Quantum Thermodynamics: A Nonequilibrium Green's Function Approach

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We establish the foundations of a nonequilibrium theory of quantum thermodynamics for noninteracting open quantum systems strongly coupled to their reservoirs within the framework of the nonequilibrium Green's functions. The energy of the system and its coupling to the reservoirs are controlled by a slow external time-dependent force treated to first order beyond the quasistatic limit. We derive the four basic laws of thermodynamics and characterize reversible transformations. Stochastic thermodynamics is recovered in the weak coupling limit.

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Nonequilibrium thermodynamics of open quantum systems is a powerful tool for the study of mesoscopic and nanoscale systems. It allows one to reliably assess the performance of energy-converting devices such as thermoelectrics or photoelectrics, by identifying the system entropy production. It enables one to meaningfully compare these different devices by discriminating the system-specific features from the universal ones and to appraise the role of quantum effects. It can also be used to verify the thermodynamic consistency of approximation schemes. Such a theory is nowadays available for systems weakly interacting with their surrounding [1–6], where it has proven very useful [7–15]. However, in case of strong system-reservoir interactions, finding definitions for heat, work, entropy, and entropy production, which satisfy the basic laws of thermodynamics is an open problem. Each proposal has its own limitations [16–23], even at equilibrium [24–30]. Reversible transformations, for instance, are never explicitly characterized. Establishing a consistent nonequilibrium thermodynamics for open quantum systems strongly coupled to their surrounding is therefore an important step towards a more realistic thermodynamic description of mesoscopic and nanoscale devices. It is also essential to improve our understanding of the microscopic foundations of thermodynamics.

In this Letter, we use the nonequilibrium Green's functions (NEGF) to establish a fully consistent nonequilibrium thermodynamic description of a fermionic single quantum level strongly coupled to multiple fermionic reservoirs. A slow time-dependent driving force controls the level energy as well as the system-reservoir interaction. We propose definitions for the particle number, the energy, and the entropy of the system, as well as for entropy production, heat, and work, which give rise to a consistent zeroth, first, second, and third law. These definitions can be seen as energy resolved versions of the weak coupling definitions used in stochastic thermodynamics. An interesting outcome of our approach is that the general form of the energy and particle currents is different from the standard form used in the NEGF and cannot be expressed as an expectation value of operators. We recover the known expressions when considering nonequilibrium steady states (i.e., in absence of driving) or in the weak coupling limit.

The total Hamiltonian that we consider is $\hat{H}(t) = \hat{H}_S(t) + \sum_\nu \hat{H}_\nu + \sum_\nu \hat{V}_\nu(t)$, where ν labels the different fermionic reservoirs (see Fig. 1), $\hat{H}_S(t) = \varepsilon(t)\hat{d}^\dagger\hat{d}$ is the fermionic single level Hamiltonian, $\hat{H}_\nu = \sum_{k\in\nu}\varepsilon_k\hat{c}_k^\dagger\hat{c}_k$ is the reservoir ν Hamiltonian, and $\hat{V}_\nu(t) = \sum_{k\in\nu}(V_k^\nu(t)\hat{d}^\dagger\hat{c}_k + \text{H.c.})$ is the level-reservoir coupling. The time dependence in the system and in the coupling is due to the external time-dependent driving force.

The central object in the NEGF theory is the single particle Green function (GF) [31]

$$G(\tau_1,\tau_2) = -i\langle T_c\hat{d}(\tau_1)\hat{d}^\dagger(\tau_2)\rangle, \tag{1}$$

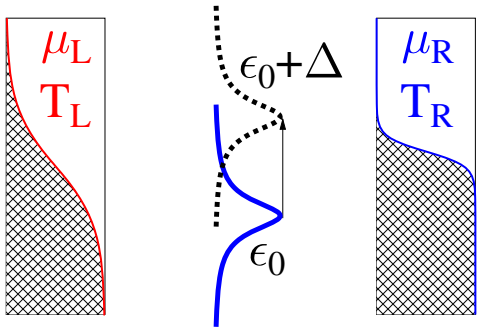

FIG. 1 (color online). Sketch of a fermionic single quantum level junction. The level is broadened by the strong coupling to the reservoirs and is driven by a time-dependent force.

