Operational thermodynamics of open quantum systems

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Accurately describing work extraction from a quantum system is a central objective for the extension of thermodynamics to individual quantum systems. The concepts of work and heat are surprisingly subtle when generalizations are made to arbitrary quantum states. We formulate an operational thermodynamics suitable for application to an open quantum systems undergoing a general quantum evolution. We derive the first law of thermodynamics for a process described by a completely-positive and trace-preserving map and show consistency with the Hatano-Sasa statement of the second law. We show that heat, from the first law, is positive when the input state of the map majorises the output state. Moreover, the change in entropy is also positive for the same majorisation condition. This makes a strong connection between the two operational laws of thermodynamics.

Introduction.— The laws of thermodynamics were forged in the furnaces of the industrial revolution, as engineers and scientists refined their picture of energy, studying heat and its interconversion to mechanical work with a view to powering the mines and factories of this new era of human endeavor. Followed by the development of statistical mechanics at the change of the centuries [1], far from its pragmatic inception, thermodynamics is now a theory with a remarkable range of applicability, successfully describing the properties of macroscopic systems ranging from refrigerators to black holes [2].

Moving on to the 21st century with both the industrial and electronic revolutions behind us, we are currently pushing technology towards and beyond the microscopic scale. With a view to devices operating at a scale where quantum mechanical laws become important we may ask whether the solid grounds of thermodynamics might be challenged, not only by the lack of a thermodynamic limit, but also by the intrinsic uncertainty synonymous with this domain. It comes as no surprise that there has been a concerted effort to understand how the laws of thermodynamics generalize to arbitrary quantum systems [3] both at and away from equilibrium. Such laws will aid in better understanding the relationship between quantum and statistical mechanics, extend our predictability for out-of-equilibrium systems and aid the design of efficient controls for thermal machines.

An important question relating to the extension of the first law of thermodynamics into such a regime is to ask “to what extent do the concepts of work and heat extend to quantum systems?” This is an avenue of research that has been open for several decades [3] [4]. Without severe assumptions regarding the set of allowed quantum states, coupling strengths and bath-properties it has so far remained a difficult question without any satisfactory general answer. Nevertheless it is central for the formulation of a concrete theory of quantum thermodynamics of both equilibrium and non-equilibrium systems. Some important steps have been made toward providing an answer (each with its own strengths), such as the formulation of quantum fluctuation relations [5], information theoretical approaches [6] (in specific see [7] for an overview), and some combination of the two [8]. Finally, central to the work presented here is a work extraction formalism for non-passivity of quantum states [9]. Despite the range of approaches a more general picture for the thermodynamics of general quantum evolutions is far from clear.

In this Letter, we take an operational approach to characterizing the energy change of an open quantum process described by a completely-positive trace-preserving (CPTP) map. In analogy to the first law of thermodynamics we discuss work done, extractable work, and heat. The concepts of ergotropy and adiabatic work allow us to state our main result: An operational first law for general quantum processes. We show that our operational first law is in agreement with widely used Hatano-Sasa version of the second law for CPTP maps [10] [11] by explicitly stating the Clausius inequality for unital and thermal maps. We then show that both operational heat and the change in von Neumann entropy are positive when the input state of the map majorises the output state.

Thermodynamics of quantum systems.— The first law of thermodynamics states that the internal energy change in a thermodynamic process can be split up into two contributions – work and heat: \(dE = \delta Q + \delta W\). For a generic quantum system, the internal energy at time \(t\) is \(E(t) = \text{tr}[\rho(t)H(t)]\), implying that the change in the internal energy \(dE\) depends only on the end points. Heat and work on the other hand are path-dependent—thus the different notation for the ‘differentials’. As an illustration we may consider the heat expended when pushing a piston into a cylinder filled with gas: It not only depends on the initial and final positions of the piston but also on how fast it is pushed. Using the derivative of the internal energy with respect to time the following two expressions are motivated [10]:

\[
\delta W = \text{tr}[\rho(t)d_tH(t)]dt \quad \text{and} \quad \delta Q = \text{tr}[H(t)d_t\rho(t)]dt,
\]

with \(d_t := d/dt\). Integrated over a specific evolution this
yields (average) values for heat and work
\[
\Delta E = \int_0^\tau \delta W + \int_0^\tau \delta Q = \langle W \rangle + \langle Q \rangle \\
= \text{tr}[\rho(\tau)H(\tau)] - \text{tr}[\rho(0)H(0)].
\] (2)

