Work Sum Rule for Open Quantum Systems

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A key question in the thermodynamics of open quantum systems is how to partition thermodynamic quantities such as entropy, work, and internal energy between the system and its environment. We show that the only partition under which entropy is nonsingular is based on a partition of Hilbert space, which assigns half the system-environment coupling to the system and half to the environment. However, quantum work partitions nontrivially under Hilbert-space partition, and we derive a work sum rule that accounts for quantum work at a distance. All state functions of the system are shown to be path independent once this nonlocal quantum work is properly accounted for. Our results are illustrated with application to a driven resonant level strongly coupled to a reservoir.

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The program of scaling the laws of thermodynamics down to the nanoscale and beyond has proven to be exceptionally challenging. While substantial progress has been made [\[1](#page-4-0)–[21](#page-5-0)], there exists little consensus on some of the most foundational questions, especially in the extreme quantum limit. Formulating the First Law of Thermodynamics for open quantum systems begs the question of how to partition quantum observables between the system and its environment. Various competing schemes have been proposed in the literature [[22](#page-5-1)–[29\]](#page-5-2), many of which involve assigning part of the interfacial energy to the system and the remainder to the environment.

In this Letter, we analyze the thermodynamics of a quasistatically driven open quantum system strongly coupled to its environment, and show that entropy is only well-defined under a Hilbert-space partition, which divides the system-environment coupling equally between the system and the environment. Internal energy, chemical work, and entropy are partitioned straightforwardly under a partition of Hilbert space. However, the partition of quantum work is nontrivial in this framework. We therefore derive a work sum rule that accounts for the thermodynamic effect of quantum work at a distance. All state functions of the system are shown to be path independent once this nonlocal quantum work is properly accounted for.

The internal energy $U(t)$ of the Universe (consisting of system + environment) with Hamiltonian $H(t)$ is

$$
U(t) := \langle H(t) \rangle, \tag{1}
$$

where $\langle \rangle$ denotes the quantum statistical average $\langle H(t) \rangle = \text{Tr}\{H(t)\rho(t)\}\,$, where $\rho(t)$ is the density matrix of the universe at time t and $Tr\{\}$ denotes the trace over the full Fock space. The (inclusive) rate of work done by external forces on the Universe is [[5](#page-4-1),[26](#page-5-3),[30](#page-5-4)]

$$
\dot{W}_{\text{ext}}(t) := \frac{d}{dt} \langle H(t) \rangle = \langle \dot{H}(t) \rangle, \tag{2}
$$

where the second equality follows from the von Neumann equation for the density matrix (see Sec. I in [[31](#page-5-5)]).

In this Letter, we consider a time-dependent quantum universe of fermions without interparticle interactions. In that case, the internal energy and power delivered can be expressed in terms of the single-particle Green's function $G[<](t, t')$ as

$$
U(t) = -i \mathbb{Tr} \{ h(t) G^{<}(t, t) \}, \tag{3}
$$

and

$$
\dot{W}_{\text{ext}}(t) = -i \mathbb{T} \mathbb{r} \{ \dot{h}(t) G^{<}(t, t) \},\tag{4}
$$

respectively, where $h(t) = h_S(t) + h_R + h_{SR}(t)$ is the matrix representation of $H(t)$ in the one-body Hilbertspace, and Tr denotes a trace over this Hilbert space (see Sec. II A in [[31](#page-5-5)] for the definition and evaluation of $G^<$). Here, $h_S(t)$ is the driven Hamiltonian of the quantum subsystem of interest, h_R is the Hamiltonian of the macroscopic reservoir, and $h_{SR}(t)$ is the coupling Hamiltonian.

