


Quantum thermodynamics of nanoscale steady states far from equilibrium

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We develop an exact quantum thermodynamic description for a noninteracting nanoscale steady state that couples strongly with multiple reservoirs. We demonstrate that there exists a steady-state extension of the thermodynamic function that correctly accounts for the multiterminal Landauer-Büttiker formula of quantum transport of charge, energy, or heat via the nonequilibrium thermodynamic relations. Its explicit form is obtained for a single bosonic or fermionic level in the wide-band limit, and corresponding thermodynamic forces (affinities) are identified. Nonlinear generalization of the Onsager reciprocity relations are derived. We suggest that the steady-state thermodynamic function is also capable of characterizing the heat current fluctuations of the critical transport where the thermal fluctuations dominate. Also, the suggested nonequilibrium steady-state thermodynamic relations seemingly persist for a spin-degenerate single level with local interaction.

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I. INTRODUCTION

Constructing a thermodynamic theory that applies consistently to nonequilibrium steady states has long been a theoretical challenge in many fields of science, not only in physics but also in chemistry and biology. Steady states differ from equilibrium states by being driven by the external environments (reservoirs) and accommodating finite flows that induce entropy production. Formulating thermodynamics for such irreversible systems is notoriously difficult; successes have been mainly achieved within the linear-response theory, where various transport coefficients can be related to fluctuations in equilibrium [1,2]. Beyond the linear-response regime, a possible thermodynamic formulation has been anticipated for the steady states [3–7]. Yet the theory has been largely unexplored, partly because basic concepts such as temperature and entropy get elusive and questioned when treating an open, irreversible system.

In recent years, it has been recognized that thermodynamic laws are consistent with quantum properties of open nanoscale systems typically connected with multiple reservoirs with different chemical potentials and temperatures (Fig. 1). The emergence of thermodynamics is somewhat unexpected; the situation is opposite to the conventional thermodynamic limit, because it involves only a few particles, even a single one. The statistical ensemble average is replaced by quantum averaging, and the applicability of thermodynamics results from its quantum nature. Nanoscale systems provide a rare and novel opportunity to study the steady-state thermodynamics, without relying on any statistical ensemble hypothesis [8].

As a realization of nonequilibrium systems, a steady-state nanoscale system is notable because of its strong coupling with the reservoirs. One cannot rely on local equilibrium hypothesis to characterize a nanosystem. Any temperature or chemical potential cannot be assigned to it *a priori*,

except for a system coupled with a single reservoir. Strongly coupled reservoirs make effective dynamics non-Markovian with memory effect, and the Lindblad form of the master equation invalidated. The nonequilibrium density matrix is represented not by the standard Gibbs ensemble but by a generalized one (the MacLennan-Zubarev type) [9–12]. A consistent thermodynamic framework is nontrivial even for noninteracting transport. It has been striven for by many approaches [13–17], but has not been fully disclosed so far.

In this paper, we develop a thermodynamic description of the steady state at nanoscale, entirely based on quantum mechanics. A steady-state extension of the Massieu-Planck function Φ_{ss} , which is determined by normalization of the reduced density matrix, is singled out as a nonequilibrium thermodynamic function. One salient feature of this quantum construction is that the resulting thermodynamic function Φ_{ss} unavoidably becomes stationary in time because the steady-state density matrix is time independent. It contrasts sharply with a naive expectation that a steady-state thermodynamic function might increase in time because it may include an increasing entropic contribution. Notwithstanding we will demonstrate that the function Φ_{ss} is viable to describe the steady-state properties far from equilibrium. To fully characterize the steady state, we also need to identify a correct set of parameters that control the steady state. A consistent choice of them includes local (inverse) temperature and chemical potential $(\bar{\beta}, \bar{\beta}\bar{\mu})$, as well as various affinities (A_a^N, A_a^E) that are thermodynamic forces to drive the system out of equilibrium [see Eqs. (21) for their definitions]. With these parameters, the significance of Φ_{ss} is compactly represented in the differential form

$$d\Phi_{ss} = \bar{N}d(\bar{\beta}\bar{\mu}) - \bar{E}d\bar{\beta} + \frac{\hbar}{2\gamma} \sum_a (I_a dA_a^N + J_a dA_a^E), \quad (1)$$

where \bar{N} or \bar{E} is the average occupancy or energy of the system, while I_a or J_a is a nonlinear inflow of particle or energy from the reservoir a . The constant γ is the total relaxation rate of the system. The relation (1) serves as a nonequilibrium extension

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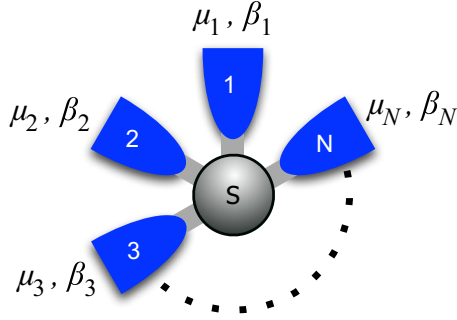


FIG. 1. An open system (S) connected with multiple reservoirs with different inverse temperatures β_a and chemical potentials μ_a for $a = 1, \dots, N$.

of the thermodynamic relation of the Massieu-Planck function (see Appendix A). Being stationary in time, the function Φ_{ss} does not refer to the internal entropy. Yet Φ_{ss} characterizes the entropy production rate by

$$\frac{dS}{dt} = \frac{2\gamma}{\hbar} \sum_a \left(A_a^E \frac{\partial \Phi_{ss}}{\partial A_a^E} + A_a^N \frac{\partial \Phi_{ss}}{\partial A_a^N} \right). \quad (2)$$

Formulas (1) and (2) will be proved to be exact for a noninteracting single bosonic or fermionic level that couples linearly with multiple reservoirs. Moreover we argue that the above thermodynamic structure in Eqs. (1) and (2), found in a noninteracting steady state, persists even in a steady state of the model with local interaction, namely, the single-impurity Anderson model.

