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Letter

Entropy-based formulation of thermodynamics in arbitrary quantum evolution

S. Alipour,[1] A. T. Rezakhani,[2] A. Chenu,[3,4,5] A. del Campo,[3,4,5,6] and T. Ala-Nissila[1,7]

[1]*QTF Center of Excellence, Department of Applied Physics, Aalto University, P.O. Box 11000, FI-00076 Aalto, Espoo, Finland*
[2]*Department of Physics, Sharif University of Technology, Tehran 14588, Iran*
[3]*Department of Physics and Materials Science, University of Luxembourg, L-1511 Luxembourg, Grand Duchy of Luxembourg*
[4]*Donostia International Physics Center, E-20018 San Sebastián, Spain*
[5]*IKERBASQUE, Basque Foundation for Science, E-48013 Bilbao, Spain*
[6]*Department of Physics, University of Massachusetts, Boston, Massachusetts 02125, USA*
[7]*Interdisciplinary Centre for Mathematical Modelling and Department of Mathematical Sciences, Loughborough University, Loughborough, Leicestershire LE11 3TU, United Kingdom*

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Given the evolution of an arbitrary open quantum system, we formulate a general and unambiguous method to separate the internal energy change of the system into an entropy-related contribution and a part causing no entropy change, identified as heat and work, respectively. We also demonstrate that heat and work admit geometric and dynamical descriptions by developing a universal dynamical equation for the given trajectory of the system. The dissipative and coherent parts of this equation contribute exclusively to heat and work, where the specific role of a work contribution from a counterdiabatic drive is underlined. Next we define an expression for the irreversible entropy production of the system which does not have explicit dependence on the properties of the ambient environment; rather, it depends on a set of the system's observables excluding its Hamiltonian and is independent of internal energy change. We illustrate our results with three examples.

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Quantum thermodynamics holds center stage at the interface of quantum information science, statistical mechanics, and quantum technologies, and it has shed new light on the laws of thermodynamics in the quantum regime [1–15]. Apart from uncovering a plethora of new phenomena, quantum thermodynamics has also motivated efforts to engineer efficient quantum machines in the laboratory [16–20].

Yet, several fundamental issues remain to be clarified. Most notably, except for particular regimes (e.g., weak-coupling, Markovian dynamics with slow driving [21]), an unequivocal definition of heat and work in arbitrary open-quantum system dynamics has been elusive. One problem is that such thermodynamic variables are not observables described by Hermitian operators [22]; rather, they are trajectory-dependent quantities [23–27]. Existing definitions mostly incorporate generators of the dynamics (besides the state) and dynamical master equations with *coherent* and *dissipative* parts [10,21,28–32]. For instance, in the widely-used *conventional* framework [21,33] the internal energy change due to the dissipative (coherent) part of the master equation has been called heat (work) change. An alternative strategy uses the potential of mean force that amounts to preaveraging the total partition function over the environmental degrees of freedom [28–32]. In addition, there are *semiclassical* approaches where a coarse-grained version of the system state in the energy eigenbasis and semiclassical definitions of heat and work have been employed [34–36].

Despite extensive efforts, some of the existing approaches neither are consistent with each other nor always reproduce expected results according to thermodynamics. Although there exist attempts to overcome such inconsistencies through *ad hoc* methods [32,37,38], the issue remains unresolved.

To further clarify the issue, we note the following drawbacks of the conventional framework. (i) Its formulation has been originally devised only for particular conditions, e.g., a Markovian dynamics with a constant or relatively slow-varying Hamiltonian, when the dissipative part does not explicitly depend on the physical system Hamiltonian. Despite this subtlety, the conventional definitions of heat and work have been applied almost arbitrarily in the literature to more general scenarios. In fact, the dissipative part of a master equation may explicitly depend on the physical Hamiltonian of the system [39,40]. Hence in such physical scenarios, external driving may also contribute to the heat exchanged. This aspect introduces further ambiguities and hinders a clear and thermodynamically consistent assignment of the heat and work concepts. (ii) Since the description of the dynamics in the form of the conventional Lindblad master equation is not unique [41], different energy values can be assigned to differently chosen generators of the same dynamics. This indicates that part of the energy assigned to the dissipative part of the master equation may not necessarily lead to an entropy change, and should be identified as work. Specifically, for initial states of the decoherence-free subspace type [42,43], the dynamics leads to no dissipation along the corresponding trajectories, and all the energy change, if any, would be of the work type. Recently, a scheme to partially remedy the shortcomings of the conventional definitions has been proposed in Ref. [44], where heat has been defined by subtracting *ergotropy* (maximum extractable work from a system in a cyclic unitary process) [45] from the dissipative part of the internal energy change by considering a *virtual* instantaneous unitary transformation. Nevertheless, an extension of ergotropy to open systems is still elusive, and since this extra transformation does not rely on the real physical dynamics, trajectory-dependent quantities such as heat can be fictitious. For an alternative approach, separating the internal energy based on conserved quantities of the dynamics, see Ref. [46].