where T_c denotes the contour ordering operator, and τ_1 and τ_2 are the contour variables. Here and below, $\hbar = k_B = 1$. When the time-dependent driving force is slow relative to the system relaxation time, the dynamics of the GF [Eq. (1)] can be evaluated using the first order gradient expansion [31–33]. Within this limit, the system dynamics is fully characterized by two quantities, the probability to find the level filled at the energy E, $\phi(t,E)$, and the retarded projection of the Green function $G^r(t,E)$. The energy dependence of these quantities results from the fact that the energy of the level is not sharply defined at $\varepsilon(t)$ as in the weak coupling limit, but gets broadened by the strong coupling to the reservoirs. As shown is [32,34–37] (see also [38]), the retarded Green function is given by

$$G^r(t,E) = [E - \varepsilon(t) - \Sigma^r(t,E)]^{-1}, \tag{2}$$

where the real and imaginary part of the total retarded self-energy, $\Sigma^r(t,E) = \Lambda(t,E) - i\Gamma(t,E)/2$, describe, respectively, the Lamb shift $\Lambda(t,E)$ and the broadening $\Gamma(t,E)$ of the system level caused by the coupling. In the weak coupling limit, $\Gamma \to 0$ and $\Lambda \to 0$. The occupation probability of the level, $\phi(t,E)$, is obtained by solving the equation of motion

$$\begin{aligned}&\{E - \varepsilon(t) - \Lambda(t,E); A(t,E)\phi(t,E)\}\\ &\quad + \{\mathrm{Re}G^r(t,E); \Gamma(t,E)\phi(t,E)\} = \mathcal{C}(t,E),\end{aligned} \tag{3}$$

where $\{f_1; f_2\}$ denotes the Poisson bracket operation $\partial_E f_1 \partial_t f_2 - \partial_t f_1 \partial_E f_2$ and $A(t,E) = -2\mathrm{Im}G^r(t,E)$ is the system spectral function describing the Lorentzian probability amplitude for finding the system at energy E

$$A(t,E) = \frac{\Gamma(t,E)}{[E - \varepsilon(t) - \Lambda(t,E)]^2 + [\Gamma(t,E)/2]^2}. \tag{4}$$

It becomes a delta function centered around $\varepsilon(t)$ in the weak coupling limit. Σ^r as well as Λ and Γ are sums of reservoirs contributions: respectively, $\Sigma^r_\nu(t,E)$, $\Lambda_\nu(t,E)$, and $\Gamma_\nu(t,E)$. Finally, the net particle current entering the level at energy E, $\mathcal{C}(t,E)$ in Eq. (3), is also the sum of different reservoirs contributions, each expressed as a difference between incoming (+) and outgoing (−) electronic currents

$$\begin{aligned}\mathcal{C}_\nu(t,E) &= \mathcal{C}^+_\nu(t,E) - \mathcal{C}^-_\nu(t,E),\\ \mathcal{C}^+_\nu(t,E) &= A(t,E)\Gamma_\nu(t,E)f_\nu(E)[1 - \phi(t,E)],\\ \mathcal{C}^-_\nu(t,E) &= A(t,E)\Gamma_\nu(t,E)\phi(t,E)[1 - f_\nu(E)],\end{aligned} \tag{5}$$

where $f_\nu(E)$ is the Fermi-Dirac distribution of reservoir ν.

In absence of time-dependent driving, ε, Λ and Γ do not depend on time. If the level is in contact with a single reservoir at temperature T and chemical potential μ, it will relax to an equilibrium state where $\phi(t,E)$ will correspond to the Fermi distribution $f(E)$ at T and μ. If another reservoir at the same T and μ is put in contact with the level, the system will remain at equilibrium with respect to the two reservoirs. In that sense, the NEGF satisfies the zeroth law of thermodynamics.