II. MODEL AND KNOWN RESULTS

The total Hamiltonian consists of $H = H_S + H_R + H_{SR}$, whose terms represent a nanoscale system (“quantum dot”), multiple reservoirs with different inverse temperatures β_a and chemical potentials μ_a (for $a = 1, \dots, N$), and the linear coupling between the system and the reservoirs (see Fig. 1). They are

$$H_S = \epsilon_d d^\dagger d, \quad (3)$$

$$H_R = \sum_a \epsilon_{ak} c_{ak}^\dagger c_{ak}, \quad (4)$$

$$H_{SR} = \sum_{a,\sigma} (V_{da} d^\dagger c_{ak} + V_{ad} c_{ak}^\dagger d), \quad (5)$$

where d^\dagger creates a particle with energy ϵ_d at the system and c_{ak}^\dagger , with energy ϵ_{ak} at the reservoir a . Particles can be bosonic or fermionic. We present the results for both cases simultaneously with composite signs (with the upper for bosonic; the lower for fermionic).

The presence of the reservoir makes the nanoscale system dissipative, inducing a finite resonant width $\gamma_a = \pi |V_{da}|^2 \rho_a$ due to the reservoir a (with its density of states ρ_a). Quantum transport across a noninteracting system can be solved exactly by several approaches, such as the scattering method, the equation-of-motion method, or the nonequilibrium Green’s function method. When we take the wide-band approximation,

the inflow of particle I_a or of energy J_a from the reservoir a is given by the Landauer-Büttler formula [18–20]

$$I_a = \frac{2}{\hbar} \sum_b \frac{\gamma_a \gamma_b}{\gamma} \int d\epsilon \rho(\epsilon) [f_a(\epsilon) - f_b(\epsilon)], \quad (6)$$

$$J_a = \frac{2}{\hbar} \sum_b \frac{\gamma_a \gamma_b}{\gamma} \int d\epsilon \rho(\epsilon) \epsilon [f_a(\epsilon) - f_b(\epsilon)]. \quad (7)$$

Here $\rho(\epsilon)$ refers to the spectral function of the system,

$$\rho(\epsilon) = \frac{\gamma/\pi}{(\epsilon - \epsilon_d)^2 + \gamma^2}, \quad \gamma = \sum_a \gamma_a, \quad (8)$$

and $f_a(\epsilon) = [e^{\beta_a(\epsilon - \mu_a)} \mp 1]^{-1}$ is the distribution function of the reservoir a . These currents are usually expressed by the transmission $\mathcal{T}_{ab}(\epsilon) = 4\pi \gamma_a \gamma_b \rho(\epsilon)/\gamma$ between the reservoirs a and b , but we prefer writing them in the above form. One can evaluate them analytically in terms of the digamma function [see Eqs. (D1) and (D2)]. Heat current flowing from the reservoir a is defined by $J_a^Q = J_a - \mu_a I_a$. The average number \bar{N} and energy \bar{E} of the system are given by

$$\bar{N} = \int d\epsilon \sum_a \frac{\gamma_a}{\gamma} \rho(\epsilon) f_a(\epsilon), \quad (9)$$

$$\bar{E} = \int d\epsilon \sum_a \frac{\gamma_a}{\gamma} \rho(\epsilon) \epsilon f_a(\epsilon). \quad (10)$$

We will show below that the steady-state thermodynamic function Φ_{ss} , which is constructed quantum mechanically, correctly produces quantum transport equations (6) and (7) as well as local quantities (9) and (10) via the differential relation (1).

III. MASSIEU-PLANCK FUNCTION

Analogous to an equilibrium system, our basic assumption is that the partition function which normalizes the density matrix bridges between a microscopic model and its thermodynamics. We suppose its steady-state extension is provided instead by normalizing the reduced density matrix $\hat{\rho}_{red}$ of the relevant system. In treating the steady state, we find it advantageous to use the Massieu-Planck function [21], which is defined by the logarithm of the (effective) partition function.

A. Single-reservoir Massieu-Planck function

As for an open system that connects with a single reservoir with β and μ , the effective thermodynamics has long been investigated [22–27]. By recasting it, the single-reservoir Massieu-Planck function is found to be (see Appendix B)

$$\Phi_1(\beta, \beta\mu) = \mp \int_{-\infty}^{\infty} d\epsilon \rho(\epsilon) \ln[1 \mp e^{-\beta(\epsilon - \mu)}]. \quad (11)$$

The energy integration actually diverges in the wide-band limit, so some regularization is needed. In Appendix C, we show the explicit analytical form of $\Phi_1(\beta, \beta\mu)$ with regularization, and examine its various thermodynamic properties that are independent of regularization. The physics of Φ_1 is transparent; the level of the open nanosystem acquires finite broadening due to coupling with the reservoir. We make a point of regarding Φ_1 as a function of β and $\beta\mu$, since they are parameters

conjugate to particle number and energy. We stress that they are originally external parameters specified by the reservoir. The implication of β and μ as thermodynamic parameters is somewhat blurred because the reduced density matrix ϱ_{red} is no longer represented by the standard Gibbs ensemble.

B. Steady-state Massieu-Planck function

One can calculate the steady-state Massieu-Planck function Φ_{ss} that couples with multiple reservoirs by normalizing the reduced density matrix ϱ_{red} . As we work on noninteracting systems, the calculation can drastically be simplified by utilizing a Gaussian nature of $\hat{\varrho}_{\text{red}}$, in light of Zubarev's relevant distributions and nonequilibrium statistical operators [9] (see also [28–30]). Asking $\bar{N} = \langle d^\dagger d \rangle$ to reproduce Eq. (9), we deduce that $\hat{\varrho}_{\text{red}}$ may well be represented in terms of relevant field operators $\psi_a(\varepsilon)$ and $\psi_a^\dagger(\varepsilon)$, satisfying

$$[\psi_a(\varepsilon), \psi_{a'}^\dagger(\varepsilon')]_{\mp} = \delta_{aa'} \delta(\varepsilon - \varepsilon'), \quad (12)$$

$$\langle \psi_{a'}^\dagger(\varepsilon') \psi_a(\varepsilon) \rangle = \delta_{aa'} \delta(\varepsilon - \varepsilon') f_a(\varepsilon), \quad (13)$$

$$d = \sum_a \int d\varepsilon \sqrt{\frac{\gamma_a}{\gamma}} \rho(\varepsilon) \psi_a(\varepsilon). \quad (14)$$

