

The second laws of quantum thermodynamics

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The second law of thermodynamics places constraints on state transformations. It applies to systems composed of many particles, however, we are seeing that one can formulate laws of thermodynamics when only a small number of particles are interacting with a heat bath. Is there a second law of thermodynamics in this regime? Here, we find that for processes which are approximately cyclic, the second law for microscopic systems takes on a different form compared to the macroscopic scale, imposing not just one constraint on state transformations, but an entire family of constraints. We find a family of free energies which generalize the traditional one, and show that they can never increase. The ordinary second law relates to one of these, with the remainder imposing additional constraints on thermodynamic transitions. We find three regimes which determine which family of second laws govern state transitions, depending on how cyclic the process is. In one regime one can cause an apparent violation of the usual second law, through a process of embezzling work from a large system which remains arbitrarily close to its original state. These second laws are relevant for small systems, and also apply to individual macroscopic systems interacting via long-range interactions. By making precise the definition of thermal operations, the laws of thermodynamics are unified in this framework, with the first law defining the class of operations, the zeroth law emerging as an equivalence relation between thermal states, and the remaining laws being monotonicity of our generalized free energies.

quantum thermodynamics | quantum information theory | statistical physics | resource theory | free energy

The original formulation of the second law, due to Clausius (1), states that “Heat can never pass from a colder to a warmer body without some other change, connected therewith, occurring at the same time.” In attempting to apply Clausius’s statement of the second law to the microscopic or quantum scale, we immediately run into a problem because it talks about cyclic processes in which there is no other change occurring at the same time, and at this scale it is impossible to design a process in which there is no change, however slight, in our devices and heat engines. Interpreted strictly, the Clausius statement of the second law applies to situations which never occur in nature. The same holds true for other versions of the second law, such as the Kelvin–Planck statement, where one also talks about cyclic processes, in which all other objects beside the system of interest are returned back to their original state. Here, we derive a quantum version of the Clausius statement, by looking at processes where a microscopic or quantum system undergoes a transition from one state to another, whereas the environment and working body or heat engine is returned back to their original state. Whereas macroscopically only a single second law restricts transitions, we find that there is an entire family of more fundamental restrictions at the quantum level. At the macroscopic scale, and for systems with short-range correlations, this entire family of second laws becomes equal to the ordinary second law, but outside of this regime these other second laws impose additional restrictions on thermodynamical transitions. What is more, one needs to be more precise about what one means by a cyclic process. At the macroscopic scale, the fact that a process is only approximately cyclic has generally been assumed to be enough to guarantee the second law.

Here, we show that this is not the case in the microscopic regime, and we therefore need to talk about “how cyclic” a process is when stating the second law. We also derive in this work, a zeroth law of thermodynamics, which is stronger than the ordinary zeroth law.

For thermodynamics at the macroscopic scale, a system in state ρ can be transformed into state ρ' provided that the free energy goes down, where the free energy for a state ρ is

$$F(\rho) = \langle E(\rho) \rangle - kTS(\rho), \quad [1]$$

with T the temperature of the ambient heat bath that surrounds the system, k the Boltzmann constant, $S(\rho)$ the entropy of the system, and $\langle E \rangle$ its average energy. This is a version of the second law, where we also use the fact that the total energy of the system and heat bath must be conserved. This criterion governing state transitions is valid if the system is composed of many particles, and there are no long-range correlations. In the case of microscopic, quantum, or highly correlated systems, a criterion for state transitions of a total system was proven in ref. 2 and named thermo-majorization. This criterion has been conjectured (3) and serves as a second law in some cases (see also the reformulation of ref. 4). However, here we will see that if elevated to such high status without sufficient care, it can be violated. Namely, we will give examples where $\rho \rightarrow \rho'$ would violate the thermo-majorization criterion, but nonetheless, the transition is possible via a cyclic process in which a working body σ —an ancilla or catalyst—is returned back into its original state. The criterion of ref. 2 is thus only relevant when additional systems are not used to aid in the transition.

This phenomenon is related to entanglement catalysis (5), where it can be shown that some forbidden transitions are

Significance

In ordinary thermodynamics, transitions are governed by a single quantity—the free energy. Its monotonicity is a formulation of the second law. Here, we find that the second law for microscopic or highly correlated systems takes on a very different form than it does at the macroscopic scale, imposing not just one constraint on state transformations, but many. We find a family of quantum free energies which generalize the standard free energy, and can never increase. The ordinary second law corresponds to the nonincreasing of one of these free energies, with the remainder imposing additional constraints on thermodynamic transitions. In the thermodynamic limit, these additional second laws become equivalent to the standard one. We also prove a strengthened version of the zeroth law of thermodynamics, allowing a definition of temperature.