We introduce the renormalized spectral function

$$\mathcal{A}(t,E) = A(1 - \partial_E\Lambda) + \Gamma\partial_E\mathrm{Re}G^r \geq 0, \tag{6}$$

which as its standard version Eq. (4), can be proven non-negative, normalized to one, and to converge to a delta in the weak coupling limit $\mathcal{A} \to 2\pi\delta(E - \varepsilon)$ [38]. We define the particle number, energy, and entropy of the system as energy-resolved versions of the standard weak coupling definitions where the energy resolution is controlled by the renormalized spectral function $\mathcal{A}$

$$\mathcal{N}(t) = \int \frac{dE}{2\pi}\mathcal{A}(t,E)\phi(t,E), \tag{7}$$

$$\mathcal{E}(t) = \int \frac{dE}{2\pi}\mathcal{A}(t,E)E\phi(t,E), \tag{8}$$

$$\mathcal{S}(t) = \int \frac{dE}{2\pi}\mathcal{A}(t,E)\sigma(t,E), \tag{9}$$

where $\sigma(t,E)$ is an energy resolved Shannon entropy

$$\begin{aligned}\sigma(t,E) = &-\phi(t,E)\ln\phi(t,E)\\ &- [1 - \phi(t,E)]\ln[1 - \phi(t,E)].\end{aligned} \tag{10}$$

When attempting to use the standard spectral function rather then the renormalized one in Eqs. (7)–(9), one fails to define a proper entropy production and second law.

The entropy [Eq (9)] was introduced in Refs. [35,36] in the context of the quantum Boltzmann equation. We emphasize that this entropy satisfies the third law. Indeed at equilibrium when $\phi(E) = f(E)$, if we take the limit $T \to 0$, $\sigma^{eq}(E) \to 0$ and therefore, $\mathcal{S}^{eq} \to 0$.

The evolution of the particle number [Eq. (7)]

$$d_t\mathcal{N}(t) = \sum_\nu \mathcal{I}_\nu(t) \tag{11}$$

is given by the sum of the energy-integrated particle currents [Eq. (5)] from reservoir ν

$$\mathcal{I}_\nu(t) = \int \frac{dE}{2\pi}\mathcal{C}_\nu(t,E). \tag{12}$$

The evolution of the energy [Eq. (8)] in turn can be expressed as a first law

$$d_t\mathcal{E}(t) = \sum_\nu \dot{\mathcal{Q}}_\nu(t) + \dot{\mathcal{W}} + \dot{\mathcal{W}}_c. \tag{13}$$

Note that the dots are not partial derivatives, but a symbolic notation for rates. The first contribution is the heat flux from reservoir ν

$$\dot{\mathcal{Q}}_\nu = \mathcal{J}_\nu(t) - \mu_\nu \mathcal{I}_\nu(t), \tag{14}$$

where the energy current from reservoir ν is the energy integral of the energy times the particle current [Eq. (5)] at that energy

$$\mathcal{J}_\nu(t) = \int \frac{dE}{2\pi} E\mathcal{C}_\nu(t,E). \tag{15}$$

The second is the mechanical work performed by the external time-dependent force

$$\dot{\mathcal{W}}(t) = \int \frac{dE}{2\pi}\left(-A\phi\partial_t[E-\varepsilon(t)-\Lambda] - \Gamma\phi\partial_t \mathrm{Re}G^r\right) \tag{16}$$

and the third is the chemical work due to the particle currents flowing from the reservoirs to the system

$$\dot{\mathcal{W}}_c = \sum_\nu \mu_\nu \mathcal{I}_\nu(t). \tag{17}$$

The evolution of the entropy [Eq. (9)] can be expressed as a second law

$$d_t\mathcal{S}(t) = \dot{\mathcal{S}}_i(t) + \sum_\nu \frac{\dot{\mathcal{Q}}_\nu(t)}{T_\nu}, \tag{18}$$

where the entropy production becomes an energy-resolved version of the weak coupling form

$$\dot{\mathcal{S}}_i(t) = \sum_\nu \int \frac{dE}{2\pi}[\mathcal{C}_\nu^+(t,E) - \mathcal{C}_\nu^-(t,E)] \ln \frac{\mathcal{C}_\nu^+(t,E)}{\mathcal{C}_\nu^-(t,E)} \geq 0, \tag{19}$$

which measures the deviation from detailed balance at each energy E, and only vanishes at equilibrium when $\forall\ \nu : f_\nu(E) = \phi(t,E)$.