Then the function Φ_{ss} normalizes $\hat{\varrho}_{\text{red}}$ as

$$\hat{\varrho}_{\text{red}} = \exp \left[-\Phi_{\text{ss}} - \sum_{a,\varepsilon} \beta_a(\varepsilon - \mu_a) \psi_a^\dagger(\varepsilon) \psi_a(\varepsilon) \right], \quad (15)$$

where $\sum_{a,\varepsilon}(\dots) = \sum_a \int d\varepsilon (\gamma_a/\gamma) \rho(\varepsilon) (\dots)$ is the summation/integral over the energy shell and the reservoirs. Determining Φ_{ss} by imposing $\text{Tr} \hat{\varrho}_{\text{red}} = 1$ is equivalent to evaluating the functional determinant. A quick, symbolic way to evaluate it is

$$\exp(\Phi_{\text{ss}}) = \prod_{a,\varepsilon} [1 \mp e^{-\beta_a(\varepsilon - \mu_a)}]_{\mp}^{-1}, \quad (16)$$

$$= \exp \left(\mp \sum_{a,\varepsilon} \ln [1 \mp e^{-\beta_a(\varepsilon - \mu_a)}] \right). \quad (17)$$

It expresses the steady-state Massieu-Planck function

$$\Phi_{\text{ss}} = \sum_a \frac{\gamma_a}{\gamma} \Phi_1(\beta_a, \beta_a \mu_a) \quad (18)$$

as a superposition of the single-reservoir contribution $\Phi_1(\beta_a, \beta_a \mu_a)$. Hence Φ_{ss} can be evaluated analytically. The manipulation of Eq. (16) is due to observing that $(\gamma_a/\gamma)\rho(\varepsilon)$ acts as (fractional) degeneracies satisfying $\sum_{a,\varepsilon} 1 = 1$; such analytical continuation is validated because it correctly reproduces the single-reservoir result in Eq. (11).

The relevant field operator $\psi_a^\dagger(\varepsilon)$ in Eqs. (12)–(15) has a clear physical meaning. One can construct the steady-state density matrix $\hat{\varrho}_{\text{ss}}$ of the total system (the system plus the reservoirs) [10,12,31–34],

$$\hat{\varrho}_{\text{ss}} \propto \exp \left[- \sum_a \sum_k \beta_a(\epsilon_{ak} - \mu_a) \psi_{ak}^\dagger \psi_{ak} \right], \quad (19)$$

where $\psi_{ak}^\dagger = \Omega c_{ak}^\dagger \Omega^\dagger$ is a scattering-state field of the reservoir a that is defined by the Møller operator Ω . The field ψ_{ak}^\dagger becomes a coherent superposition of fields c_{ak}^\dagger and d^\dagger . Accordingly, field d is solved to be a superposition of the scattering fields involving all the reservoir fields, as in Eq. (14). It accounts for quantum coherence between the system and the reservoirs. The average density is $\langle \psi_{ak}^\dagger \psi_{ak} \rangle = f_a(\epsilon_{ak})$, and the canonical (anti)commutation relation is preserved. The relevant field $\psi_a(\varepsilon)$ is nothing but an energy representation of the scattering-state field ψ_{ak} .

We cannot emphasize too much the peculiar nature of Eq. (18). Although such a superposition is a common trait of quantum mechanics, Eq. (18) tells us that the function Φ_{ss} that describes the irreversible steady state (with the increasing entropy) is unchanged in time and given by a superposition of the Φ_1 's of the single reservoirs, each of which refers to the entropy-preserving, reversible system. One may notice such a trait of superposition in the expression of average number \bar{N} or energy \bar{E} [Eqs. (9) or (10)], but it is far from obvious that one can use Φ_{ss} to describe quantum transport I_a and J_a . To fully disclose the steady-state thermodynamics, one needs to find the relevant controlling parameters for it.

C. Affinities

Finding the correct set of appropriate controlling parameters arbitrary away from equilibrium is quite nontrivial, but it is imperative to establish the steady-state thermodynamic relations. The function Φ_{ss} of Eq. (18) depends on $2N$ independent external parameters $\{\beta_a, \mu_a\}_{a=1,\dots,N}$ specified by the reservoirs. Among them, we expect that two *local* parameters (temperature and chemical potential) regulate the average particle number and energy [Eqs. (9) and (10)], while all other $2N - 2$ parameters (the difference of temperatures and/or chemical potentials) drive the system out of equilibrium and cause irreversible processes. The latter parameters are called thermodynamic forces or affinities. Among them, we will identify the relevant parameters $(\bar{\beta}, \bar{\beta}\mu, \{A_a^N, A_a^E\})$ defined in Eqs. (21) below, which describe the quantum transport as well as thermodynamic properties. This constitutes our main result, with the steady-state thermodynamic function (18).

One can identify affinities and their associated currents by examining the internal entropy production rate [21]. In the system we consider, it is balanced with the entropy inflow, so that we find

$$\frac{dS}{dt} = - \sum_a \beta_a (J_a - \mu_a I_a). \quad (20)$$

Its positivity follows because the distribution function $f_a(\varepsilon)$ is a decreasing function regarding $\beta_a(\varepsilon - \mu_a)$ [10,15,32,35]. The form of Eq. (20) tells us to introduce two types of affinities associated with each reservoir: chemical affinities A_a^N to generate particle currents, which is a deviation of $\beta\mu$, and thermal affinities A_a^E to generate energy currents, which is a deviation of $-\beta$. Those deviations must be defined from some reference values, $\bar{\beta}$ and $\bar{\beta}\mu$, which in turn regulate \bar{N} and \bar{E} . We choose to introduce affinities for conserved particle and energy currents rather than heat currents. The conservation laws are fulfilled by the condition $\sum_a \gamma_a A_a^E = \sum_a \gamma_a A_a^N = 0$. Hence

a pair of $A_a^{N,E}$ are redundant (see Appendix E for an explicit construction). All things considered, we come to make the following choice of local quantities $\bar{\beta}$ and $\bar{\beta}\mu$, and affinities:

$$A_a^N = \beta_a \mu_a - \bar{\beta}\mu, \quad \bar{\beta}\mu = \sum_a \frac{\gamma_a}{\gamma} \beta_a \mu_a, \quad (21a)$$

$$A_a^E = -\beta_a + \bar{\beta}, \quad \bar{\beta} = \sum_a \frac{\gamma_a}{\gamma} \beta_a. \quad (21b)$$