These definitions for work and heat fit the understanding that heat corresponds to a change in the state and accordingly, entropy, a function of state. For unitary evolution it vanishes by virtue of the Liouville-von Neumann equation: \( d_\tau \rho(t) = i[H(t), \rho(t)] \). The part corresponding to work, on the other hand, does not relate to a change of the state or its entropy but rather to a change in the energy landscape.

In general, however, it is not easy to compare work and heat for a generic quantum process. This is because the integrands in the last equation are often neither well-defined nor easy to measure. Only for systems with well-defined descriptors of \( \rho(t) \) and \( d_\tau \rho(t) \), we have a closed form for work and heat. For instance, Markov systems are described in a time local form \( d_\tau \rho(t) = C[\rho(t)] \), leading to the well-known results \([10]\). For non-Markovian systems, on the other hand, \( d_\tau \rho(t) \) is not well defined since it may depend on the state of the system at previous times.

Specifically, we may be interested in a process whose dynamical transformation from the initial state to a final state is well defined in terms of a CPTP map, but whose intermediate dynamics is not. Far from being an academic issue, this is a very realistic and practical problem. For instance, in a chemical process the Hamiltonian dynamics as well as the reduced state at all times are generally not known. The end points however may be accessible and the process may be described as stochastic process. In this scenario it is very reasonable to ask what the different energy contributions to the process are even if the time-resolved evolution is not known. In this Letter we give a first law of thermodynamics for quantum processes in terms of equilibrium quantities like adiabatic work and heat, and an operational quantity called ergotropy.

**Ergotropy and cyclic work extraction.**—We proceed with a brief review of work extraction in cyclic unitary evolution \([9, 12]\): Given a quantum state \( \rho \) on a finite-dimensional Hilbert space and a Hamiltonian \( H \) we may ask how much work can be extracted via a cyclic unitary process. Cyclicity here means that the system Hamiltonians at the beginning and the end of the process have to be identical, i.e., \( H \equiv H(0) = H(\tau) \). For unitary evolution any change in internal energy \( \langle H \rangle \) is due to work. We express the Hamiltonian in its increasing spectral decomposition: \( H := \sum \epsilon_n |\epsilon_n\rangle \langle \epsilon_n| \) with \( \epsilon_{n+1} \geq \epsilon_n \ \forall \ n \). The state \( \rho \) on the other hand is expressed in its decreasing eigen-decomposition: \( \rho := \sum r_n |r_n\rangle \langle r_n| \) with \( r_{n+1} \leq r_n \ \forall \ n \). The goal is to transform \( \rho \) into a state with lower internal energy, extracting the difference in internal energy in the process.

After maximal cyclical work extraction, no more work can be extracted using a cyclic unitary process and the system ends up in a so-called passive state \( \pi \) \([9, 12]\). A passive state is unique up to degeneracies in the Hamiltonian \([13]\). Passive states are diagonal in the Hamiltonian’s eigenbasis with decreasing populations for increasing energy levels. That is, a state \( \rho \), as defined above, is passive when \( |r_n| = |\epsilon_n| \ \forall \ n \). Note that Gibbs states are always passive.

The maximum work that can be extracted from a non-passive state \( \rho \) with respect to a Hamiltonian \( H \) via a cyclic unitary process (yielding a passive state \( \pi \)) is called ergotropy \([9]\):

\[
W := \text{tr}[\rho H] - \text{tr}[\pi H] \\
= \sum_n r_n \epsilon_n \left[ (|\epsilon_n| r_n)^2 - \delta_{mn} \right].
\] (3)

Ergotropy is always positive and has the opposite sign from the conventional definition of work. Its relation to entanglement has recently been explored in \([14]\). We may write \( W(\rho, H) \) in order to explicitly state the dependence on the pair \((\rho, H)\) of state and Hamiltonian.