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possible, if we can use an additional system σ as a catalyst, i.e., we may have $\rho \rightarrow \rho'$ and yet $\rho \otimes \sigma \rightarrow \rho' \otimes \sigma$. In the case of thermodynamics, the catalyst σ may be thought of as a working body or heat engine which undergoes a cyclic process and is returned back into its original state. In deciding whether one can transform ρ into ρ' , one therefore needs to ask whether there exists a working body or other ancillas σ for which $\rho \otimes \sigma \rightarrow \rho' \otimes \sigma$ (Fig. 1). Thus, thermo-majorization (Fig. 2) should only be applied to total resources including all possible catalysts and working bodies and not the system of interest itself. In the case of entanglement theory, and when the catalyst is returned in exactly the same state, the criteria for when one pure state may be transformed into another have been found (6, 7) and they are called trumping conditions. We will generalize and adapt the trumping conditions to enable their application to the case of thermodynamics.

Family of Second Laws

Here, we consider all possible cyclic thermodynamical processes, and show that transition laws are affected by using ancillary systems which are returned back to their initial state. Rather than a single free energy that determines which transitions are possible, we find necessary and sufficient conditions for thermodynamic transitions which form not just one but a family of second laws. We define the free energies

$$F_\alpha(\rho, \rho_\beta) := kT D_\alpha(\rho || \rho_\beta) - kT \log Z, \quad [2]$$

with the Rényi divergences $D_\alpha(\rho || \rho_\beta)$ defined as

$$D_\alpha(\rho || \rho_\beta) = \frac{\text{sgn}(\alpha)}{\alpha - 1} \log \sum_i p_i^\alpha q_i^{1-\alpha}, \quad [3]$$

where p_i are the eigenvalues of ρ and q_i the eigenvalues of the thermal state of the system $\rho_\beta = e^{-\beta H_S} / Z$ with Hamiltonian H_S , partition function $Z = \sum_{i,g} e^{-\beta E_i}$, and $\beta = 1/T$.

We can then state quantum second laws, and ones that hold for states block diagonal in the energy basis. In the latter case, we find the following set of second laws:

In the presence of a heat bath of single fixed temperature, the free energies $F_\alpha(\rho, \rho_\beta)$ do not increase for $\alpha \geq 0$. That is, $\forall \alpha \geq 0$, $F_\alpha(\rho, \rho_\beta) \geq F_\alpha(\rho', \rho_\beta)$, where ρ and ρ' are the initial and final state, respectively. Moreover, if $F_\alpha(\rho, \rho_\beta) \geq F_\alpha(\rho', \rho_\beta)$ holds $\forall \alpha \geq 0$,

then there exists a catalytic thermal operation that transforms ρ to ρ' .

We say that the $F_\alpha(\rho, \rho_\beta)$ are monotones—the system always gets closer to the thermal state, thus the function always decreases. By including an auxiliary system as described in ref. 2, the above statement of the second law is equivalent to the case where one changes the Hamiltonian of the system, in which case one could write $F_\alpha(\rho, \rho_\beta) \geq F_\alpha(\rho', \rho'_\beta)$, where the initial Hamiltonian H_S has been changed via external control to the final Hamiltonian H'_S , with ρ_β and ρ'_β being the respective thermal states. This is described in *SI Appendix, section I*. Note that in fact $F_\alpha(\rho, \rho_\beta)$ is a monotone for all $\alpha \in (-\infty, \infty)$ but because we are allowed to borrow a pure state and return it in a state arbitrarily close to its initial state, only $\alpha \geq 0$ is relevant, as can be seen by noting that if any of the probabilities p_i in Eq. 3 are zero, then for $\alpha < 0$, $F_\alpha(\rho, \rho_\beta)$ diverges and will thus always be monotonic.

This set of limitations is less stringent than thermo-majorization. Not only do these second laws provide limitations, but they are also sufficient—whenever the free energies of some state ρ are all greater than for another state ρ' , one can transform ρ into ρ' . We prove this in *SI Appendix*. Note that the monotonicity of [2] establishes a continuous family of conditions, one for each value of α . However, in the case of larger systems, one can perform a quick check, namely: we find that for any distribution p we can construct smoothed distributions that are very close to p , and in terms of these smoothed distributions, check two conditions in terms of the two free energies for $\alpha = 0, \infty$ found in ref. 2. If such conditions are satisfied on the smoothed distribution, it implies that the infinite set of conditions is satisfied as well.