In the presence of a single reservoir, the second law [Eq. (18)] implies $\dot{\mathcal{Q}} \leq T\partial_t S(t)$. When integrated along transformations connecting an initial and final equilibrium point, we recover Clausius inequality $\mathcal{Q} \leq T\Delta S^{eq}$. Introducing the nonequilibrium grand potential

$$\Omega(t) = \mathcal{E}(t) - \mu\mathcal{N}(t) - T\mathcal{S}(t) \tag{20}$$

and using the first law [Eq. (13)], the second law [Eq. (18)] can also be rewritten as

$$T\dot{\mathcal{S}}_i(t) = \dot{\mathcal{W}}(t) - d_t\Omega(t) \geq 0. \tag{21}$$

For a transformation starting and ending at equilibrium, we thus recover Kelvin's statement of the second law $\mathcal{W}(t) \geq \Delta\Omega^{eq}$, where $\Omega^{eq} = T\int(dE/2\pi)\mathcal{A}(t,E)\ln f(E)$.

For reversible transformations, the inequalities resulting from the positivity of the entropy production become equalities. Such transformation occurs when the level is in contact with a single reservoir and subjected to a quasistatic driving (much slower than the level relaxation time). In this case, the entropy production vanishes to first order $\dot{\mathcal{S}}_i(t) = 0$, while to the same order heat and mechanical work become state functions $\dot{\mathcal{Q}}(t)/T = d_t\mathcal{S}^{eq}$ and $\dot{\mathcal{W}}(t) = d_t\Omega^{eq}$.

We can also prove that (as for weak coupling [46]) the nonequilibrium grand potential is always larger then the equilibrium one, i.e., $\Omega(t) \geq \Omega^{eq}$. Indeed, using Eq. (20) and Eqs. (7)–(9), we find that

$$\Omega(t) - \Omega^{eq} = T\int \frac{dE}{2\pi}\mathcal{A}(t,E)D(t,E) \geq 0, \tag{22}$$

where the energy-resolved relative entropy reads

$$D(t,E) = \phi(t,E)\ln\frac{\phi(t,E)}{f(E)} + [1-\phi(t,E)]\ln\frac{1-\phi(t,E)}{1-f(E)} \geq 0. \tag{23}$$

The non-negativity of [Eq. (22)] follows from $\mathcal{A}, D \geq 0$.

We consider in Fig. 2, the quantum level in contact with a single reservoir. Its energy is driven by the external force according to the protocol described in the caption. Figure 2(a) depicts the heat flux [Eq. (14)] and entropy production [Eq. (19)] increase with time as the distribution ϕ departs from its equilibrium value. The reversible transformation ($\dot{S}_i = 0$) is reached in the very slow driving limit when $\omega_0 \to 0$, as shown on Fig. 2(b).

We note that the system energy [Eq. (8)] and particle number [Eq. (7)] as well as the energy and particle currents [Eqs. (15) and (12)] that we introduced cannot be expressed in term of expectation values of operators. One may interpret this as a manifestation of the fact that defining a boundary between the system and the reservoirs in case of strong interaction is an ambiguous task. The main argument in favor of the proposed definitions is that they lead to a consistent nonequilibrium thermodynamics at slow driving.

