. Therefore, one has to replace it with universal informational second law.

**SUPPLEMENTARY NOTE 4: VIOLATIONS OF LAWS OF THERMODYNAMICS**

In order to highlight how the laws of thermodynamics break down in the presence of correlations, let us discuss the following two examples. In the first, the system $S$ is purely classically correlated with the bath $B$ at temperature $T$, while in the other they are jointly in a pure state and share quantum entanglement. In both the examples the Hamiltonians of the system and bath ($H_S$ and $H_B$) remain unchanged throughout the processes.

**Example 1 – Classical correlations.**

$$\rho_{SB} = \sum_i p_i |i\rangle\langle i|_S \otimes |i\rangle\langle i|_B \xrightarrow{U_{SB}} \rho'_{SB} = |\phi\rangle\langle \phi|_S \otimes \sum_i p_i |i\rangle\langle i|_B.$$  

**Example 2 – Entanglement.**

$$|\Psi\rangle_{SB} = \sum_i \sqrt{p_i} |i\rangle_0 |i\rangle_B \xrightarrow{U_{SB}^c} |\Psi'\rangle_{SB} = |\phi\rangle_0 \otimes |\phi\rangle_B,$$

where in both examples $|\phi\rangle_X = \sum_i \sqrt{p_i} |i\rangle_X$ with $X \in \{S, B\}$ and $1 > p_i \geq 0$ for all $i$. Note that the unitaries, $U_{SB}^c$ and $U_{SB}^e$, leave the local energies of system and bath unchanged, and $U_{SB}^e$ does not change the bath state.

**Violations of first law**

In Example 1, the Helmholtz free energy of the system increases $F(|\phi\rangle_S) > F(\rho_S)$ and therefore a work $-\Delta W_S = \Delta F_S > 0$ is performed on the system. To assure the energy conservation of the system, an equal amount of heat is required to be transferred to the bath. Surprisingly, however, no heat is transferred to the bath as it remains unchanged. Thus $\Delta E_S = -\Delta W_S + \Delta Q$, i.e. the energy conservation is violated and so the first law.

A further violation can also be seen in Example 2 involving system-bath quantum entanglement. In this case, a non-zero work $-\Delta W_S = \Delta F_S > 0$ has been performed on the system, and a heat flow to the bath is expected. In contrast, there is a negative heat flow to the bath! Therefore, it violates the first law, i.e. $\Delta E_S \neq -\Delta W_S + \Delta Q$. 


Violations of second law and anomalous heat flows

We now show how correlation could result in a violation of the Kelvin-Planck statement of the second law, which states: No process is possible whose sole result is the absorption of heat from a reservoir and the conversion of this heat into work. In Example 1, no change in the local bath state indicates that there is no transfer of heat. However, the change in the Helmholtz free energy of the local system is $-\Delta W = \Delta F > 0$. Thus, a non-zero amount of work is performed on the system without even absorbing heat from the bath ($\Delta Q = 0$).

The situation becomes more striking in Example 2, with initial system-bath entanglement. In this case, $-\Delta W = \Delta F > 0$ amount of work is performed on the system. However, not only is there no heat flow from the bath to the system, but there is a negative heat flow to the bath! Thus, the second law is violated.

We next see how the presence of correlations can lead to anomalous heat flows and thereby a violation of the Clausius statement based second law. Such violations were known for the other definition of heat $\Delta Q = -\Delta E_B$ (see [? ] and references therein). Here we show that such violations are also there with new heat definition $\Delta Q = -kT\Delta S_B$. Let $\rho_{AB} \in \mathcal{H}_A \otimes \mathcal{H}_B$ be an initial bipartite finite dimensional state whose marginals $\rho_A = \text{Tr}_B \rho_{AB}$ are thermal states at different temperatures $T_A$ and $T_B$ and with Hamiltonians $H_A$ and $H_B$. In absence of initial correlations between the baths A and B, any energy preserving unitary will respect Clausius’ statement of the second law. However, if initial correlations are present, this will not be necessarily the case.

Consider a state transformation $\rho'_{SB} = U_{AB}\rho_{AB}U_{AB}^\dagger$ where $U_{AB}$ is an energy preserving unitary acting on $\rho_{AB}$. As the thermal state minimizes the free energy, the final reduced states $\rho'_S$ and $\rho'_B$ have increased their free energy,

$$\Delta E_A - kT_A \Delta S_A \geq 0 \quad (28)$$
$$\Delta E_B - kT_B \Delta S_B \geq 0 \quad (29)$$

where $T_{A/B}$ is the initial temperature of the baths, and $\Delta E_{A/B}$ and $\Delta S_{A/B}$ are the change in internal energy and entropy respectively.

By adding Supplementary Eqs. (28) and (29), and considering energy conservation, we get

$$T_A \Delta S_A + T_B \Delta S_B \leq 0 . \quad (30)$$

Due to the conservation of total entropy, the change in mutual information is simply $\Delta I(A : B) = \Delta S_A + \Delta S_B$, with $I(A : B) = S_A + S_B - S_{AB}$. This allows us to rewrite Supplementary Eq. (30) in terms of only the entropy change in A as

$$T_A - T_B) \Delta S_A \leq -T_B \Delta I(A : B). \quad (31)$$

If the initial state $\rho_{AB} = \rho_A \otimes \rho_B$ is uncorrelated, then the change in mutual information is necessarily positive $\Delta I(A : B) \geq 0$, and

$$k(T_A - T_B)\Delta S_A = -\Delta Q_A \frac{T_A - T_B}{T_A} \leq 0 . \quad (32)$$

To see that this equation is precisely the Clausius statement, consider without loss of generality that A is the hot bath and $T_A - T_B > 0$. Then, Supplementary inequality (32) implies an entropy reduction of the hot bath $\Delta S_A \leq 0$ i.e. a heat flow from the hot bath to the cold one.

However, if the the system is initially correlated, the process can reduce the mutual information, $\Delta I(A : B) < 0$, and Supplementary Eq. (31) allows a heat flow from the cold bath to the hot one.

Violations of zeroth law

The zeroth law establishes the notion of thermal equilibrium as an equivalence relation, in which temperature labels the different equivalent classes. To see that the presence of correlations also invalidates the zeroth law, we show that the transitive property of the equivalence relation is not fulfilled. Consider a bipartite system AC in an initial correlated state $\rho_{AC}$, like in Examples 1 and 2, and a third party B which is in a thermal state at the same temperature of the marginals $\rho_A$ and $\rho_C$. Then, while the subsystems AB and BC are mutually in equilibrium, the subsystems AC are not, clearly violating transitivity. There are several ways to realize that the parties AC are not in equilibrium. One way is to see that any energy preserving unitary, except for the identity, decreases the amount of correlations between the parties, $\Delta I(A : C) < 0$, which implies that the initial state is not stable. This can be shown from Supplementary Eq. (30) for the particular case of equal temperatures and the definition of mutual information.

Another way is to see that the Boltzmann free energy follows $F(\rho_{AC}) > F(\rho_A \otimes \rho_C)$.

Violations of Landauer’s erasure principle

Another thermodynamic principle that breaks down when correlations are present is Landauer’s erasure principle. Landauer postulated that in order to erase one bit of information in the presence of a bath at temperature $T$, an amount of heat needed to be dissipated is $kT \log 2$. As noted in [? ], when the system is classically correlated, there exists erasing process which does not increase entropy of the bath (see Example 1). The situation becomes more striking when the system shares quantum entanglement with the bath. This is the case of Example 2 with initial entanglement, where instead of increasing, an erasing process reduces the entropy of the bath and the bath is cooled down.