We consider a quantum system that can exchange energy and particles with a reservoir in equilibrium at temperature T and chemical potential μ . The system is driven quasistatically so that it remains in equilibrium with the reservoir throughout the driving protocol. Under these conditions, the equal-time Green's function takes the quasiequilibrium form

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$$
G^{<(0)}(t,t) = \int d\epsilon f(\epsilon) A^{(0)}(t,\epsilon), \tag{5}
$$

where $A^{(0)}(t, \epsilon) = \delta[\epsilon - h(t)]$ is the spectral function in the quasistatic limit (see Sec. II Λ in [31]) and $f(c) = (1 +$ quasistatic limit (see Sec. II A in [[31\]](#page-5-5)) and $f(\epsilon) = (1 + \epsilon)$ $e^{\beta(e-\mu)}$ ⁻¹ is the Fermi-Dirac distribution. Here, the super-
script $\Lambda^{(n)}$ denotes the order in time derivatives of the driving script $A^{(n)}$ denotes the order in time derivatives of the driving Hamiltonian. We note that Eq. [\(5\)](#page-0-4) is exact to all orders in the system-reservoir coupling $h_{SR}(t)$, but omits terms involving $\dot{h}(t)$, $\ddot{h}(t)$, etc.

The relevant thermodynamic quantities for such a universe are evaluated as follows (see Sec. II B in [[31](#page-5-5)]). The internal energy is given by

$$
U^{(0)}(t) = \int d\varepsilon \, g(t,\varepsilon) f(\varepsilon) \varepsilon,\tag{6}
$$

where $g(t, \epsilon) = \mathbb{T} \{A^{(0)}(t, \epsilon)\}\$ is the quasistatic density of states. Similarly, the quasistatic power delivered may be states. Similarly, the quasistatic power delivered may be computed using Eqs. [\(4\)](#page-0-5) and [\(5\)](#page-0-4) as

$$
\dot{W}_{\text{ext}}^{(1)}(t) = \int d\epsilon f(\epsilon) \text{Tr}\{\dot{h}(t)A^{(0)}(t,\epsilon)\}.
$$
 (7)

The quasistatic entropy is given by

$$
\langle S(t) \rangle = S^{(0)}(t) = \int d\varepsilon \, g(t, \varepsilon) s(\varepsilon), \tag{8}
$$

where the entropy operator $S(t) = -\ln \rho(t)$, $s(\epsilon) =$ $\beta(\epsilon - \mu)f(\epsilon) + \ln(1 + e^{-\beta(\epsilon - \mu)})$, and we set $k_B = 1$. The mean number of particles is given by mean number of particles is given by

$$
N^{(0)}(t) = \int d\varepsilon \, g(t, \varepsilon) f(\varepsilon). \tag{9}
$$

The grand canonical potential of the Universe is $\Omega^{(0)}(t) = U^{(0)}(t) - TS^{(0)}(t) - \mu N^{(0)}(t)$, which can be
expressed as expressed as

$$
\Omega^{(0)}(t) = \int d\varepsilon \, g(t, \varepsilon) \omega(\varepsilon), \tag{10}
$$

where $\omega(\epsilon) = -(1/\beta) \ln(1 + e^{-\beta(\epsilon-\mu)})$.
The first variations of the thermo-

The first variations of the thermodynamic quantities satisfy $\dot{\Omega}^{(1)}(t) = \dot{U}^{(1)}(t) - T\dot{S}^{(1)}(t) - \mu \dot{N}^{(1)}(t)$. Importantly,
the first variation of O is equal to the external work the first variation of Ω is equal to the external work

$$
\dot{W}_{\text{ext}}^{(1)}(t) = \dot{\Omega}^{(1)}(t) \tag{11}
$$

(see Secs. III A and III B in [[31](#page-5-5)] for derivations).

A central quantity in scanning probe microscopy is the local density of states $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$ $[8-10,12,13,16,34-45]$, which provides a firm experimental basis to construct local thermodynamic state functions in real quantum systems. The local density of states of subsystem γ is defined as [\[46,](#page-6-0)[47\]](#page-6-1) $g_{\gamma}(t, \epsilon) := \mathbb{T}_{\mathbb{F}}\{\Pi_{\gamma}\delta[\epsilon - h(t)]\}$, where $\Pi_{\gamma} = \int_{x \in \gamma} dx |x\rangle\langle x|$ is
the prejection operator onto subgress μ of the single the projection operator onto subspace γ of the singleparticle Hilbert-space. $g_{\gamma}(t, \epsilon)$ gives the *local spectrum* of the nonlocal operator $h(t)$.