Viewing Φ_{ss} Eq. (18) as a function of the above set of parameters $(\bar{\beta}, \bar{\beta}\mu, \{A_a^N, A_a^E\})$ of Eqs. (21), it is now straightforward to find (see Appendix E for details)

$$\bar{N} = \frac{\partial \Phi_{ss}}{\partial (\bar{\beta}\mu)}, \quad \bar{E} = -\frac{\partial \Phi_{ss}}{\partial \bar{\beta}}, \quad (22)$$

$$I_a = \frac{2\gamma}{\hbar} \frac{\partial \Phi_{ss}}{\partial A_a^N}, \quad J_a = \frac{2\gamma}{\hbar} \frac{\partial \Phi_{ss}}{\partial A_a^E}. \quad (23)$$

The expression in Eq. (2) of the entropy production rate dS/dt follows from Eqs. (20) and (23).

IV. DISCUSSION

A. Local temperature

Our definitions of local temperature $\bar{\beta}$ and chemical potential $\bar{\mu} = \bar{\beta}\mu/\bar{\beta}$ are motivated by the theoretical consistency of the thermodynamic formulation. Alternatively, one may probe local quantities by measurements such as the scanning thermal technique [36]. Those *probed* quantities, β_p and μ_p , are determined by the no-flow condition of charge and energy when attaching the probe reservoir [37–39]:

$$\int d\varepsilon \rho(\varepsilon) \varepsilon^k [\bar{f}(\varepsilon) - f_p(\varepsilon)] = 0 \quad (\text{for } k = 0, 1), \quad (24)$$

where $\bar{f}(\varepsilon) = \sum_a (\gamma_a/\gamma) f_a(\varepsilon)$ is the effective distribution of the reservoirs. In a general nonlinear setting far from equilibrium, parameters β_p and μ_p may differ from $\bar{\beta}$ and $\bar{\mu}$. However, as for the linear deviation, the probed quantities β_p and μ_p agree with $\bar{\beta}$ and $\bar{\mu}$, because the effective distribution $\bar{f}(\varepsilon)$ can be expanded as $\bar{f}(\varepsilon) \approx f_p(\varepsilon) + f_p'(\varepsilon)[(\bar{\beta} - \beta_p)\varepsilon - (\bar{\beta}\mu - \beta_p\mu_p)]/\beta_p$. We also note that the scale $\bar{\mu}$ has played an important role in characterizing nonlinear electronic transport in the Kondo regime through an interacting dot [40].

Explicit forms of local quantities and affinities in Eqs. (21) are outcomes of the wide-band approximation, which is well justified for quantum coherent transport through a nanostructure. If γ_a were to acquire substantial energy dependence, one could nonetheless construct Φ_{ss} by generalizing Eq. (18) to take an energy-dependent superposition for each energy shell. However, it is quite nontrivial in this situation to identify appropriate controlling parameters that enable us to construct the thermodynamic description.

B. Maxwell relations and nonlinear generalization of the Onsager relations

The existence of the function Φ_{ss} that satisfies the differential form of Eq. (1) has important consequences for the steady-state thermodynamic structure. One can derive various steady-state extensions of Maxwell relations by using the

symmetry of second derivatives. For instance, we see the $\bar{\beta}\mu$ dependence of the current can be obtained by the chemical affinity dependence of the occupancy, as in

$$\frac{\partial^2 \Phi_{ss}}{\partial A_a^N \partial (\bar{\beta}\mu)} = \frac{\partial \bar{N}}{\partial A_a^N} = \frac{\hbar}{2\gamma} \frac{\partial I_a}{\partial (\bar{\beta}\mu)}. \quad (25)$$

Many other relations are derived similarly. One can make a nonlinear generalization of the Onsager reciprocity relations by using the symmetry $\partial^2 \Phi_{ss}/\partial A_a^E \partial A_a^N$:

$$\frac{2\gamma}{\hbar} \frac{\partial \Phi_{ss}}{\partial A_a^E \partial A_a^N} = \frac{\partial I_a}{\partial A_a^E} = \frac{\partial J_a}{\partial A_a^N}, \quad (26)$$

which is valid for nonlinear responses. In the linear-response limit (or the zero-affinity limit), the above gives the usual Onsager reciprocal relations between the cross coefficients.

C. Implication in the interacting system

We have demonstrated that the function Φ_{ss} characterizes the steady state, based on a noninteracting transport model through a single level. Notwithstanding, the validity of the thermodynamic structure in Eqs. (1) and (2) seems to go beyond noninteracting systems to include a steady-state with local interaction. Let us consider the spin-degenerate fermionic single level with local interaction connecting with the multiple reservoirs, namely, the nonequilibrium single-impurity Anderson model. For that system, we can still derive the Landauer-Büttiker type formulas (6) and (7) by help of nonequilibrium Green's functions [20,40–43], where the many-body effect is encapsulated only in the spectral function $\rho(\varepsilon) = -\text{Im } G^R(\varepsilon)/\pi$. Moreover, in the wide-band limit, the current conservation of charge and energy enforces Eqs. (9) and (10) even with interaction [40]. Note, the expression of \bar{N} may be understood as a generalization of the Friedel sum rule [44–46] which holds at the zero temperature.

