Temperature and voltage measurement in quantum systems far from equilibrium

Abhay Shastry and Charles A. Stafford
Department of Physics, University of Arizona, 1118 East Fourth Street, Tucson, Arizona 85721, USA
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We show that a local measurement of temperature and voltage for a quantum system in steady state, arbitrarily far from equilibrium, with arbitrary interactions within the system, is unique when it exists. This is interpreted as a consequence of the second law of thermodynamics. We further derive a necessary and sufficient condition for the existence of a solution. In this regard, we find that a positive temperature solution exists whenever there is no net population inversion. However, when there is a net population inversion, we may characterize the system with a unique negative temperature. Voltage and temperature measurements are treated on an equal footing: They are simultaneously measured in a noninvasive manner, via a weakly coupled *thermoelectric probe*, defined by requiring vanishing charge *and* heat dissipation into the probe. Our results strongly suggest that a local temperature measurement without a simultaneous local voltage measurement, or vice versa, is a misleading characterization of the state of a nonequilibrium quantum electron system. These results provide a firm mathematical foundation for voltage and temperature measurements far from equilibrium.

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I. INTRODUCTION

Scanning probe microscopy [1–5] revolutionized the field of nanoscience and enabled the measurement of local thermodynamic observables such as voltage [6] and temperature [7] in nonequilibrium quantum systems. The ability to define local thermodynamic variables in a system far from equilibrium is of fundamental interest because it is a necessary step toward the construction of nonequilibrium thermodynamics [8–15]. Many experiments in mesoscopic electrical transport utilize voltage probes as circuit elements [16–19], and scanning potentiometers are now a mature technology [20–22], routinely achieving subangstrom spatial resolution to study a host of novel physical phenomena [23–26]. In contrast, scanning thermometry [7] has proven significantly more challenging [27], but is currently undergoing a rapid evolution toward nanometer resolution [28–31], leading to important insights into transport and dissipation at the nanoscale [32–36]. A fundamental challenge for theory is to develop a rigorous mathematical description of such local thermodynamic measurements. Until now, mainly operational definitions [12,37–46] have been advanced, leading to a competing panoply of often contradictory definitions of such basic observables as temperature and voltage.

The second law of thermodynamics is one of the cornerstones of physics. The origin of the second law was rooted in empirical observations in the early nineteenth century, and its theoretical explanation emerged with the gradual development of the statistical foundation of thermodynamics. The statistical basis of the second law places it in a league of its own, among the laws of physics. A quote on the subject, at once exalting and to the point, by the famous astrophysicist Sir Arthur Eddington reads as follows [47]: "The law that entropy always increases holds, I think, the supreme position among the laws of Nature. If someone points out to you that your pet theory of the universe is in disagreement with Maxwell's equations—then so much the worse for Maxwell's equations. If it is found to be contradicted by observation—well, these experimentalists do bungle things sometimes. But if your theory is found to be against the second law of thermodynamics I can give you no hope; there is nothing for it but to collapse in deepest humiliation." Any theory which purports to describe the measurement of temperature, voltage, or other thermodynamic parameters, must therefore satisfy this fundamental requirement, and as Eddington notes, regardless of the nature of microscopic interactions.

We examine statements of the second law of thermodynamics, accompanied with mathematical proofs, and their consequences, in the context of local noninvasive measurements of temperature and voltage in nonequilibrium quantum electron systems. We derive our results from very general considerations, i.e., for electron transport in steady state, arbitrarily far from equilibrium, and for arbitrary interactions within the quantum electron system. Our considerations apply to any system of fermions, charged or neutral. For noninteracting electrons, our results also hold for arbitrarily large (invasive) probe couplings. While our analyses in this article are presented in a theorem-proof format, their motivation draws from physical principles. We show that the uniqueness of the temperature and voltage measurement is a consequence of the second law of thermodynamics and that, in order to obtain a unique measurement, it is necessary to measure both temperature and voltage simultaneously. Simply put, this is because electrons carry both energy and charge.