The partitioned thermodynamic quantities $U_{\gamma}(t)$, $S_{\gamma}(t)$, $N_{\gamma}(t)$, and $\Omega_{\gamma}(t)$ are defined [[48](#page-6-2)–[50](#page-6-3)] by simply replacing $g(t, \epsilon)$ by $g_{\gamma}(t, \epsilon)$ in Eqs. [\(6\),](#page-1-0) [\(8\)](#page-1-1), [\(9\),](#page-1-2) and [\(10\),](#page-1-3) and are the quantum statistical averages of partitioned quantum observables $H|_{\gamma}$, $S|_{\gamma}$, and $N|_{\gamma}$, respectively, where $O|_{\gamma}$ is the Fockspace operator corresponding to the following operator defined on the single-particle Hilbert space [\[51\]](#page-6-4) (see Sec. IV in [\[31\]](#page-5-5))

$$
o|_{\gamma} = \frac{1}{2} \{ \Pi_{\gamma}, o \},
$$
 (12)

where σ is the single-particle Hilbert-space operator corresponding to the global Fock-space operator $O = \sum_{\gamma} O_{\gamma}$, and the anticommutator (defined as ${a, b} = ab + ba$) ensures the Hermiticity of $O|_{\gamma}$.

In analogy with Eq. (11) , we define the *rate of thermo*dynamic work done on subsystem γ as $\dot{W}^{(1)}_Y(t) := \dot{\Omega}^{(1)}_Y(t)$,
leading to the First I aw of Thermodynamics for a quantum leading to the First Law of Thermodynamics for a quantum subsystem $U^{(1)}_Y(t) = T\dot{S}^{(1)}_Y(t) + \mu \dot{N}^{(1)}_Y(t) + \dot{W}^{(1)}_Y(t)$. However, in general, $\dot{W}_{\gamma}^{(1)}(t) \neq \langle \dot{H} |_{\gamma}(t) \rangle$, so that the rate of thermodynamic work done on a given subsystem is not thermodynamic work done on a given subsystem is not equal to the expectation value of the power operator partitioned on that subsystem. Instead,

$$
\dot{W}_{\gamma}^{(1)}(t) \coloneqq \dot{\Omega}_{\gamma}^{(1)}(t) = \langle \dot{H} |_{\gamma}(t) \rangle + I_{\gamma}^{W}(t), \tag{13}
$$

where $I_Y^W(t)$ represents the instantaneous quantum flow of
free energy into subsystem x induced by the external drive free energy into subsystem γ induced by the external drive $H(t)$. $I_Y^W(t)$ can be thought of as (the rate of) quantum work
at a distance. Quantum work is inherently poplocal because at a distance. Quantum work is inherently nonlocal because even if the external drive is local, the quantum states acted upon are nonlocal. This nonlocal work predicted in driven quantum systems is reminiscent of the phenomena of measurement-induced energy teleportation [[52](#page-6-5),[53](#page-6-6)] or conditional work at a distance [\[54\]](#page-6-7) in autonomous quantum systems.

The Hamiltonian of the open quantum system is $H(t)$ = $H_S(t) + H_R + H_{SR}(t)$, where $H_S(t) = \sum_{n,m} [h_S(t)]_{nm} d_n^{\dagger} d_m$ is the system Hamiltonian and $H_R = \sum_k \epsilon_k c_k^{\dagger} c_k$ is the reservoir Hamiltonian. Finally, the coupling Hamiltonian reservoir Hamiltonian. Finally, the coupling Hamiltonian $H_{SR}(t)$, describing the interface between the system and the reservoir, is $H_{SR}(t) = \sum_{k,n} [V_{kn}(t) c_{k}^{\dagger} d_n + \text{H.c.}]$. Definition [\(12\)](#page-1-5) implies that the Hamiltonian partitioned on the system is

$$
H|_S(t) = H_S(t) + \frac{1}{2} H_{SR}(t),
$$
\n(14)

so that the coupling Hamiltonian is partitioned equally between the system and the reservoir.

The following sum rule for external work can be derived for open systems (see Sec. V in [[31](#page-5-5)] for a derivation)

$$
\dot{W}_{\text{ext}}^{(1)}(t) = \dot{\Omega}_S^{(1)}(t) + \delta \dot{\Omega}_R^{(1)}(t), \tag{15}
$$

where $\delta\Omega_R = \Omega_R - \Omega_{R,0}$ is the change in the reservoir grand potential induced due to the change in its spectrum as a result of its coupling to the system. A similar construction was introduced by Friedel [[55](#page-6-8)] to describe the cloud of screening charge induced in a metal due to the presence of an impurity.