Therefore the structure of Eqs. (6) and (10) is intact even for the single-impurity Anderson model in the wide-band limit. From these, we can deduce that a small deviation of Φ_{ss} should take a form of Eq. (1). Equivalently, it can be written as

$$d\Phi_{ss} = \mp \int d\varepsilon \rho(\varepsilon) \sum_a \frac{\gamma_a}{\gamma} d \ln[1 \mp e^{-\beta_a(\varepsilon - \mu_a)}], \quad (27)$$

$$= \int d\varepsilon \rho(\varepsilon) \sum_a \frac{\gamma_a}{\gamma} f_a(\varepsilon) d[\beta_a \mu_a - \beta_a \varepsilon]. \quad (28)$$

The deviation $d[\beta_a \mu_a - \beta_a \varepsilon]$ is taken by regarding $(\bar{\beta}, \bar{\beta}\mu, A_a^N, A_a^E)$ as independent parameters, which gives Eq. (1). The form (28) is surprising when one recalls that the local interaction makes the reduced density matrix $\hat{\rho}_{\text{red}}$ non-Gaussian and the spectral function $\rho(\varepsilon)$ dependent on the parameters. We suspect that there is some cancellation between the quadratic and quartic contributions, similar to the nonequilibrium Ward identities [47], because $\hat{\rho}_{ss}$ still takes the Gaussian form of Eq. (19) in terms of scattering-state fields, even for an interacting dot [10,12,31–34]. An actual mechanism is missing though.

D. Characterizing low-temperature heat current fluctuations

In the context of the large deviation approach to equilibrium statistical mechanics [48], the free energy characterizes not only average quantities but also their fluctuations, being the cumulant generating function (CGF). Since the CGF of steady-state currents has been known [49–51], it would be desirable to see a connection with Φ_{ss} , but a generic link between the CGF and Φ_{ss} is missing. Nevertheless we can show that the heat transport at low temperature gives a concrete example of how Φ_{ss} is capable of characterizing the fluctuations.

The average heat current between two reservoirs (with different temperatures $\beta_{1,2}$ and the same chemical potential $\mu = 0$) exhibits the universal behavior [52,53]

$$J^Q \approx \frac{c\pi}{12\hbar} \left(\frac{1}{\beta_1^2} - \frac{1}{\beta_2^2} \right), \quad (29)$$

with the transmission coefficient $c = \mathcal{T}_{12}(\mu)$. It is known that the low-temperature current fluctuations are dominated by the thermal (Johnson-Nyquist) noise, even for the extreme nonequilibrium situation $\beta_1/\beta_2 \gg 1$ [54]. Since one can connect the thermal noise with the thermal conductance, one may well say that Φ_{ss} characterizes those noises.

For perfect (or “critical”) transmission $c \approx 1$ which is realized for $\gamma_1 = \gamma_2$, one can develop conformal field theory to construct the CGF $F(\lambda)$ to characterize low-temperature heat current fluctuations [55–57], which is found to be

$$F(\lambda) \approx \frac{c\pi}{12\hbar} \left[\frac{i\lambda}{\beta_1(\beta_1 - i\lambda)} - \frac{i\lambda}{\beta_2(\beta_2 + i\lambda)} \right]. \quad (30)$$

It corresponds to the the low-temperature limit of Ref. [51]. What is interesting in the present context is that Bernard and Doyon [56] have noticed that the function $dF(\lambda)/d\lambda$ is related to the nonlinear heat current J by what they call the extended fluctuation relations. Then by comparing with Eq. (1), we come to see that the CGF $F(\lambda)$ is directly given in terms of Φ_{ss} :

$$F(\lambda) = \frac{\gamma}{\hbar} [\Phi_{ss}(\bar{\beta}, A^E + 2i\lambda) - \Phi_{ss}(\bar{\beta}, A^E)]. \quad (31)$$

with $\bar{\beta} = (\beta_1 + \beta_2)/2$ and $A^E = -\beta_1 + \beta_2$. Indeed, the low-temperature behavior of Φ_{ss} is readily evaluated from Eqs. (C11) and (18) as

$$\Phi_{ss} \approx \frac{c\pi}{12\gamma} \left(\frac{1}{\beta_1} + \frac{1}{\beta_2} \right), \quad (32)$$

and putting it into Eq. (31) exactly reproduces Eq. (30). It is noted that the fluctuation theorem $F(\lambda) = F(iA^E - \lambda)$ is equivalent to the inversion symmetry $\Phi_{ss}(\bar{\beta}, A^E) = \Phi_{ss}(\bar{\beta}, -A^E)$ in this case.

V. CONCLUSION

In summary, we have developed a thermodynamic description of the nonequilibrium steady state that connects with multiple reservoirs, and demonstrated that the steady-state Massieu-Planck function Φ_{ss} can characterize consistently its quantum transport properties of charge, energy, or heat. The positive entropy production rate caused by irreversible processes is also characterized by Φ_{ss} . We have evaluated explicitly for a single bosonic or fermionic level that connects

with multiple reservoirs, and argued that the same thermodynamic structure persists even for a steady state with local interaction and that the heat current fluctuations are related to the function Φ_{ss} at low temperature.

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APPENDIX A: THERMODYNAMIC RELATIONS OF THE EQUILIBRIUM MASSIEU-PLANCK FUNCTION

We recall some basic thermodynamic relations of the equilibrium Massieu-Planck function Φ_{eq} , which is defined as the logarithm of the (grand) partition function [21]. With a given thermodynamic potential $\Omega(T, \mu)$ as a function of the temperature T and the chemical potential μ , the function Φ_{eq} can be written as $\Phi_{eq} = -\Omega(T, \mu)/T$. Below we show how it is beneficial to regard Φ_{eq} as a function of the inverse temperature $\beta = 1/T$ and $\beta\mu$. The volume of the system is irrelevant and ignored because we treat a nanoscale system.