For $\alpha \rightarrow 1$, $F_\alpha(\rho, \rho_\beta)$ is equal to the ordinary Helmholtz free energy $F(\rho)$, hence our conditions include the ordinary second law (combined with energy conservation), and we thus see that it is merely one of many constraints on thermodynamical state transitions.

In the macroscopic regime, and for systems which are not highly correlated, then $F_\alpha(\rho, \rho_\beta) \approx F_1(\rho, \rho_\beta)$ for all α (2, 8), which explains why the single constraint given by the usual second law is more or less adequate in this limit. It was previously found that the quantity $F_{\min}(\rho)$, defined in ref. 2, gives the maximal amount of work extractable from a system in contact with a reservoir under all thermal operations (2) (by transforming it to a thermal state in equilibrium with the bath). This is also the relevant quantity in a model of alternating adiabatic and isothermal operations (9). We see this in our newly derived second laws as

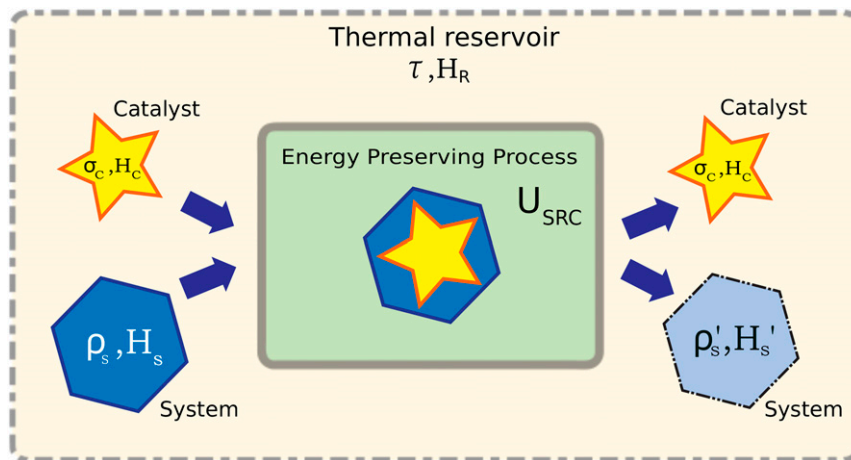


Fig. 1. In the microregime, when can a state ρ_S with Hamiltonian H_S be transformed to a state ρ'_S and Hamiltonian H'_S ? To do so, one can couple the system to a heat bath $\rho_\beta = e^{-\beta H_R} / Z$ with Hamiltonian H_R and use any devices as long as they are returned back in their original state (thus we may think of them as a catalyst— σ) and we are allowed to perform any action as long as we preserve the overall energy (see below for a more detailed description of these operations, which we call catalytic thermal operations). Loosely speaking, our second law says that ρ_S can transit to ρ'_S if and only if ρ'_S is closer to the thermal state ρ_β of the system at inverse temperature β with respect to all Rényi divergences. In the thermodynamic limit, all these quantities converge so that we recover the usual second law.

thought of as a distance measure between states, as was done with the thermo-majorization criterion in ref. 4 and we will henceforth refer to it as the work distance from ρ to ρ' .

Approximately Cyclic Processes

As in the context of entanglement theory (17–20), we consider thermodynamics as a resource theory (2, 21–23), where we are allowed to implement a class of operations, and then quantify the resources which cannot be created under the class of operations. For thermodynamics, various classes of operations have been considered in the microregime (2, 9, 21, 24–32). In particular, we consider thermal operations (2, 21, 23) where we allow the systems of interest ρ to be coupled to a thermal reservoir ρ_R in the thermal state at temperature T , and we allow arbitrary unitaries between the system, working body, and reservoir which conserve energy. Energy conservation is important, because we need to account for all sources of energy which might get added to our system. Operations which pump energy into or from the system can be incorporated into the paradigm by including the source of energy as an ancillary system. This paradigm is equivalent to ones in which we allow interaction Hamiltonians rather than unitaries, or where we allow for a Hamiltonian which changes with time, provided that all ancillas are carefully accounted for (23).