An interesting and important case to consider is that where only the system is time-dependent so that $H|_{S}(t) = H_{S}(t)$ and $\dot{H}|_R(t) = 0$. Nonetheless, generically $\delta \dot{\Omega}_R(t) \neq 0$, so the work sum rule becomes

$$
\langle \dot{H}_S(t) \rangle \stackrel{\dot{H}_{SR}=0}{=} \dot{\Omega}_S^{(1)}(t) + \delta \dot{\Omega}_R^{(1)}(t). \tag{16}
$$

Thus, even if external forces act only inside the system, the instantaneous thermodynamic work done on the reservoir is nonzero. For this case, Eqs. [\(13\)](#page-1-6) and [\(16\)](#page-2-0) imply that the rate of nonlocal quantum work done on the system is $I_S^{W \dot{H}_{\text{S R}}=0} - \delta \dot{\Omega}_R^{(1)}$, or minus the rate of nonlocal quantum work done on the reservoir.

Using the internal energy $U_S(t) = \langle H|_S(t) \rangle$, the First Law for the open system becomes

$$
\frac{d}{dt}\langle H_S(t) + \frac{1}{2}H_{SR}(t)\rangle = T\dot{S}_S^{(1)} + \mu \dot{N}_S^{(1)} + \dot{W}_S^{(1)}, \quad (17)
$$

where S_S is the entropy partitioned on the system Hilbert space, and

$$
\dot{W}_S^{(1)} = \dot{W}_{\text{ext}}^{(1)} - \delta \dot{\Omega}_R^{(1)} = \langle \dot{H} |_{S} \rangle + I_S^W \tag{18}
$$

is the rate of thermodynamic work done on the system.

Using the nonequilibrium Green's function (NEGF) formalism [\[56](#page-6-9)–[58](#page-6-10)], each term in the First Law [Eq. [\(17\)\]](#page-2-1) can be expressed in terms of the quasistatic system Green's functions (see Sec. V in [[31](#page-5-5)] for derivations). The terms contributing to the (partitioned) quasistatic power [Eq. [\(7\)](#page-1-7)] delivered by external forces are

$$
\langle \dot{H}_S(t) \rangle = \int \frac{d\epsilon}{\pi} f(\epsilon) \mathbb{I} \mathfrak{m} \mathbb{Tr} \{ \dot{h}_S(t) \mathcal{G}^{A(0)}(t, \epsilon) \}, \qquad (19)
$$

$$
\langle \dot{H}_{SR}(t) \rangle = \int \frac{d\epsilon}{\pi} f(\epsilon) \mathbb{I} \mathbb{m} \mathbb{Tr} \{ \dot{\Sigma}^A(t, \epsilon) \mathcal{G}^{A(0)}(t, \epsilon) \}, \quad (20)
$$

where $G^{A(0)}$ and Σ^A are the quasistatic advanced system Green's function and self-energy, respectively, defined in Sec. II in [[31](#page-5-5)]. The rate of nonlocal work done on the system is

$$
I_S^W(t) = -\frac{1}{2} \langle \dot{H}_{SR}(t) \rangle + \int \frac{d\epsilon}{\pi} \text{Im} \mathbb{Tr} \left\{ \frac{\partial \mathcal{G}^{A(0)}(t, \epsilon)}{\partial t} \frac{\partial \Sigma^A(t, \epsilon)}{\partial \epsilon} - \frac{\partial \mathcal{G}^{A(0)}(t, \epsilon)}{\partial \epsilon} \frac{\partial \Sigma^A(t, \epsilon)}{\partial t} \right\} \omega(\epsilon), \tag{21}
$$

where the second term on the rhs may be interpreted as an instantaneous flow of free energy into the system induced by the time-dependent external drive, while the nonlocality of the first term on the rhs is trivial since H_{SR} is itself nonlocal.

We note that in the broadband limit $[\partial_{\epsilon} \Sigma^{A}(t, \epsilon) = 0]$ the rate of nonlocal quantum work vanishes, in contrast to the notion proposed in Ref. [\[25\]](#page-5-12) and discussed in Ref. [\[29\]](#page-5-2) (see Sec. VI in [[31](#page-5-5)]).