In this Research Letter, on pure thermodynamic grounds supplemented with dynamical and geometric arguments, we circumvent the issue and ambiguities discussed above and put forward a set of universal definitions for heat, work, and entropy production based explicitly on trajectories in the state space of a quantum system (note the difference with quantum jump *trajectories* arising in stochastic dynamics of monitored systems [47,48]). Our framework is general and independent of the dynamics which generates the trajectory, and it is thus applicable to any time-continuous evolution.

In particular, following standard thermodynamics wherein energy change in a reversible isentropic process is identified as work [49,50], we define heat solely as the part of the internal energy of the system which can be associated with entropy change. While the first law of thermodynamics treats heat and work on an equal footing, the second law distinguishes them. Essentially, heat is a form of disorganized energy, and some disorganization (entropy) will flow with it [51]. Hence heat is introduced as the part of the energy change which can be accompanied by an entropy change, whereas the energy transfer in the form of work definitely does not lead to any entropy change. We use this key feature to identify heat and work and accordingly obtain a computable expression for the irreversible entropy production. We show that this quantity varies due to the change in variables of the system other than its energy. This clarifies why in general scenarios heat and entropy changes are not necessarily monotonic with respect to each other.

Consider the evolution of the system in a time interval $t \in [0, t_f]$ described by a time-dependent density matrix $\varrho(t)$, expressed in its spectral decomposition as

$$\varrho(t) = \sum_{k=1}^{D} r_k(t) |r_k(t)\rangle\langle r_k(t)|. \tag{1}$$

One can consider $\{\varrho(t)\}$ as a *trajectory* in the state space of the system, starting from given $\{\varrho(0)\}$ (for brevity, we drop all time dependence henceforth unless necessary). Note that the change in ϱ can be decomposed as $d\varrho = \text{đ}\varrho^{(\mathrm{ev})} + \text{đ}\varrho^{(\mathrm{ep})}$, where $\text{đ}\varrho^{(\mathrm{ev})} = \sum_k dr_k |r_k\rangle\langle r_k|$ is the change due to the variation of the eigenvalues and $\text{đ}\varrho^{(\mathrm{ep})} = \sum_k r_k(|dr_k\rangle\langle r_k| + |r_k\rangle\langle dr_k|)$ is due to the variation in the eigenprojectors, where đ denotes inexact differential. In addition, we observe that the entropy $S(\varrho) = -\mathrm{Tr}[\varrho \ln \varrho]$ changes only when the eigenvalues vary,

$$dS = -\sum_k dr_k \ln r_k = -\mathrm{Tr}[\text{đ}\varrho^{(\mathrm{ev})} \ln \varrho]. \tag{2}$$

In a system with the physical Hamiltonian H, the system internal energy $U = \mathrm{Tr}[\varrho H]$ changes along the trajectory with $t \to t + dt$ as $U \to U = U + dU$, with $dU = \mathrm{Tr}[d\varrho\, H] + \mathrm{Tr}[\varrho\, dH]$. Here, the contribution associated with $\text{đ}\varrho^{(\mathrm{ev})}$ is solely related to the change of the eigenvalues, which is in line with how the entropy changes. Hence we define the heat change as

$$\text{đ}\mathbb{Q} = \mathrm{Tr}[\text{đ}\varrho^{(\mathrm{ev})} H] = \sum_k dr_k \langle r_k|H|r_k\rangle \tag{3}$$

and assign the remaining variations to work change,

$$\begin{aligned}\text{đ}\mathbb{W} &= \mathrm{Tr}[\varrho\, dH] + \mathrm{Tr}[\text{đ}\varrho^{(\mathrm{ep})} H] \\ &= \sum_k r_k(\langle r_k|dH|r_k\rangle + \langle dr_k|H|r_k\rangle + \langle r_k|H|dr_k\rangle),\end{aligned} \tag{4}$$

such that

$$dU = \text{đ}\mathbb{Q} + \text{đ}\mathbb{W} \tag{5}$$

encompasses the first law of thermodynamics.