In order to have a meaningful definition of temperature, the Hamiltonian must be bounded below ($\langle H \rangle \geqslant -c$ for some finite $c \in \mathbb{R}$). By the same token, a system can, in principle, exhibit negative temperatures if the energy averaged over the spectrum is bounded above ($\langle H \rangle \leqslant c$ for some finite $c \in \mathbb{R}$). These are well-known results in statistical physics, and we highlight their role in the context of local noninvasive measurements of temperature and voltage. We derive a condition, that is both necessary and sufficient, for the existence of a joint temperature and voltage measurement. This condition corresponds physically to a nonequilibrium system that does not exhibit local population inversion. We obtain also, as a corollary of the former condition, the result that nonequilibrium systems exhibiting local population inversion can be characterized with a negative temperature which is also unique. Population inversion is the working principle behind important Fermionic devices such as the maser and laser [48–51].

In this article we consider a probe that couples exclusively to the electronic degrees of freedom. Out of equilibrium, the temperature distributions of different microscopic degrees of freedom (e.g., electrons, phonons, nuclear spins) do not, in general, coincide, so that one has to distinguish between measurements of the electron temperature [37,38,44,52] and the lattice temperature [53,54]. This distinction is particularly acute in the extreme limit of elastic quantum transport [55], where electron and phonon temperatures are completely decoupled. It should be emphasized that the electrons within the system are free to undergo arbitrary interactions, e.g., with photons, phonons, other electrons, etc. However, *direct* heat transport into the probe via black-body radiation, phonons, etc. is excluded. Inclusion of these additional heat transfer processes into the probe leads to a temperature measurement that is simply a combination of the temperatures of the various microscopic degrees of freedom [45].

The article is organized as follows. We outline the formalism in Sec. II, and introduce a postulate that helps put our results on sound mathematical footing. In Sec. III we discuss our theory of local thermodynamic measurements, explain the idea behind noninvasive measurements, and also derive some useful expressions for further analysis. In Sec. IV we provide several statements of the second law of thermodynamics and show their relation to the uniqueness of temperature and voltage measurements. In Sec. V we start by defining certain useful quantities and proceed to derive the condition for the existence of a solution. We also discuss here the case of broadband probes in order to further illustrate the physical meaning behind our results, and conclude that probes operating in the broadband limit can be considered to be *ideal*. We consider the other extreme as well, i.e., narrowband probes and conclude that they are unsuitable for thermoelectric measurements. Our results are illustrated for a two-level system which is detailed in Sec. V D. We conclude with a summary of our central findings in Sec. VI, contrasting our approach to prior theoretical work, and discuss possible future directions. Some key results on the local properties of fermions in a nonequilibrium steady state are presented in Appendix A, which are needed in our analysis of the measurement problem. Appendix B provides a detailed analysis of the noninvasive-probe limit.

II. FORMALISM

We use the nonequilibrium Green's function formalism (NEGF) for describing the motion of electrons within a quantum conductor. A general expression for the nonequilibrium steady-state electrical current ($\nu = 0$) [56] and the electronic contribution to the heat current ($\nu = 1$) [57] flowing into a macroscopic electron reservoir P is

$$I_p^{(\nu)} = -\frac{i}{h}\int_{-\infty}^{\infty} d\omega(\omega-\mu_p)^\nu \,\mathrm{Tr}(\Gamma^p(\omega)\{G^<(\omega) + f_p(\omega)[G^>(\omega)-G^<(\omega)]\}), \tag{1}$$

where $\Gamma^p(\omega)$ is the tunneling width matrix describing the coupling of the probe to the system,

$$f_p(\omega) = \{1+\exp[(\omega-\mu_p)/k_B T_p]\}^{-1} \tag{2}$$

is the Fermi-Dirac distribution of the probe, and $G^<(\omega)$ and $G^>(\omega)$ are the Fourier transforms of the Keldysh "lesser" and "greater" Green's functions [58], describing the nonequilibrium electron and hole distributions within the system, respectively (see Appendix A for details). The spectral function of the system is $A(\omega) = [G^<(\omega) - G^>(\omega)]/2\pi i$.