In this article we consider a natural additional ingredient, namely, we allow additional resources such as a heat engine and working body (or ancilla system) C in state ρ_C^{in} which must be returned in its initial form. Considering such resources is crucial because the experimenter who is trying to extract work or otherwise manipulate a system should be allowed as much ingenuity as possible. However, to ensure that work is not being added to the system from the heat engine itself, we demand that the thermodynamic process be cyclic, in the sense that the additional resources are returned in state ρ_C^{out} , which is approximately equal to its original state ρ_C^{in} . In essence, how cyclic a process is can be understood in terms of how good an approximation ρ_C^{in} is to ρ_C^{out} . In the macroscopic case, the notion of cyclicity in a process has not been deemed important—it was assumed that it was enough to take the initial and final state of the heat engine as being close to the same macroscopic state. We find that this is not the case, even if one demands that the initial and final states are arbitrarily close. This is increasingly important when we are manipulating microscopic systems, where small differences can have more noticeable effects. We find that depending on the desired approximation, i.e., depending on how cyclic we demand the process to be, there are several different regimes of second laws.

The simplest case is where $\rho_C^{\text{in}} = \rho_C^{\text{out}}$, that is, the process is perfectly cyclic and the catalyst is restored to its original form. In this setting, we have the second law as stated above. However, no real process is perfectly cyclic, and so it is important to consider the case where $\rho_C^{\text{in}} \approx \rho_C^{\text{out}}$. This requires us to derive approximate transformation conditions which we expect to also find application in entanglement theory, and are contained in *SI Appendix, section G*. We find that the form the second law takes when the process is not perfectly cyclic depends on the degree to which our process is cyclic. We find three separate regimes which are quantified by how cyclic the process is, in terms of how close ρ_C^{in} is to ρ_C^{out} .

In the first regime, we demand that the change in the working body through a cycle is small, in the sense that $D_{\text{work}}(\rho_C^{\text{out}} > \rho_C^{\text{in}}) \leq \epsilon$. In other words, any change in the working body could be corrected by applying a small amount of work. In this case, we recover the second laws as stated above.

The second regime is when the change in the working body has error inversely proportional to the number of particles it has, i.e., $\|\rho_C^{\text{in}} - \rho_C^{\text{out}}\|_1 \leq \epsilon/\log N$, where N is the dimension of the catalyst. In this case, we retrieve the standard second law. The ordinary free energy continues to govern whether a thermodynamical transition is possible, whereas the Rényi divergences do not. We thus see that the ordinary second law can arise in the

macroscopic limit, or if we allow processes which deviate from being cyclic in a manner which is constant per number of particles in the working body. We detail this in *SI Appendix, section F4* by showing that when the standard second law holds, we can construct a catalyst to enable the transition while being returned with small error per particle.

Finally, we consider the regime where we simply demand that the process is close to cyclic regardless of the size of the ancillary system, i.e., $\|\rho_C^{\text{in}} - \rho_C^{\text{out}}\|_1 \leq \epsilon$. Because ϵ can be arbitrarily small, one would imagine that for such an approximately cyclic process one recovers a second law of some sort. Nonetheless, we find that for any ϵ , no matter how small, one can construct a working body, and cyclic process, such that one can pump heat from a cold reservoir to a hot reservoir, in violation of the Clausius statement of the second law. In fact, we can make arbitrary state transformations by taking the size of the working body to be so large that work can be extracted from a single heat bath, while barely modifying the state of the working body. This is related to a phenomenon in entanglement theory known as embezzling (33). For example, for any desired approximation ϵ there exists a dimension d such that the catalyst $\rho_C^{\text{in}} = \sum_{j=1}^d 1/j|j\rangle\langle j|$ allows us to transform any initial state ρ to any final state ρ' such that $\|\rho_C^{\text{in}} - \rho_C^{\text{out}}\|_1 \leq \epsilon$.

Discussion

The second law is often seen as arising from an experimenter's lack of control over the system of interest. Here we see that this is not the case—we obtain our fundamental limitations even in the case where the experimenter can access the microscopic degrees of freedom of the heat bath and couple it in an arbitrary way with the system. The reason that such fine control does not lead to a violation of the second law is related to the fact that a Maxwell's demon with microscopic control over a system cannot violate the second law—a demon which knows the positions and momenta of the particles of a system, must record this information in a memory, which then needs to be reset at the end of a cyclic process (34, 35). For the same reason, an ability to access the degrees of freedom of the heat bath would also require work to perform such a memory resetting step. Remarkably, although the limitations are derived assuming that one can perform all possible operations, work distillation and formation is achievable using a very limited set of operations—namely, changing the energy levels of the system, and putting parts of the system in thermal contact with the reservoir.