Dynamical analysis of the heat and work definitions. We use a recently proposed *trajectory-based shortcut to adiabaticity* (TB-STA) framework for open-system dynamics, which identifies a particular dynamical equation of motion that generates any given time-dependent density matrix or trajectory [52]. This technique, based on the general Lindblad-like equation described later, is not limited by the system-environment coupling strength or initial correlation. Since the TB-STA equation describes a given trajectory, its coherent and dissipative parts are directly related to the coherences and dissipation in the course of the evolution, which contrast with the conventional Markovian Lindblad master equation. Our main result shows that, among all possible dynamical equations describing a given trajectory, TB-STA allows us to unambiguously separate the change of the internal energy into heat and work for any open system. We prove that energy change assigned to the (so-called) dissipative part of the equation is accompanied by entropy change, and it is therefore *heat*. The coherent part, which does not involve entropy change, corresponds to a *dissipative work*, which for its relation to nonunitarity of the dynamics entails the implicit existence of some ambient environment.

Having a trajectory $\{\varrho(t)\}$ at hand, an associated differential equation describing its dynamics is given by the following Lindblad-like equation [52]:

$$\dot{\varrho} = -i[\mathbb{h}, \varrho] + \mathbb{D}_{\mathrm{CD}}[\varrho], \tag{6}$$

where

$$\mathbb{h} = i\sum_k (|\dot{r}_k\rangle\langle r_k| - \langle r_k|\dot{r}_k\rangle |r_k\rangle\langle r_k|), \tag{7}$$

$$\mathbb{D}_{\mathrm{CD}}[\varrho] = \sum_{kj} \mathbb{c}_{kj}\left(\mathbb{L}_{kj}\varrho\mathbb{L}_{kj}^{\dagger} - \frac{1}{2}\{\mathbb{L}_{kj}^{\dagger}\mathbb{L}_{kj}, \varrho\}\right), \tag{8}$$

$$\mathbb{L}_{kj} = |r_k\rangle\langle r_j|, \quad \mathbb{c}_{kj} = (1 - \delta_{r_j 0})\dot{r}_k/(Dr_j), \tag{9}$$

with the dot denoting time differentiation. Several remarks are in order: (i) $\mathbb{h}$ operates as a *counterdiabatic* (CD) Hamiltonian which alone generates a parallel transport $|r_k(0)\rangle \to |r_k(t)\rangle\ \forall k$ (cf. CD Hamiltonians in the energy sense [53,54]). (ii) In the CD dissipator $\mathbb{D}_{\mathrm{CD}}$ the anticommutator identically vanishes, whence the dissipator reduces exclusively to jumps in the instantaneous eigenbasis of ϱ. (iii) The dynamical equation (6) is universal, irrespective of any physical setting (system Hamiltonian and the environment) in which the system has obtained this trajectory. In the standard technical sense [55,56], this dynamical equation for a trajectory is different from the master equation of the system. The same physical system prepared in a different initial condition with the same physical setting can yield a different trajectory—hence a different dynamical equation associated with the new trajectory. (iv) A Lindblad-like master equation for general open-system dynamics has also been introduced recently [57].

Comparison with the conventional definitions. Along the trajectory, the conventional definitions are $\text{đ}Q=\mathrm{Tr}[d\varrho\, H]$ for heat change and $\text{đ}W=\mathrm{Tr}[\varrho\, dH]$ for work exchange; hence $dU=\text{đ}Q+\text{đ}W$ [cf. Eq. (5)]. Now rewrite $\text{đ}Q$ as

$$\text{đ}Q=\sum_k dr_k\langle r_k|H|r_k\rangle+\sum_k r_k(\langle dr_k|H|r_k\rangle+\langle r_k|H|dr_k\rangle). \tag{10}$$