Introducing the local nonequilibrium distribution function of the system, as sampled by the probe, defined by [13]

$$f_s(\omega) \equiv \frac{\mathrm{Tr}\{\Gamma^p(\omega)G^<(\omega)\}}{2\pi i\,\mathrm{Tr}\{\Gamma^p(\omega)A(\omega)\}}, \tag{3}$$

and the effective probe-system transmission function

$$\mathcal{T}_{ps}(\omega) = 2\pi\,\mathrm{Tr}\{\Gamma^p(\omega)A(\omega)\}, \tag{4}$$

Eq. (1) can be written in a form analogous to the two-terminal Landauer-Büttiker formula [13]:

$$I_p^{(\nu)} = \frac{1}{h}\int_{-\infty}^{\infty} d\omega(\omega-\mu_p)^\nu \mathcal{T}_{ps}(\omega)[f_s(\omega)-f_p(\omega)]. \tag{5}$$

We note that the expression in Eq. (5) for the steady-state currents flowing into a macroscopic (probe) reservoir coupled to a nonequilibrium quantum system is completely general, and allows for arbitrary interactions within the quantum system which is in an arbitrary nonequilibrium steady state. It is simply a rewriting of the fully general result of Eq. (1).

Since the spectral function $A(\omega)$ is positive-semidefinite and the probe-system coupling $\Gamma^p(\omega)$ is positive-semidefinite (see Appendix A), we note that

$$\begin{aligned}\mathrm{Tr}\{A(\omega)\Gamma(\omega)\} &= \mathrm{Tr}\{A(\omega)^{1/2}A^{1/2}(\omega)\Gamma(\omega)\}\\ &= \mathrm{Tr}\{A^{1/2}(\omega)\Gamma(\omega)A^{1/2}(\omega)\}\\ &\geqslant 0,\end{aligned} \tag{6}$$

where $A^{1/2}(\omega)$ is the positive-semidefinite square root of $A(\omega)$. $A^{1/2}(\omega)\Gamma(\omega)A^{1/2}(\omega)$ becomes positive-semidefinite when $A^{1/2}(\omega)$ and $\Gamma(\omega)$ are positive-semidefinite [59] and therefore we have

$$\mathcal{T}_{ps}(\omega) \geqslant 0, \quad \forall\omega\in\mathbb{R}. \tag{7}$$

We note that $f_s(\omega)$ satisfies the property of a distribution function, namely,

$$0 \leqslant f_s(\omega) \leqslant 1, \quad \forall\omega\in\mathbb{R}, \tag{8}$$

as shown in Appendix A. We start our analysis with the following postulate, and explain its physical significance.

Postulate 1. The local probe-system transmission function $\mathcal{T}_{ps} : \mathbb{R} \to [0,\infty)$ and the nonequilibrium distribution function $f_s : \mathbb{R} \to [0,1]$ are measurable over any interval $[a,b] \in \mathbb{R}$, and $\mathcal{T}_{ps}(\omega)$ satisfies

$$0 < \int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega) < \infty \tag{9}$$

and

$$\left|\int_{-\infty}^{\infty} d\omega\, \omega\mathcal{T}_{ps}(\omega)\right| < \infty. \tag{10}$$

The measurability of $\mathcal{T}_{ps}(\omega)$ and $f_s(\omega)$ is taken to lend meaning to the currents in Eq. (5). We point out that the finiteness of the two integrals given in Eqs. (9) and (10) is more relevant to our discussion of existence in Sec. V. Our result on uniqueness, as stated in Theorem 2, is somewhat stronger and requires only that the function $\mathcal{T}_{ps}(\omega)$ grow slower than exponentially for large values of energy (for $\omega \to \pm\infty$).

On physical grounds, the probe-sample transmission function $\mathcal{T}_{ps}(\omega)$ can be argued to have a compact support (nonzero only for some finite interval $[\omega_-,\omega_+] \subset \mathbb{R}$). It is easy to see that $\mathcal{T}_{ps}$ must have a lower bound ω_- such that $\mathcal{T}_{ps}(\omega) = 0\ \forall\ \omega < \omega_-$, since physical Hamiltonians must have a finite ground-state energy. However, for energies larger than the probe work function (ω_+), it can be argued that the particle will merely pass through the probe and not contribute to the steady state currents into the probe. $\mathcal{T}_{ps}(\omega)$ then has a compact support and satisfies Eqs. (9) and (10). In Sec. V we comment upon the limiting case where the measure of $\omega\mathcal{T}_{ps}(\omega)$ in Eq. (10) tends to infinity. The absolute value on the left-hand side in Eq. (10) is somewhat redundant since the limiting case must have $\omega_+ \to \infty$ while $\omega_- \to -\infty$ is ruled out based on the principle that any physical spectrum has a finite ground-state energy. We note that Eqs. (9) and (10) also imply