Starting with the thermodynamic relation $d\Omega = -Nd\mu - SdT$, the thermodynamic relation of Φ_{eq} becomes

$$d\Phi_{eq} = -\Omega d\beta + N\beta d\mu - S \frac{d\beta}{\beta} = Nd(\beta\mu) - Ed\beta, \quad (A1)$$

where we identify the entropy $S = -\beta\Omega + \beta(E - N\mu)$. One can check the above directly by making a quantum statistical construction of the partition function $e^{\Phi_{eq}}$. For noninteracting bosonic and fermionic particles with levels $\{\epsilon_\alpha\}$, one finds

$$\Phi_{eq}(\beta, \beta\mu) = \mp \sum_{\alpha} \ln[1 \mp e^{-\beta(\epsilon_\alpha - \mu)}]. \quad (A2)$$

Then it is easy to see

$$\left. \frac{\partial \Phi_{eq}(\beta, \beta\mu)}{\partial(\beta\mu)} \right|_{\beta} = \sum_{\alpha} f(\epsilon_\alpha) = N, \quad (A3)$$

$$\left. \frac{\partial \Phi_{eq}(\beta, \beta\mu)}{\partial\beta} \right|_{\beta\mu} = - \sum_{\alpha} \epsilon_\alpha f(\epsilon_\alpha) = -E, \quad (A4)$$

$$\left. \frac{\partial \Phi_{eq}(\beta, \beta\mu)}{\partial\beta} \right|_{\mu} = -(E - \mu N), \quad (A5)$$

where $f(\epsilon) = [e^{\beta(\epsilon - \mu)} \mp 1]^{-1}$ is the distribution function. The entropy $S = -\partial\Omega/\partial T|_{\mu}$ is given by

$$S = \Phi_{eq} - \beta \left. \frac{\partial \Phi_{eq}}{\partial\beta} \right|_{\mu} = \Phi_{eq} + \beta(E - \mu N). \quad (A6)$$

The first equality means that Φ_{eq} is a Legendre transform of S regarding β with a fixed μ . One finds the entropy S taking a familiar Shannon-like form:

$$S = - \sum_{\alpha} \{ f(\epsilon_\alpha) \ln f(\epsilon_\alpha) \mp [1 \pm f(\epsilon_\alpha)] \ln [1 \pm f(\epsilon_\alpha)] \}. \quad (A7)$$

APPENDIX B: EFFECTIVE SINGLE-LEVEL THERMODYNAMICS COUPLED WITH A SINGLE RESERVOIR

The calculation of the effective free energy of a single level ϵ_d that couples with a single reservoir (with the inverse temperature β and the chemical potential μ) has been long known for fermionic systems [23] as well as bosonic systems [25–27]. As in equilibrium, we then recast it to find the single-reservoir Massieu-Planck function Φ_1 in Eq. (11) of the main text. Though the system coupled with a single reservoir no longer obeys the Gibbs ensemble, we may regard the reservoir's parameters β and μ as local thermal parameters of this open system, because of the thermodynamic relation

$$N = \frac{\partial \Phi_1(\beta, \beta\mu)}{\partial(\beta\mu)} = \int_{-\infty}^{\infty} d\varepsilon \rho(\varepsilon) f(\varepsilon), \quad (\text{B1})$$

$$E = -\frac{\partial \Phi_1(\beta, \beta\mu)}{\partial\beta} = \int_{-\infty}^{\infty} d\varepsilon \rho(\varepsilon) \varepsilon f(\varepsilon). \quad (\text{B2})$$

Comparing Eq. (A2) and Eq. (11), we see the spectral function $\rho(\varepsilon)$ play a role of degeneracy at each energy shell. Physical quantities are expressed simply by replacing the summation $\sum_{\alpha}(\dots)$ by the energy integral $\int d\varepsilon \rho(\varepsilon)(\dots)$. Particularly, the local entropy $S = \Phi_1 + \beta(E - N\mu)$ becomes

$$S = -\int d\varepsilon \rho(\varepsilon) \{f(\varepsilon) \ln f(\varepsilon) \mp [1 \pm f(\varepsilon)] \ln [1 \pm f(\varepsilon)]\}. \quad (\text{B3})$$

This form of the entropy indicates that some entities having distribution $f(\varepsilon)$ are present at each energy shell, when put in the context of information theory. This is why we have introduced field operators at each energy shell in the main text.

APPENDIX C: ANALYTICAL FORM OF $\Phi_1(\beta, \beta\mu)$ AND REGULARIZATION

For the Lorentzian spectrum of Eq. (8), it is possible to obtain the analytical form of the single-reservoir Massieu-Planck function Φ_1 , hence the steady-state Φ_{ss} via Eq. (18). The function Φ_1 turns out to be divergent due to the zero-temperature contribution in the wide-band limit [58]. To suppress such divergence, we need to introduce a finite band width of the reservoir. The explicit form of Φ_1 is useful to connect several different expressions found in the literature; it also clarifies the nature of the divergence and shows directly that physical quantities are independent of the regularization of such divergence.

A central role is played by the following integral formula:

$$\begin{aligned} & \int_{-\infty}^{\infty} \frac{d\varepsilon}{(\varepsilon - \epsilon_d + i\gamma)(e^{\beta(\varepsilon - \mu)} \mp 1)} \\ &= \int_{-\infty}^{\infty} \frac{\mp \theta(\mu - \varepsilon) d\varepsilon}{\varepsilon - \epsilon_d + i\gamma} - \frac{d}{dz} \log_e \mathcal{F}(z), \end{aligned} \quad (\text{C1})$$

where the first term corresponds to the zero-temperature contribution while the second term, to the finite temperature. The latter can be evaluated explicitly (see [59], Sec. 3.415). We find it useful to express it by the complex function $\mathcal{F}(z)$

that is normalized by $\mathcal{F}(z) \rightarrow 1$ for large $|z|$:

$$\mathcal{F}(z) = \begin{cases} \frac{\Gamma(z) z^{\frac{1}{2}-z} e^{-z}}{\sqrt{2\pi}}, & (\text{B}), \\ \frac{\sqrt{2\pi} z^z e^{-z}}{\Gamma(\frac{1}{2}+z)}, & (\text{F}), \end{cases} \quad (\text{C2})$$

as a function of the dimensionless complex parameter

$$z = x + iy = \frac{\beta}{2\pi}(\gamma + i\xi), \quad \xi = \epsilon_d - \mu. \quad (\text{C3})$$