In absence of driving, the system eventually reaches a steady state (equilibrium or nonequilibrium), where the system properties such as $\phi(t,E)$, $\mathcal{A}(t,E)$ and Eqs. (7)–(9) become time independent. In this case, we find that $\mathcal{J}_\nu(t) = -\mathrm{Tr}[\hat{H}_\nu d_t\hat{\rho}(t)]$ and $\mathcal{I}_\nu(t) = -\sum_{k\in\nu}\mathrm{Tr}[\hat{c}_k^\dagger\hat{c}_k d_t\hat{\rho}(t)]$ [38]. The first and second law at steady state simplify to

$$\dot{\mathcal{W}}_c = -\sum_\nu \dot{\mathcal{Q}}_\nu(t), \qquad \dot{\mathcal{S}}_i(t) = -\sum_\nu \frac{\dot{\mathcal{Q}}_\nu(t)}{T_\nu} \geq 0. \tag{24}$$

Since in the weak coupling limit A and $\mathcal{A}$ become delta functions, we recover the usual definitions of stochastic

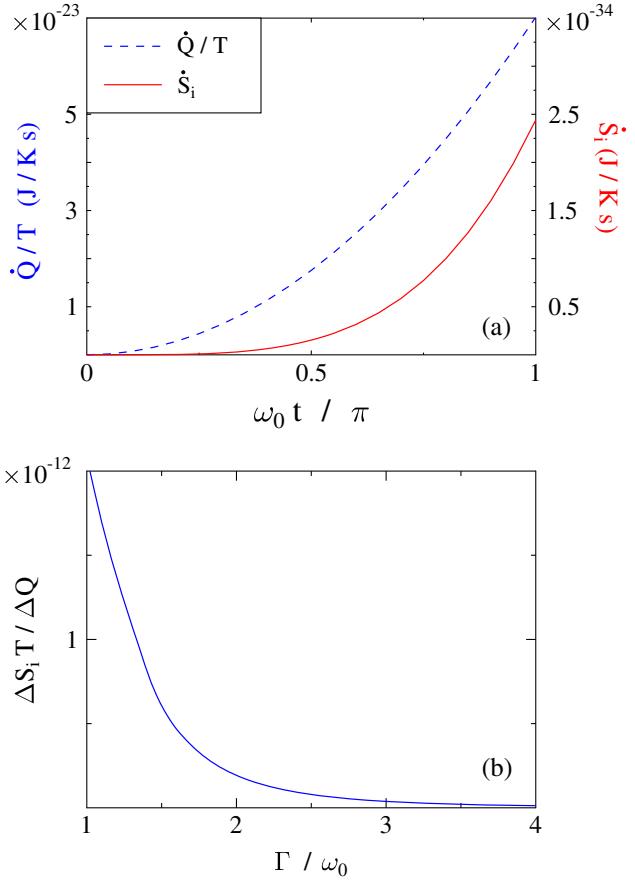

FIG. 2 (color online). Heat flux [Eq. (14)] and entropy production [Eq. (19)], for the quantum level in contact with a single reservoir at $T = 300$ K. The external force drives the level energy as $\varepsilon(t) = \varepsilon_0 + \Delta(1 - \cos\omega_0 t)/2$ from ε_0 at $t = 0$ to $\varepsilon_0 + \Delta$ at $t = \pi/\omega_0$, where $\varepsilon_0 = -0.02$ eV and $\Delta = 0.02$ eV. Entropy production (solid line, red) and heat flux (dashed line, blue) are depicted in (a) as functions of time for $\Gamma = 0.01$ eV and $\omega_0 = 0.01$ eV. The ratio of their time-integrated values is depicted in (b) as a function of the driving rate ω_0.