**Adiabatic work.**—Before we give the main result we need to define one more concept. Consider now a non-cyclic, unitary process with different initial and final Hamiltonian \( H \) and \( H' := \sum \epsilon'_n |\epsilon'_n\rangle \langle \epsilon'_n| \) again with \( \epsilon'_{n+1} \geq \epsilon'_n \). If we restrict the change in the Hamiltonian from \( H \) to \( H' \) to be adiabatic in the quantum sense, i.e., the eigenstates of the Hamiltonian remain eigenstates at each instant \([15]\); the final state will be a passive state with respect to the final Hamiltonian if the initial state was passive with respect to the first Hamiltonian. Since this transformation is unitary there is no heat, and we call this the adiabatic work:

\[
\langle W \rangle_{ad} = \text{tr}[\pi'_m H'] - \text{tr}[\pi H],
\] (4)

where \( \pi'_m = \sum r_n |\epsilon'_n\rangle \langle \epsilon'_n| \). Now we move to the open dynamics regime.

**First law for open quantum evolution.**—Non-unitary dynamics between two time-steps of a quantum process is described by a CPTP map: \( \rho \rightarrow \rho' := M(\rho) \). Such maps are ubiquitous in modern quantum theory. This type of evolution is equivalent to coupling the system to an initially uncorrelated ancilla, letting system and ancilla unitarily evolve together, and decoupling the system again. Our goal in this Letter is write down a first law of thermodynamics for such a process. However, we do not have any knowledge about the energetics for the ancilla. Moreover, we do not have any information about the state of the system or the Hamiltonian at any intermediary times. Therefore computing work and heat in Equation 2 is not possible for such a process.

Instead, we begin by considering the change in internal energy due to this evolution:

\[
\Delta E = \text{tr}[\rho H'] - \text{tr}[\rho H],
\] (5)

where we have labeled the initial and final system Hamiltonian \( H \) and \( H' \). To this we add and subtract \( \text{tr}[\pi H'] \), \( \text{tr}[\pi'_m H'] \), and \( \text{tr}[\pi H] \) to get

\[
\Delta E = \text{tr}[\pi H] - \text{tr}[\rho H] + \text{tr}[\rho H'] - \text{tr}[\pi H'] \\
+ \text{tr}[\pi'_m H'] - \text{tr}[\pi H] + \text{tr}[\pi H'] - \text{tr}[\pi'_m H'].
\] (6)
The first two pairs terms are simply the ergotropies $-W(\rho, H)$ and $W(\rho', H')$, while the next pair is adiabatic work. Let us define $\Delta W = W(\rho', H') - W(\rho, H)$ and the operational heat $\langle Q \rangle_{op} = \text{tr}[\pi' H'] - \text{tr}[\pi'' H']$ to get our main result:

$$\Delta E = \Delta W + \langle W \rangle_{ad} + \langle Q \rangle_{op}. \tag{7}$$

The last equation tells us that the internal energy change in a general quantum process can be split up into a work-like, a heat-like and a third, genuine out-of-equilibrium contribution that equals the difference in ergotropy between initial and final state. The definition of heat is justified as the eigenvalues of equilibrium change in the internal energy change between $\rho$ and $\rho'$ (purple) using fictitious operational process of extracting ergotropy $\rho \to \pi$ (blue), the equilibrium processes of doing adiabatic work $\pi \to \pi'$ (green), equilibrium heating $\pi_m \to \pi'$ (orange), and finally depositing ergotropy $\rho' \to \pi'$ (blue) to close the loop. The relationship between these quantities is is given in [Equation 1](#).

Consequently, we can interpret the CPTP map as a sequence of fictitious thermodynamic processes: first we extract work from $\rho$ by transforming it to $\pi$; then we adiabatically change $H$ to $H'$ and hence $\pi$ to $\pi_m'$; we then heat $\pi_m'$ by changing its spectrum to $\pi'$; and finally we deposit work onto $\pi'$ and change into $\rho'$ [15]. These steps are illustrated in [Figure 1](#).