The first sum originates from a change in the eigenvalues of the state, contributing when not all dr_k's vanish (similar to the reason for the change in S); the second sum is solely associated with the change of the eigenvectors. The first sum can be rewritten as $\mathrm{Tr}[\mathbb{D}_{\mathrm{CD}}[\varrho]H]dt$, which is related to the *dissipative* part of the trajectory dynamical equation (6); whereas the second sum can be recast as $-i\mathrm{Tr}[[H,\mathbb{h}]\varrho]dt$, which is associated with the *coherent* part of the dynamical equation—hence dissipative *work*. From this perspective, we can consider the following relations as a justification of our definitions:

$$\text{đ}\mathbb{Q}=\text{đ}Q-\text{đ}\mathbb{W}_{\mathrm{CD}}=\mathrm{Tr}[\mathbb{D}_{\mathrm{CD}}[\varrho]H]dt, \tag{11}$$

$$\text{đ}\mathbb{W}=\text{đ}W+\text{đ}\mathbb{W}_{\mathrm{CD}}=\mathrm{Tr}[\varrho(\dot{H}-i[H,\mathbb{h}])]dt, \tag{12}$$

where $\text{đ}\mathbb{W}_{\mathrm{CD}}$ is an environment-induced dissipative work,

$$\begin{aligned}\text{đ}\mathbb{W}_{\mathrm{CD}}&=\sum_k r_k(\langle dr_k|H|r_k\rangle+\langle r_k|H|dr_k\rangle)\\&=-i\mathrm{Tr}[[H,\mathbb{h}]\varrho]dt,\end{aligned} \tag{13}$$

and is generated due to the CD evolution. In this framework the work is due to both driving the system through varying its physical Hamiltonian ($\text{đ}W$) and the CD evolution along the trajectory due to the environment ($\text{đ}\mathbb{W}_{\mathrm{CD}}$). Importantly, unlike in Ref. [53], the CD term here is not related to an external physical CD control of the Hamiltonian; rather, it corresponds to the natural CD evolution along the state trajectory.

Relation to the semiclassical definitions. In the *semiclassical* formulation of thermodynamics for quantum systems, heat and work are defined differently [36]. Using the instantaneous eigenbasis of the system Hamiltonian $H=\sum_n E_n|E_n\rangle\langle E_n|$, we obtain $U=\sum_n p_nE_n$, where $p_n=\langle E_n|\varrho|E_n\rangle$ is the population of the energy eigenstate $|E_n\rangle$. From the identity $dU=\sum_n dp_n\, E_n+\sum_n p_n\, dE_n$, one can read the semiclassical heat and work variations as

$$\text{đ}q=\sum_n dp_n\, E_n, \tag{14}$$

$$\text{đ}w=\sum_n p_n\, dE_n. \tag{15}$$

However, if instead of using the instantaneous eigenbasis of H, we evaluate the trace in U in the instantaneous eigenbasis of ϱ [Eq. (1)], then $U=\sum_k r_kH_k$ and $dU=\sum_k dr_k\, H_k+\sum_k r_k\, dH_k$, where $H_k=\langle r_k|H|r_k\rangle$. As in the semiclassical setting, one can now read the first term as the heat change and the second one as the work change. We observe that these are equivalent to the TB-STA definitions [Eqs. (11) and (12)], $\text{đ}\mathbb{Q}=\sum_k dr_k\, H_k$ and $\text{đ}\mathbb{W}=\sum_k r_k\, dH_k$. One can argue that the instantaneous eigenbasis of ϱ is preferred in calculating the energy contributions [58]. A closer inspection of $\text{đ}q$ reveals that it includes the energy (not heat) change $\text{đ}\mathbb{W}_{\mathrm{CD}}$ which we have assigned to the CD Hamiltonian in the form of work,

$$\text{đ}q=\text{đ}\mathbb{Q}+\text{đ}\mathbb{W}_{\mathrm{CD}}+\sum_n E_n(\langle dE_n|\varrho|E_n\rangle+\langle E_n|\varrho|dE_n\rangle). \tag{16}$$

Thus, since $\text{đ}q$ has contributions from both heat and work exchanges (in our sense), we conclude that in general the semiclassical definitions of heat and work fail to properly account for various contributions to the internal energy change.