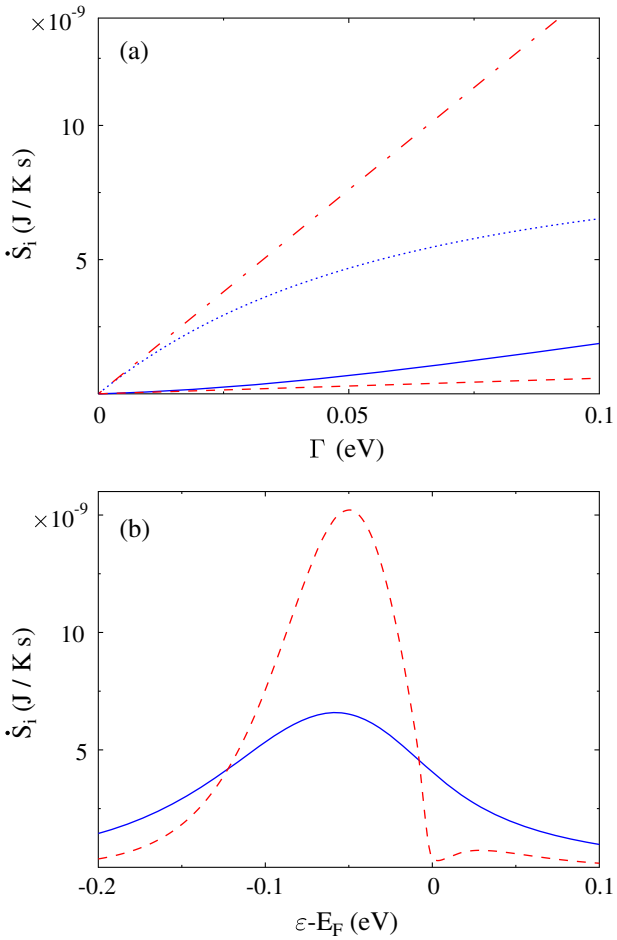

FIG. 3 (color online). Entropy production for the quantum level at steady state between two reservoirs $\nu = L, R$ (with $T_L = 300$ K, $T_R = 10$ K, $\mu_L = -0.05$ eV, $\mu_R = E_F = 0$) as a function of: (a) the interaction strength to the reservoirs $\Gamma = 2\Gamma_L = 2\Gamma_R$, (b) the position of the level ε. The strong coupling entropy production [Eq. (19)] is depicted for $\varepsilon = -0.05$ eV (dotted line, blue) and $\varepsilon = 0.05$ eV (solid line, blue), and its weak coupling counterpart for $\varepsilon = -0.05$ eV (dash-dotted line, red) and $\varepsilon = 0.05$ eV (dashed line, red). The energy grid used spans from -3 to 3 eV with step 10^{-6} eV.

thermodynamics [3,47,48] for a master equation with Fermi's golden rule rates describing the evolution of the occupation probability of the level [8,49].

Figure 3(a) depicts the entropy production of the quantum level at steady state between two reservoirs with different temperatures and chemical potentials. The entropy production is plotted as a function of the coupling strength with the reservoirs, when this device operates as a thermoelectric. As the coupling strength increases, the discrepancy between the entropy production [Eq. (19)] and its weak coupling counterpart (dotted vs dash-dotted and solid vs dashed lines) becomes more pronounced. We note that the weak coupling prediction can overestimate (dash-dotted line) or underestimate (dashed line) the entropy production [Eq. (19)]. Figure 3(b) depicts the same two entropy productions for $\Gamma = 0.1$ eV as functions of the position of the level. In the weak coupling regime, this system satisfies the condition of tight coupling (energy and particle current are proportional) [50–53] which enables it to operate reversibly at finite bias, as seen at $\varepsilon = 0.0017$ eV. However, the level broadening induced by the strong coupling to the reservoirs completely breaks the tight coupling property and reversibility is lost.

The main message of this Letter is that it is possible to formulate a consistent nonequilibrium thermodynamics for driven open quantum systems strongly coupled to their reservoirs. No such theory existed before and the definitions we used seem to be the only ones rendering such a formulation possible. We considered a fermionic level coupled to fermionic reservoirs, but our approach can be straightforwardly extended to any noninteracting fermionic or bosonics systems. It can probably be extended to describe interacting systems, but considering fast drivings

remains out of its reach since it relies on treating slow time-dependent driving forces (i.e., gradient expansion). We are now in the position to address important problems such as characterizing the dissipation caused by connecting or disconnecting a system from its reservoirs, or assessing the difference in performance between strongly coupled and weakly coupled energy-converting devices.

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