The first term on the right-hand side of Eq. (C1) is divergent, which we need to suppress by introducing finite band width of the reservoir,

$$\int_{-D}^{\infty} \frac{\mp \theta(\mu - \varepsilon) d\varepsilon}{\varepsilon - \epsilon_d + i\gamma} = \mp \log_e(z/z_D), \quad (\text{C4})$$

with the dimensionless cutoff $z_D = \beta[\gamma + i(D + \epsilon_d)]/2\pi$. Formula (C1) allows us to evaluate the average occupation number N and energy E in Eqs. (B1) and (B2) straightforwardly:

$$N(z) = \frac{1}{\pi} \text{Im} \left[\pm \log_e(z/z_D) + \frac{d}{dz} \log_e \mathcal{F}(z) \right], \quad (\text{C5})$$

$$= \begin{cases} -\frac{1}{2} + \frac{1}{\pi} \text{Im} \left[\psi(z) + \frac{1}{2z} \right], & (\text{B}), \\ \frac{1}{2} - \frac{1}{\pi} \text{Im} \left[\psi\left(\frac{1}{2} + z\right) \right], & (\text{F}), \end{cases} \quad (\text{C6})$$

$$E(z) = \frac{1}{\pi} \text{Im} \left[(\epsilon_d - i\gamma) \left(\pm \log_e(z/z_D) + \frac{d}{dz} \log_e \mathcal{F}(z) \right) \right], \quad (\text{C7})$$

$$= \begin{cases} \frac{1}{\pi} \text{Im} \left\{ (\epsilon_d - i\gamma) \left[\psi(z) + \frac{1}{2z} - \log_e z_D \right] \right\}, & (\text{B}), \\ -\frac{1}{\pi} \text{Im} \left\{ (\epsilon_d - i\gamma) \left[\psi\left(\frac{1}{2} + z\right) - \log_e z_D \right] \right\}, & (\text{F}). \end{cases} \quad (\text{C8})$$

It is noted that while the average energy E reduces to $\epsilon_d f(\epsilon_d)$ in the isolated limit $\gamma \rightarrow +0$, it diverges for any finite γ because of the zero-temperature contribution. Yet its finite-temperature contribution is well defined.

One can construct the function Φ_1 by integrating the above expression of N or E . Those forms suggest that we may write it as

$$e^{\Phi_1} = A |\mathcal{F}(z)|^2, \quad (\text{C9})$$

where A is a factor coming from the zero-temperature contribution. Seeing Φ_1 vanish at large y , one determines A as

$$\log_e A = \pm 2 \text{Re} \int_z^{z_D} dz' \log_e(z'/z_D) = |(z_D/z)^z|^{\mp 2}. \quad (\text{C10})$$

This leads to the single-reservoir Massieu-Planck function,

$$\Phi_1(\beta, \beta\mu) = \begin{cases} \ln \left| \frac{z^{\frac{1}{2}} \Gamma(z)}{\sqrt{2\pi} z_D^z e^{-z}} \right|^2, & (\text{B}), \\ \ln \left| \frac{\sqrt{2\pi} z_D^z e^{-z}}{\Gamma(\frac{1}{2}+z)} \right|^2, & (\text{F}). \end{cases} \quad (\text{C11})$$

It is always good to check the isolated limit $\gamma \rightarrow +0$. In this limit, we find

$$|\mathcal{F}(x + iy)|^2 \rightarrow e^{\pm\pi(|y|-y)}(1 \mp e^{-2\pi y})^{\mp 1}, \quad (\text{C12})$$

$$A \rightarrow e^{\pm\pi(y-|y|)}. \quad (\text{C13})$$

Hence $e^{\Phi_1} \rightarrow [1 \mp e^{-2\pi y}]^{\mp 1}$. This is nothing but the partition function of the isolated system in Eq. (A2).

In addition to N and E , we can obtain various thermodynamic quantities by differentiating $\Phi_1(\beta, \beta\mu)$. They take a simple form in terms of the function $\mathcal{F}(z)$. For instance, the entropy S becomes

$$S = \Phi_1 - \beta \frac{\partial \Phi_1}{\partial \beta} \Big|_{\mu} = 2 \operatorname{Re} \left[\left(1 - z \frac{d}{dz} \right) \log_e \mathcal{F}(z) \right], \quad (\text{C14})$$

$$= \begin{cases} 2 \operatorname{Re} \left[\log_e \left(\frac{\Gamma(z) z^{\frac{1}{2}}}{\sqrt{2\pi}} \right) + z - \frac{1}{2} - z\psi(z) \right], & (\text{B}) \\ 2 \operatorname{Re} \left[\log_e \left(\frac{\sqrt{2\pi}}{\Gamma(\frac{1}{2} + z)} \right) - z + z\psi(\frac{1}{2} + z) \right]. & (\text{F}) \end{cases} \quad (\text{C15})$$

The vanishing of S at zero temperature follows from the fact $\mathcal{F}(z) \rightarrow 1$ for large z . The specific heat C becomes

$$C = \beta^2 \frac{\partial^2 \Phi_1}{\partial \beta^2} \Big|_{\mu} = 2 \operatorname{Re} \left[z^2 \frac{d^2}{dz^2} \log_e \mathcal{F}(z) \right], \quad (\text{C16})$$

$$= \begin{cases} 2 \operatorname{Re} \left[-\frac{1}{2} - z + z^2 \psi'(z) \right], & (\text{B}) \\ 2 \operatorname{Re} \left[z - z^2 \psi'(\frac{1}{2} + z) \right]. & (\text{F}) \end{cases} \quad (\text{C17})$$

The result of specific heat for fermionic systems agrees with that of [23] (with $\xi = 0$), while for the bosonic systems, it agrees with that of the damped harmonic oscillator [26,27].