It is important to notice that the work and heat represented here do not correspond to the work and heat in [Equation 1](#). To solve for [Equation 1](#) we would have to take the full dynamics and the ancilla into account.

The present relations rather take the form of an operational first law: Each part of the sum requires a different type of operation. To change $\Delta W$ cyclic unitary evolution suffices. For a general unitary process, $\langle W \rangle_{ad}$ can be drawn in addition. For $\langle Q \rangle_{op} \neq 0$ a general CPTP map is necessary (a non-unital one to achieve $\langle Q \rangle_{op} < 0$ as will be demonstrated below).

It has long been convenient to express the properties of out-of-equilibrium systems by using quantities that relate to equilibrium states and hence can be computed in a straightforward fashion. Furthermore, the equilibrium quantities relate to measurements that can predict the non-equilibrium properties of systems of interest. Similar to other works in statistical mechanics relating to fluctuation theorems [5] we break up the out-of-equilibrium energy change into equilibrium quantities adiabatic work and heat and an operational quantity ergotropy. We now relate these results to the second law of thermodynamics.

**Connecting first and second laws.**— Interestingly, a second law for CPTP maps is well-known [11]. In the context of equilibrium thermodynamics, the Clausius inequality states that the thermodynamic entropy of any system and its environment is non-decreasing. For such systems, owing to the notions of temperature $\beta^{-1}$, thermodynamic entropy $\Delta S$ and heat $\langle Q \rangle$ being well defined, the second law can be stated as $\Delta S \geq \beta \langle Q \rangle$.

To generalize this to the quantum regime, von Neumann entropy, $S(\rho) := \text{tr}[\rho \log(\rho)]$, is considered in the place of thermodynamic entropy (being equivalent for thermal states). The second law for arbitrary states undergoing CPTP evolution is a direct consequence of the property of contractivity of relative entropy under CPTP maps. Here, relative entropy is defined as $S[\rho||\sigma] := \text{tr}[\rho \log(\rho) - \rho \log(\sigma)]$ [17]. Under generic CPTP evolution described by a map $M$, relative entropy obeys contractivity [18], namely $S[\rho||\sigma] \geq S[M(\rho)||M(\sigma)]$. Since we are interested in the change in entropy $\Delta S := S(M(\rho)) - S(\rho)$, we have the choice of a reference state $\sigma$. The obvious choice of $\sigma$ is the fixed point $\epsilon$ of the map $M$, namely $M(\epsilon) = \epsilon$. Rearranging the contractivity inequality, we arrive at the quantum version of the Hatano-Sasa inequality [19]. This inequality is given by $\Delta S \geq -\text{tr}[\{M(\rho) - \rho\} \log(\epsilon)]$.

While the first law relates to the partitioning of energy into heat and work, the (Clausius form of the) second law relates only to the increase in entropy. Specifically, the quantum Hatano-Sasa inequality is valid for CPTP evolution where neither heat nor temperature are well-defined quantities. Hence, in general it is difficult to verify the internal consistency between a quantum generalization of the first law and a similar generalization of the second law that is applicable to arbitrary CPTP dynamics. However, we establish a relation between the two laws by considering thermal maps.

**Thermal maps.**— We call a map thermal if it has a thermal state for a fixed point: $\epsilon = \pi_\beta = \exp(-\beta(H - F))$ at some inverse temperature $\beta$, where $F$ is the (Helmholtz) free energy. We remind the reader that a thermal state is a passive state. In order to make the connection to the second law a cyclic process is considered, i.e., $H = H'$. The input and output states $\rho$ and $\rho'$ are not restricted and can both be out-of-equilibrium. The quantum Hatano-Sasa inequality now re-
where two states satisfy \( \rho \). This restatement of the quantum Hatano-Sasa inequality is interesting from the standpoint that the change in entropy could be thought of as being lower bounded by the sum of two terms, the change in entropy and the operational heat. Since these terms are measurable and operationally well defined, the second law can be reinterpreted as being a statement about the lower bound on the entropy increase being set by two operationally relevant quantities derived from the first law.