The work sum rule and Hilbert-space partition of the thermodynamics derived above are illustrated with an application to the driven resonant-level model. (See Sec. VIII in [\[31\]](#page-5-5) for an extension to multiple reservoirs and analysis of a driven two-level system coupled to two reservoirs.) The system Hamiltonian is $H_S(t) = \varepsilon_s(t) d^{\dagger} d$, the reservoir is modeled as a semi-infinite tight-binding chain with hopping integral t_0 , $H_R = t_0 \sum_{j=1}^{\infty} (c_j^{\dagger} c_{j+1} + \text{H.c.})$, and the inter-
face between system and reservoir is modeled by $H_{SR}(t) = V(t)d^{\dagger}c_1$ + H.c. Two driving protocols are investigated: in *protocol 1*, the level $\varepsilon_s(t)$ is varied while V is held fixed. In *protocol* 2, both $\varepsilon_s(t)$ and $V(t)$ are varied.

A verification of the work sum rule for protocol 1 [Eq. [\(16\)\]](#page-2-0) is shown in Fig. [1](#page-2-2), where the nonlocal quantum work on the reservoir is clearly visible as the difference between $\Delta\Omega_s$ (dashed blue curve) and the total work done W_{ext} (green dots). The nonlocal quantum work done on the reservoir may be positive [Fig. [1\(a\)\]](#page-2-2) or negative [Fig. [1\(b\)\]](#page-2-2) depending on system parameters, and increases in magnitude as the coupling V between the system and reservoir increases.

FIG. 1. Verification of the work sum rule [Eqs. [\(15\)](#page-2-3), [\(16\)](#page-2-0)] for an open quantum system: the resonant-level model. Only the level $\varepsilon_s(t)$ is driven (a) from 1 to 1.5, and (b) from -1.5 to -1. The reservoir is maintained at temperature $T = 0.02$ and chemical potential $\mu = 0$, with $t_0 = 1.25$.

Let us compare our analysis of the thermodynamics of a quasistatically driven open quantum system with some previous frameworks found in the literature [[22](#page-5-1)–[25,](#page-5-12)[27](#page-5-13)]. These frameworks can be described as α partitions of the internal energy, where $\alpha \in [0, 1]$ describes the fraction of
the coupling Hamiltonian H_{on} included in the internal the coupling Hamiltonian H_{SR} included in the internal energy of the open system. In the α partition, the internal energy of the open quantum system is [\[59\]](#page-6-11)

$$
\alpha \cdot U_S(t) = \langle H_S(t) + \alpha H_{SR}(t) \rangle, \tag{22}
$$

and the rate of external work done on the system is identified by some authors as [[23](#page-5-14)]

$$
\alpha \cdot \dot{W}_S(t) = \langle \dot{H}_S(t) + \alpha \dot{H}_{SR}(t) \rangle. \tag{23}
$$

The Hilbert-space partition proposed in this Letter corresponds to setting $\alpha = 1/2$ in Eq. [\(22\)](#page-3-0). However, as discussed above, the rate of external work done on the system is given by Eq. [\(18\),](#page-2-4) and cannot in general be expressed as in Eq. [\(23\)](#page-3-1) for any value of α .

For independent quantum particles, the entropy operator $-\ln \hat{\rho}^{(0)}$ under quasistatic driving is also a one-body observable, and can be partitioned in the same way [[60](#page-6-12)]. One finds for the α partition of the entropy of subsystem γ (see Sec. VII in [[31](#page-5-5)])

$$
\alpha \cdot S_{\gamma}^{(0)}(t) = 2\alpha \int d\epsilon \, g_{\gamma}(t,\epsilon) s(\epsilon)
$$

$$
+ (1 - 2\alpha) \int d\epsilon \int d\epsilon' \,\mathbb{Tr}\{\tilde{A}_{\gamma}(t,\epsilon)\tilde{A}_{\gamma}(t,\epsilon')\}
$$

$$
\times \{-f(\epsilon) \ln[f(\epsilon')] - [1 - f(\epsilon)] \ln[1 - f(\epsilon')] \},
$$
(24)

where $\tilde{A}_{\gamma}(t, \epsilon) = \Pi_{\gamma} \delta[\epsilon - h(t)] \Pi_{\gamma}$.
Although it might appear that