We have derived a family of fundamental limitations on thermodynamical state transformations for both quasi-classical states and fully quantum states. Because these limitations are given in terms of generalizations of the free energy, they can be thought of as second laws, combined with the first law, i.e., energy conservation. For an isolated system, one could take the second law to be the increase in the Rényi entropies, which holds if the allowed class of operations are mixtures of unitaries. This can be thought of as resulting from a coarse-graining or lack of information about the full dynamics, but we do not consider this approach in detail here.

Thinking of thermodynamics as a resource theory allows us to reformulate the laws of thermodynamics in a very natural way. In essence, the zeroth law defines the set of allowed free states (the thermal state), the first law the set of allowed operations (namely, energy conserving unitaries), and the second law is derived from these conditions to specify the set of allowed transitions. This has the advantage of separating out laws of fundamental physics, e.g., that evolution be unitary and energy conserving, from those of thermodynamics.

To state the zeroth and first law of thermodynamics more explicitly, let us define the set of catalytic thermal operations introduced here.

- Catalytic thermal operations:

Given a system in initial state ρ_S with respect to Hamiltonian H_S , and a fixed parameter $\beta > 0$ (denoting the inverse temperature of the thermal reservoir), we can

- i) borrow a catalyst system in state σ with regard to some Hamiltonian H_C and return it in a state at least ε close to σ . What one means by close will determine which family of second laws apply, and in the subsequent statement we invoke the most stringent conditions, namely closeness in terms of work distance,
- ii) add an arbitrary number of copies of states τ from a set \mathcal{S} . The zeroth law, stated below, will imply that the only choice is $\mathcal{S} = \{(\tau, H_R) | \tau = e^{-\beta H_R} / Z_R\}$ with any Hamiltonian H_R , and Z_R being the partition function of H_R ,
- iii) perform any unitary operation U such that $[U, H_S + H_R + H_C] = 0$ (the first law), and
- iv) perform partial trace over systems R and C .

Note that the demand that the unitary commutes with the total Hamiltonian implies that energy is conserved. Conversely, it is easy to see that the only processes that conserve energy for an arbitrary state must commute with the Hamiltonian. We emphasize that the equivalence of this paradigm to others has already been addressed in ref. 23. We here view the first law not as something which is a consequence of thermodynamics, but rather one which defines what thermodynamics is.

•Zeroth law:

Let $\mathcal{S} = \{(\tau, H_R) | \tau = \rho_\beta^R = e^{-\beta H_R} / Z\}$ is a thermal state for H_R . If $(\tau, H_R) \notin \mathcal{S}$, then arbitrary state transitions are possible and the theory becomes trivial. This is derived in *SI Appendix, section D*.

We thus see that the ordinary zeroth law is replaced by the following fact: if our class of operations includes energy-conserving operations and the ability to add an arbitrary number of copies of some state τ corresponding to a Hamiltonian H_R , then the only pair of (τ, H_R) which does not make for a trivial theory (in the

sense that all state transformations would be possible), is if $\tau = \rho_\beta$, where ρ_β is the thermal state (2) with respect to H_R . This is related to the fact that the thermal state is the only state that is completely passive, i.e., one cannot draw work from arbitrary number of copies of the state (36). Taking a resource perspective, we now see we have an equivalence relation on \mathcal{S} which defines for us the notion of temperature. More precisely, we will call (τ_1, H_{R_1}) and (τ_2, H_{R_2}) equivalent resources if and only if no work can be gained from $\tau_1^{\otimes \ell_1} \otimes \tau_2^{\otimes \ell_2}$ for arbitrary number of copies ℓ_1 and ℓ_2 . Thus, the ordinary zeroth law is replaced here by a unique condition which tells us what class of free states makes thermodynamics nontrivial.

Here, our derivation of the second laws is information theoretic in nature, requiring none of the assumptions usually required for the second law to hold. This includes ergodicity, mixing, coarse-graining of degrees of freedom, and lack of control over the system. Monotonicity of $F_\alpha(\rho, \rho_\beta)$ thus provides a powerful tool to determine what sorts of thermodynamical transitions are possible on the quantum scale, or equally well for systems which have long-range interactions. From a foundational perspective, the laws of thermodynamics take on a very simple and elegant form—a class of operations and a set of statistical distances to the free state ρ_β which can never decrease. One hopes that such information theoretic laws can be used to discover a broad range of thermodynamical phenomena at the quantum level.

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