**Majorisation, entropy, and heat.**— We now give a condition for when heat \( \langle Q \rangle_{op} \) is strictly positive. In order to do so we need to introduce the concept of majorisation. A state \( \rho \) is said to majorise \( \rho' \) (written \( \rho \succ \rho' \)) if the eigenvalues of the two states satisfy

\[
\sum_{m=1}^{n} r_m \geq \sum_{m=1}^{n} r'_m \quad \forall \quad n
\]

where \( \rho' = \sum_{m=1}^{n} r'_m |n'_m \rangle \langle n'_m | \) with \( r'_{n+1} \geq r'_n \).

The thermo-majorisation condition provides a sufficient criterion for heat to be positive: If \( \rho \succ \rho' = M(\rho) \) then \( \langle Q \rangle_{op} \geq 0 \). Since the eigenvalues of the states do not change during the ergotropy extraction phase \( \rho \succ \rho' \) implies \( \pi \succ \pi' \).

Examining the expression for \( \langle Q \rangle_{op} \) we have:

\[
\langle Q \rangle_{op} = \text{tr}[\pi' H] - \text{tr}[\pi H] = \sum_{n} (r'_n - r_n) \epsilon_n
\]

where each term in the last line is positive \([20]\).

Moreover, if \( \pi \succ \pi' \) then \( f(\pi') \geq f(\pi) \) for any Schur-concave function \( f \) \([21]\). The means \( S(\rho') \geq S(\rho) \) and therefore \( \Delta S \geq 0 \). The implication is rather profound: The thermo-majorisation condition guarantees that both the entropy change and heat are positive simultaneously. This can be thought of as a version of the second law. The second law in the context of equilibrium thermodynamics guarantees that \( \langle Q \rangle \geq 0 \). \( \Delta S \geq 0 \) and that the latter is at least as big as \( \beta \) times the former. But in the context of quantum thermodynamics, no such guarantee exists in general. Consequently, one can have cooling transformations that reduce the entropy of the states \([19]\). Thermo-majorisation strongly restricts the set of allowed transformations to only be ones that increase both the heat and the entropy of the systems under interest.

**Unital maps.**— Unital maps take the maximally mixed state onto itself: \( M_u(1/d) = 1/d \). This simple condition has strong consequences: The quantum Hardy-Littlewood-Polya theorem \([22]\) demonstrates that \( \rho \succ M_u(\rho) \) for any \( \rho \) if \( M_u \) is unital. As a consequence of the majorisation arguments above both \( \langle Q \rangle_{op} \) and \( \Delta S \) are thus positive for any input state \( \rho \) and unital map \( M_u \).

For all non-unital maps, like the thermal ones described above, there exist at least one state \( \rho \) that is majorised by the outcome state \( \rho' \) and in those cases will increase the amount of accessible work, i.e., the last term in \( \text{Equation 10} \) is negative and therefore heat is negative. The change in entropy for such a process will also be negative. The directionality that comes with the second law of thermodynamics is here reflected in the asymmetry between unital and non-unital maps: A heat-like increase of energy (and entropy) of a state only requires a unital map, whilst extraction corresponding to a negative heat-like contribution necessitates a non-unital channel. The representation of such a channel in terms of an ancilla makes clear that the second law is not ‘violated’ here.

**Conclusion.**— In summary, we have established a relation that gives an explicit energy balance for a general quantum process. Crucially, we have formulated what we consider an operational framework for the thermodynamics of an open quantum system. The important feature of our framework is that it relies only on CPTP maps for processes and input and output states. For such processes we have operationally defined heat and connected it to an operational second law. Both heat and change in entropy are shown to be positive when the input majorises the output, making a strong connection between the operational laws.

The universal nature of our framework should render it of large interest to a range of different communities such as quantum opticians and chemists interested in the thermodynamics of open quantum systems. It can be a useful tool when combined with recent experimental and theoretical progress on the extraction of work and heat statistics using so called phase estimation schemes \([23]\). Using the present formalism with these schemes to test the ideas of this Letter is currently work in progress.

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