Although it might appear that one could construct the partitioned thermodynamics for arbitrary values of α , a severe problem arises in α -S_γ if $\alpha \neq 1/2$. For $\alpha > 1/2$, the term beginning on the second line Eq. [\(24\)](#page-3-2) is negative and unbounded, while for $\alpha < 1/2$, the partitioned entropy of the subspace complementary to γ is negative and unbounded. Since entropy is a non-negative quantity, any partition that yields a negative subsystem entropy should be ruled out on principle as unphysical. Moreover, in fermionic systems there are generically tightly bound core states with occupancy $f \to 1$ and high-lying scattering states with occupancy $f \to 0$, for both of which Eq. [\(24\)](#page-3-2) is undefined for $\alpha \neq 1/2$. Thus, we are forced to conclude that the only physically allowable partitioning of the entropy is $\alpha = 1/2$, namely, the Hilbert-space partition as previously proposed in Ref. [\[49\]](#page-6-13).

Figure $2(a)$ shows a comparison of the small physical value of the system entropy for $\alpha = 1/2$ and the large (positive or negative) unphysical entropy for $\alpha \neq 1/2$ for

FIG. 2. Results for the α partition of the entropy [Eq. [\(24\)\]](#page-3-2) in the resonant-level model with $\mu = 0$, $V = 1$, and $t_0 = 1.25$. (a) Comparison of α -S_S to the entropy under Hilbert-space partition ($\alpha = 1/2$) at $T = 0.02$. (b) Coefficient of $(1 - 2\alpha)$ in Eq. [\(24\)](#page-3-2) (second term on the rhs) versus inverse temperature for various values of the level energy $\varepsilon_s = 0, \pm 1, \pm 2$.

the resonant-level model with $\varepsilon_s = -1$, $V = 1$, $\mu = 0$, and $T = 0.02$. Under Hilbert-space partition, the system entropy is bounded by $0 \leq S_s \leq \ln 2$. Note that the slope of the green curve representing α -S_S tends to $-\infty$ as $T \to 0$, indicating a severe contradiction of the Third Law of Thermodynamics for $\alpha \neq 1/2$, as pointed out previously in Refs. [\[23](#page-5-14)[,49\]](#page-6-13). Figure [2\(b\)](#page-3-3) plots the coefficient of $(1 - 2\alpha)$ in Eq. [\(24\)](#page-3-2) (second term on the rhs) as a function of inverse temperature at $\mu = 0$ for several values of the resonant level ε_s , indicating that this unphysical contribution to the entropy partition diverges α 1/T as $T \to 0$, as can be readily understood from Eq. [\(24\)](#page-3-2) and the functional form of the Fermi-Dirac distribution.

Figure [3\(a\)](#page-4-3) plots W_S [defined in Eq. [\(18\)](#page-2-4)] and $\alpha-W_S$ [defined in Eq. [\(23\)](#page-3-1)] for two different paths in protocol 2 (see inset) as a function of α . The quasistatic work for a system in the grand canonical ensemble at constant T and μ should be independent of path. It is clear that α -W_S, the definition of the work done on the system proposed in Ref. [[23](#page-5-14)], is only path independent in the limit $\alpha \rightarrow 1$, wherein $\lim_{\alpha \to 1} \alpha W_s = W_{ext}$, which unsurprisingly is indeed path independent. However, our definition [Eq. [\(18\)\]](#page-2-4) of the work W_S done on the Hilbert space of the system, including nonlocal quantum work, is path independent and generally not equal to the total external work W_{ext} for nonzero system-reservoir coupling.

Using their definitions of internal energy [Eq. [\(22\)](#page-3-0)] and of work done on the system [Eq. [\(23\)](#page-3-1)], Esposito, Ochoa, and Galperin [\[23\]](#page-5-14) use the thermodynamic identity

$$
T\Delta S_S^{\text{EOG}} = \Delta(\alpha - U_S) - \mu \Delta N_S - \alpha - W_S \tag{25}
$$

to define the change in system entropy $\Delta S_S^{\rm EOG}$ for a process. ΔS_S^{EOG} is plotted in Fig. [3\(b\)](#page-4-3) as a function of α for two different paths in protocol 2. Also plotted is the change in