APPENDIX D: NONLINEAR CURRENT OF CHARGE, ENERGY, AND HEAT

One can also find analytical expressions of nonlinear currents of particle, energy, or heat; they are well-defined and independent of the cutoff. From the thermodynamic relation (1) [or equivalently from Eqs. (6) and (7)], we can write currents of particle and energy as

$$I_a = \frac{2}{\hbar} \sum_b \frac{\gamma_a \gamma_b}{\gamma} [N(z_a) - N(z_b)], \quad (\text{D1})$$

$$J_a = \frac{2}{\hbar} \sum_b \frac{\gamma_a \gamma_b}{\gamma} [E(z_a) - E(z_b)]. \quad (\text{D2})$$

Here $N(z_a)$ and $E(z_a)$ are defined by Eqs. (C5) and (C8) for each reservoir by choosing $z_a = (\beta_a/2\pi)(\gamma + i\epsilon_d - i\mu_a)$ with β_a and μ_a of the reservoir [see Eq. (C3)]. The difference of N or E is independent of cutoff. Indeed, we see

$$N(z_a) - N(z_b) = \frac{1}{\pi} \operatorname{Im} \left[\pm \log_e \left(\frac{z_a}{z_b} \right) + \frac{\mathcal{F}'(z)}{\mathcal{F}(z)} \Big|_{z_b}^{z_a} \right], \quad (\text{D3})$$

$$E(z_a) - E(z_b) = \frac{1}{\pi} \operatorname{Im} \left\{ (\epsilon_d - i\gamma) \left[\pm \log_e \left(\frac{\beta_b z_a}{\beta_a z_b} \right) + \frac{\mathcal{F}'(z)}{\mathcal{F}(z)} \Big|_{z_b}^{z_a} \right] \right\}. \quad (\text{D4})$$

Therefore the finite-temperature contributions are written in terms of the digamma function $\psi(z) = \Gamma'(z)/\Gamma(z)$ via Eq. (C2). Likewise, heat current $J_a^Q = J_a - \mu_a I_a$ from the reservoir a can be expressed analytically by using the above expressions.

APPENDIX E: DERIVATION OF EQ. (22) OR (23)

Assigning an affinity that is associated with each current turns out to be delicate, particularly in multiterminal settings. Because of current conservation, the inflow at the reservoir a must involve outflows to other reservoirs. In order to establish Eqs. (22) and (23), it is crucial to specify how one varies a relevant parameter by fixing others, as in equilibrium thermodynamics.

In the following, we choose to use x_a for $\beta_a \mu_a$ or $-\beta_a$, to describe particle or energy transport, while we will introduce y_a for affinities later. Adopting this notation, we write the steady-state Massieu-Planck function as $\Phi_{\text{ss}} = \sum_a (\gamma_a/\gamma) \Phi_1(x_a)$, where Φ_1 is defined by Eq. (11) in the text. Now we define the local parameter \bar{x} and its affinity y_a by

$$\bar{x} = \sum_{a=1}^N \frac{\gamma_a}{\gamma} x_a, \quad y_a = x_a - \bar{x}. \quad (\text{E1})$$

Affinities y_a satisfy the sum rule $\sum_a (\gamma_a/\gamma) y_a = 0$. This comes from the condition that any variation of affinities does not affect \bar{x} . In this way, we map the N parameters $\{x_a\}$ into \bar{x} and $\{y_a\}$, where we can eliminate one of $\{y_a\}$. Alternatively, we can express x_a as a function of \bar{x} and the differences of $\{y_a\}$ by

$$x_a = \bar{x} + \sum_{b=1}^N \left(\delta_{ab} - \frac{\gamma_b}{\gamma} \right) y_b = \bar{x} + \sum_{b=1}^N \frac{\gamma_b}{\gamma} (y_a - y_b). \quad (\text{E2})$$

This parametrization is singled out by requiring to fix \bar{x} by any variation of the N -independent parameters $\{y_1, \dots, y_N\}$. Though we can safely cross out one of $\{y_a\}$ at any moment, we prefer retaining all of them for a symmetrical reason. In either way, we can obtain the currents fulfilling the current conservation by varying y_a in the above.

Equation (22) or (23) can be derived by taking derivatives regarding \bar{x} or y_a by assuming \bar{x} and y_a are independent variables:

$$\bar{\mathcal{N}} = \frac{\partial \Phi_{\text{ss}}}{\partial \bar{x}} = \sum_{a=1}^N \frac{\gamma_a}{\gamma} \frac{\partial x_a}{\partial \bar{x}} \frac{\partial \Phi_1(x_a)}{\partial x_a} = \sum_{a=1}^N \frac{\gamma_a}{\gamma} \mathcal{N}_a, \quad (\text{E3})$$

$$\mathcal{I}_a = \frac{2}{\hbar} \frac{\partial \Phi_{\text{ss}}}{\partial y_a} = \frac{2\gamma}{\hbar} \sum_{b=1}^N \frac{\gamma_b}{\gamma} \frac{\partial x_b}{\partial y_a} \frac{\partial \Phi_1(x_b)}{\partial x_b}, \quad (\text{E4})$$

$$= \frac{2\gamma}{\hbar} \sum_{b=1}^N \frac{\gamma_b}{\gamma} \left(\delta_{ab} - \frac{\gamma_a}{\gamma} \right) \mathcal{N}_b = \frac{2}{\hbar} \sum_{b=1}^N \frac{\gamma_a \gamma_b}{\gamma} (\mathcal{N}_a - \mathcal{N}_b), \quad (\text{E5})$$

Here we assign, for particle transport, $\bar{\mathcal{N}} = \bar{N}$, $\mathcal{I}_a = I_a$, and $\mathcal{N}_a = N(z_a)$; for energy transport, $\bar{\mathcal{N}} = \bar{E}$, $\mathcal{I}_a = J_a$, and $\mathcal{N}_a = E(z_a)$. The current conservation $\sum_a \mathcal{I}_a = 0$ immediately follows from the above expression of \mathcal{I}_a .

Linear current \mathcal{I}_a at terminal a is proportional to $\gamma_a(x_a - \bar{x}) = \gamma_a y_a$. Choosing the parametrization (E2), it shows that positive affinity y_a plays a role in in-

ducing the inflow at the terminal a but the outflows at all the other terminals, in the linear-response regime.

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