Irreversible or internal entropy production. The entropy change dS [Eq. (2)], together with the heat change $\text{đ}\mathbb{Q}$ [Eq. (11)], give the change in the irreversible entropy $\mathbb{S}$ as

$$\text{đ}\mathbb{S}\equiv dS-\beta\,\text{đ}\mathbb{Q}=\sum_k dr_k\langle r_k|(\mathbb{H}-\beta H)|r_k\rangle, \tag{17}$$

where $\mathbb{H}=-\ln\varrho$ and β is the nonequilibrium, instantaneous inverse temperature of the system given by (if $k_B\equiv 1$) [59]

$$\beta=(\partial S/\partial U)_{x_2,x_3,\ldots}=\mathrm{Cov}(H,\mathbb{H})/\mathrm{Cov}(H,H). \tag{18}$$

Here, $\{x_i\}$ are a set of independent variables obtained from the expectation values of a complete set of traceless orthonormal observables $\{O_i\}_{i=2}^{D^2-1}$, with $O_0=\mathbb{1}/\sqrt{D}$ (the normalized identity operator of dimension D) and $O_1=(H-\mathrm{Tr}[H]/D)/\sqrt{D\,\mathrm{Cov}(H,H)}$. In addition, $\mathrm{Cov}(X,Y)\equiv\mathrm{Tr}[XY]/D-\mathrm{Tr}[X]\,\mathrm{Tr}[Y]/D^2$. In terms of the $\{x_i\}$ variables we have $\text{đ}\varrho^{(\mathrm{ev})}=[D\,\mathrm{Cov}(H,H)]^{-1/2}\text{đ}\mathbb{Q}\,H+\sum_{i\geqslant 2}\text{đ}x_i\,O_i$, where $\text{đ}x_i=\mathrm{Tr}[\text{đ}\varrho^{(\mathrm{ev})}\,O_i]$. As a result, we obtain $dS=\beta\text{đ}\mathbb{Q}+\sum_{i\geqslant 2}\mathrm{Tr}[O_i\mathbb{H}]\,\text{đ}x_i$, whence

$$\text{đ}\mathbb{S}=\sum_{i\geqslant 2}\mathrm{Tr}[O_i\mathbb{H}]\,\text{đ}x_i, \tag{19}$$

which is indeed independent of the choice of $\{x_i\}$ [60]. We observe that $\text{đ}\mathbb{S}$ is determined only in terms of the system variables, independent of the existence of an environment in a thermal equilibrium, and the system Hamiltonian does not play any explicit role therein; it is an irrelevant observable—see Supplemental Material [60] for further discussion. We remark that, besides using $\text{đ}\mathbb{Q}$ in our definition of irreversible entropy production (17), unlike the earlier literature [61] here β is associated with the system (not with a large environment or heat bath). A discussion of why β is associated with the system can be found in Supplemental Material [60]. Another alternative expression for $\text{đ}\mathbb{S}$ can be obtained in terms of the relative entropy $S(\varrho\|\varrho_{\mathrm{eq}})=\mathrm{Tr}[\varrho\ln\varrho-\varrho\ln\varrho_{\mathrm{eq}}]$ between the state ϱ and the instantaneous canonical Gibbs state $\varrho_{\mathrm{eq}}\equiv e^{-\beta H}/\mathrm{Tr}[e^{-\beta H}]$ as [60] $\text{đ}\mathbb{S}=-dS(\varrho\|\varrho_{\mathrm{eq}})+\beta\text{đ}\mathbb{W}_{\mathrm{CD}}+\mathrm{Tr}[(\varrho-\varrho_{\mathrm{eq}})d(\beta H)]$. We also note that interestingly, from Eq. (19) an inequality can also be derived as [60]

$$\text{đ}\mathbb{S} + b(\mathbb{O})\,\text{đ}B \geqslant 0, \tag{20}$$

which is reminiscent of a generalized Clausius inequality [62,63] with all quantities depending explicitly on the system. Here, $b(\mathbb{O})$ is the spread of $\mathbb{O} = \sum_{i\geqslant 2} O_i \otimes O_i$ and $\text{đ}B = -\sum_k \ln r_k \sum_{k'|dr_{k'}\geqslant 0} dr_{k'}$.