FIG. 3. α partition of the thermodynamics in the resonant-level model, where both the level $\varepsilon_s(t)$: $0 \to 1$ and coupling $V(t)$: $0.6 \to$ 0.4 are driven along two different paths [see protocols in inset (a)]. Here, $T = 0.02$, $\mu = 0$, and $t_0 = 1.25$. (a) Two definitions of work: W_S [defined in Eq. [\(18\)](#page-2-4)] and α - W_S [defined in Eq. [\(23\)](#page-3-1)] versus α . (b) The change in the statistical mechanical partition of the entropy $\Delta(\alpha-S_S)$ [Eq. [\(24\)](#page-3-2)] and ΔS_S^{EOG} [Eq. [\(25\)](#page-3-4)], proposed in Ref. [\[23\]](#page-5-14),
versus α for the same processes. The bound $|\Delta S_{\alpha}| < \ln 2$ under versus α for the same processes. The bound $|\Delta S_S| \leq \ln 2$ under Hilbert-space partition is shown as red dashed lines.

 α -S_S, the α partition of statistical mechanical entropy [Eq. [\(24\)](#page-3-2)]. Although our model is slightly different than that used in Ref. [\[23\]](#page-5-14) (we utilize a semi-infinite 1D tightbinding model of the reservoir while Ref. [[23](#page-5-14)] uses a phenomenological self-energy), the quantitative results for the two models are comparable (see Sec. II A in [\[31\]](#page-5-5)).

As shown in Fig. [3\(b\)](#page-4-3) and in Ref. [\[23\]](#page-5-14), ΔS_S^{EOG} is path independent only for $\alpha = 1$. Based on this fact, Esposito, Ochoa, and Galperin suggest that $\alpha = 1$ should be chosen in the partition of the internal energy [Eq. [\(22\)\]](#page-3-0), despite the acknowledged violation of the Third Law of Thermodynamics for $\alpha = 1$. It is noteworthy that the magnitude of ΔS_S^{EOG} greatly exceeds ln2 [see red dashed lines in Fig. [3\(b\)](#page-4-3)], the maximum entropy of the system under Hilbert-space partition. In fact, $\Delta S_{S}^{EO}|_{\alpha=1} = 5.57$ is
opposite in sign and 80.9 times larger in magnitude than the opposite in sign and 80.9 times larger in magnitude than the actual entropy change $\Delta S_s = -0.068$ for the process shown in Fig. [3.](#page-4-3) We would argue that there is no room in the Hilbert space of the system for so much entropy, no matter how the level is broadened and shifted due to a finite coupling to the reservoir.

The problem with the entropy [[23](#page-5-14)] defined thermodynamically via Eq. [\(25\),](#page-3-4) which is not equal to the statistical mechanical partition α -S_S for any value of α , stems from the incorrect definition [Eq. [\(23\)](#page-3-1)] of the work done on the system, which does not take into account the nonlocal quantum work $\int I_S^W dt$. Once the correct definition of work [Eq. [\(18\)\]](#page-2-4) is used, the Hilbert-space partition ($\alpha = 1/2$) of the statistical mechanical entropy [Eq. [\(24\)](#page-3-2)] satisfies the First Law [Eq. [\(17\)](#page-2-1)], and is path independent, as shown in Fig. [3\(b\)](#page-4-3).

In this Letter, we have derived a work sum rule describing the thermodynamic effects of nonlocal quantum work. An open quantum system is analogous to a quantum impurity problem, and nonlocal quantum work is a thermodynamic effect analogous to the screening charge that must be included in the Friedel sum rule [\[55\]](#page-6-8). We emphasize that our thermodynamic partition of the entropy is different than the usual information-theoretic partition based on the reduced state of a quantum subsystem [\[61\]](#page-6-14); due to the nonlocality of quantum information, the reducedstate description introduces entanglement entropy that is not associated with any thermodynamic process [[51](#page-6-4)].

The thermodynamic partition proposed in this Letter is based on a partition of Hilbert space, and is consistent with the analysis of Ref. [[22](#page-5-1)], which circumnavigates the issue of partitioning quantum work. Alternative attempts [\[23\]](#page-5-14) to partition the thermodynamics of open quantum systems have failed due to an incorrect partition of quantum work.

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