For dynamically *closed quantum systems* (where $\dot{\varrho} = -i[H, \varrho]$, assuming $\hbar \equiv 1$), we have $\dot{r}_k = 0\,\forall k$; hence $dU = \text{đ}\mathbb{W} = \text{đ}W$ and $\text{đ}\mathbb{S} = dS = \text{đ}Q = \text{đ}\mathbb{Q} = \text{đ}\mathbb{W}_{\text{CD}} = 0$. This case also naturally includes phase-space-preserving cooling processes [64]. For dynamically *open quantum systems* weakly coupled to a large environment, when H is constant or slowly driven, the dynamics obeys a Markovian Lindblad master equation $\dot{\varrho} = \mathcal{L}[\varrho]$, where $\mathcal{L}[\varrho] = -i[H + H_{\text{Lamb}}, \varrho] + \mathcal{D}[\varrho]$, with H_{Lamb} being the environment-induced Lamb-shift correction and $\mathcal{D}[\varrho] = \sum_\alpha c_\alpha (L_\alpha \varrho L_\alpha^\dagger - (1/2)\{L_\alpha^\dagger L_\alpha, \varrho\})$ $(c_\alpha > 0)$ being the quantum dissipator [55,56] [cf. Eq. (8)]. Additionally, if the system starts at $\varrho_{\text{eq}}(0)$ and also $\varrho_{\text{eq}}(t)$ is the unique instantaneous steady state of the dynamics [65,66], one can prove that $\varrho(t) \approx \varrho_{\text{eq}}(t)\,\forall t$ [67], namely, the trajectory remains near the steady state, and also $dS - \beta\,\text{đ}Q \geqslant 0$ [10,21,33]. This implies that $\text{đ}\mathbb{S} - \beta\,\text{đ}\mathbb{W}_{\text{CD}} \geqslant 0$, where $\text{đ}\mathbb{W}_{\text{CD}}$ [Eq. (13)] is an energetic cost associated with how different the real trajectory is from the quasistatic one—compare with the general relation (20). This relation can be considered as a manifestation of the Clausius inequality [62,63]. Note that in general, $\text{đ}\mathbb{W} \neq \text{đ}W$ and $\text{đ}\mathbb{Q} \neq \text{đ}Q = \text{Tr}[\mathcal{D}[\varrho]H]dt$ [cf. Eq. (11)]; whereas in the *quasistatic* regime $\varrho(t) = \varrho_{\text{eq}}(t)$, we have $|r_k\rangle = |E_k\rangle$, $\mathbb{H} = \beta H$, and thus $\text{đ}\mathbb{W}_{\text{CD}} = 0$, $\text{đ}\mathbb{Q} = \text{đ}Q$, $\text{đ}\mathbb{S} = 0$, and $dS = \beta\text{đ}Q$. The latter is a manifestation of the thermodynamic *reversibility* [49].

Example I: A qubit in a Markovian environment. Consider a qubit with $H = \omega_0\sigma_z$ that weakly interacts with a Markovian environment such that $\dot{\varrho} = -i[\omega_0\sigma_z, \varrho] + \gamma(\sigma_x\varrho\sigma_x - \varrho)$, where ω_0 and γ are positive constants and $\sigma_x = |0\rangle\langle 1| + |1\rangle\langle 0|$ and $\sigma_z = |0\rangle\langle 0| - |1\rangle\langle 1|$ are the x and z Pauli operators [55]. The quantity $\dot{U} = \text{Tr}[H\dot{\varrho}] = -2\gamma\omega_0\text{Tr}[\sigma_z\varrho]$ is fully due to the dissipative part of the dynamics. We consider three different initial states (for details, see Supplemental Material [60]). (i) The first is the thermal state $\varrho_{\text{eq}}(0)$, with $\beta(0)$ being the initial inverse temperature of the system (this can differ from that of the environment, and the latter need not be in a thermal state). In this case, we obtain $\dot{\mathbb{Q}} = \dot{U} = 2\gamma\omega_0 e^{-2\gamma t}\tanh[\beta(0)\omega_0]$, $\dot{q} = \dot{\mathbb{Q}}$, and $\dot{w} = \dot{\mathbb{W}}$. (ii) The second is the pure state $|\psi(0)\rangle = (|0\rangle + |1\rangle)/\sqrt{2}$. One can see that $\dot{U} = \dot{\mathbb{Q}} = \dot{\mathbb{W}} = \dot{q} = \dot{w} = 0$. (iii) Starting from $\varrho(0) = (\mathbb{1} + [\sigma_x + \sigma_z]/2)/2$, we can obtain the heat, work, and internal energy change in the system (Fig. 1). It can be shown that $\dot{q} = \dot{U} = -(\gamma/2)e^{-2\gamma t}$ and $\dot{w} = 0$.

Example II: A qubit in a dephasing environment. Consider the dephasing master equation for a qubit, $\dot{\varrho} = -i[\omega_0\sigma_z, \varrho] + \gamma(\sigma_z\varrho\sigma_z - \varrho)$. Generally, in this process the system energy is preserved, and there is no net energy exchange between the system and the environment. However, the system and the environment can still exchange heat and work. This can be contrasted with Landauer's principle, where the environment does work to erase phase information and the system releases heat back into the environment. Let $\omega_0 = 1$ and $\gamma = 0.1$ and consider the following two different cases. (i) The initial state is $|\psi(0)\rangle = (|0\rangle + \sqrt{2}|1\rangle)/\sqrt{3}$: We obtain that $\dot{\mathbb{Q}} = -\dot{\mathbb{W}}_{\text{CD}} = (8/15)/[8 + e^{2t/5}]$ and $\dot{S} = 4\ln[(1 + 2\Delta)/(1 - 2\Delta)]/(15e^{t/5}\sqrt{8 + e^{2t/5}})$, where $\Delta = (1/6)e^{-t/5}\sqrt{8 + e^{2t/5}}$. This process can be compared with an isothermal process in ideal gases, where the energy is constant and the heat exchanged is equal to the work exchanged with the opposite sign. However, since this dynamics does not exactly model an ideal gas (its internal energy is not proportional to its temperature), β varies and thus the process is not isothermal. (ii) The initial state is $|\psi(0)\rangle = (|0\rangle + |1\rangle)/\sqrt{2}$: In this case, $dU = \text{đ}\mathbb{Q} = \text{đ}\mathbb{W}_{\text{CD}} = 0$. However, $dS \neq 0$, which means that the whole entropy change is due to the irreversible entropy production. This can be compared with a free expansion process in classical (nonideal) gases. For a further example of a driven qubit, see Ref. [68].

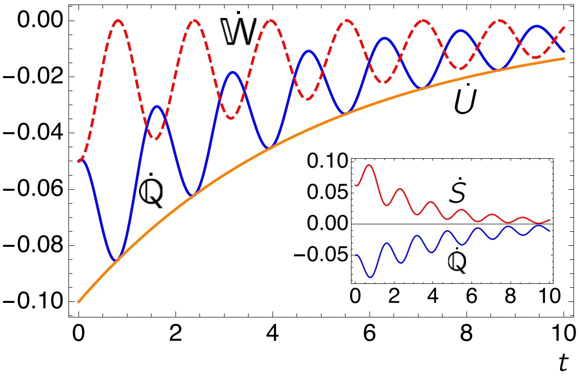

FIG. 1. Rates of internal energy (solid orange curve), heat (solid blue curve), and work (dashed red curve) changes vs time (in natural units $\hbar \equiv k_B \equiv 1$), for a qubit in a heat bath as given in example I, with $\gamma = 0.1$ and $\omega_0 = 1$. The integrated values are $U = -0.25$, $\mathbb{Q} = -0.138$, and $\mathbb{W} = -0.112$, respectively, where $X = \int_0^{10} dt\,\dot{X}$ with $X \in \{U, \mathbb{Q}, \mathbb{W}\}$. Inset compares $\dot{S}$ and $\dot{\mathbb{Q}}$.

Another example, a quantum damped harmonic oscillator in an environment of oscillators, has been worked out in Supplemental Material [60]. It appears that at all times the state of the oscillator is a Gibbs state with a time-dependent inverse temperature and a constant Hamiltonian. Because only the eigenvalues of the state change in time and the eigenvectors remain constant, $\text{đ}\mathbb{W}_{\text{CD}}$ vanishes. Hence heat and work here reduce to the values obtained from the conventional definitions.

Summary. We have revisited the assignment of thermodynamic quantities to an open quantum system strongly coupled to an environment. In general, there is no unique way of separating the system internal energy from that of the environment. Despite this fundamental issue, by introducing a dissipative work, we have shown that it is possible to consistently split the internal energy change into work change (causing no entropy change) and heat change (which can cause entropy change). The key ingredient is to use the trajectory-based description of the state of the system and its associated equation of motion, which is universally valid for any coupling strength and yields a spectral decomposition of the system density matrix, separating the changes in the eigenvalues from those of the eigenvectors. We have compared our entropy-based definitions with the conventional and semiclassical ones and have argued that these two approaches are inadequate. More importantly, by using a definition of nonequilibrium temperature in quantum systems, we have obtained the irreversible entropy production in a general dynamical process. We have demonstrated that the irreversible entropy production is a function of the system variables other than the energy and does not have any explicit dependence on the environmental degrees of freedom. We have also derived an inequality which can be contrasted with the generalized Clausius inequality. The consistency of our formalism has been illustrated in paradigmatic scenarios.

Note added. Recently, we learned about another independent related study [69].

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