$$0 < \int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega) f_s(\omega), \quad \int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega) f_p(\omega) < \infty \tag{11}$$

and

$$\int_{-\infty}^{\infty} d\omega\ \omega \mathcal{T}_{ps}(\omega) f_s(\omega), \quad \int_{-\infty}^{\infty} d\omega\ \omega \mathcal{T}_{ps}(\omega) f_p(\omega) < \infty. \tag{12}$$

III. LOCAL MEASUREMENTS

The local voltage and temperature of a nonequilibrium quantum system, as measured by a scanning thermoelectric probe, is defined by the simultaneous conditions of vanishing net charge dissipation *and* vanishing net heat dissipation into the probe [13,15,44,45,52,60]:

$$I_p^{(\nu)} = 0, \quad \nu \in \{0,1\}, \tag{13}$$

where $\nu = 0,1$ correspond to the electron number current and the electronic contribution to the heat current, respectively. Equation (13) gives the conditions under which the probe is in local equilibrium with the sample, which is itself arbitrarily far from equilibrium.

We define the system's local temperature and voltage using a probe that is weakly coupled via a tunnel barrier (see Fig. 1). The other end of this scanning probe [4,5] is the macroscopic electron reservoir whose temperature and voltage are both adjusted until Eq. (13) is satisfied. A weakly coupled probe is a useful theoretical construction for our analysis, and the extension of our results beyond the weak-coupling limit is an open question. We explain the physical basis of weak coupling below, and derive some useful formulas.

Noninvasive measurements

When the coupling of the probe to the system is weak, we may take $\mathcal{T}_{ps}(\omega)$ in Eq. (4) and the local nonequilibrium distribution function $f_s(\omega)$ to be independent of the probe temperature T_p and chemical potential μ_p. While both $\mathcal{T}_{ps}(\omega)$ and $f_s(\omega)$ depend upon the local probe-system coupling in

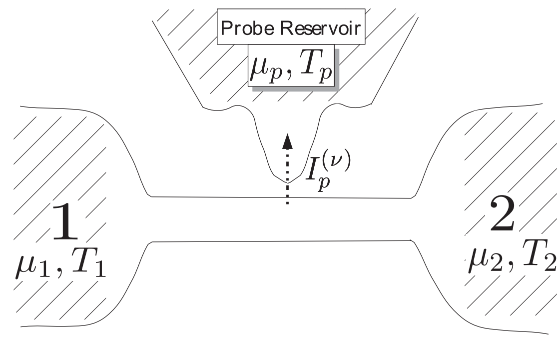

FIG. 1. Illustration of the measurement setup: The quantum conductor represented below is in a nonequilibrium steady state. A weakly coupled scanning tunneling probe noninvasively measures the local voltage (μ_p) and local temperature (T_p) *simultaneously*: By requiring both a vanishing net charge exchange ($I_p^{(0)} = 0$) *and* a vanishing net heat exchange ($I_p^{(1)} = 0$) with the system. The nonequilibrium steady state has been prepared, in this particular illustration, via the electrical and thermal bias of the strongly coupled reservoirs (1 and 2). The measurement method itself is completely general and does not depend upon (a) how such a nonequilibrium steady state is prepared, (b) how far from equilibrium the quantum electron system is driven, and (c) the nature of interactions within that system.

an obvious manner, the weak-coupling condition essentially implies that the nonequilibrium steady state of the system is unperturbed by the introduction of the probe terminal. The voltage and temperature of the probe itself play no role in preparing the nonequilibrium steady state. In other words, the probe does *not* drive the system but merely exchanges energy and particles across a weakly coupled tunnel barrier and constitutes a *noninvasive* measurement. For a precise analysis of the conditions necessary for a noninvasive probe, see Appendix B.

Given a system prepared in a certain nonequilibrium steady state (e.g., by a particular bias of the strongly coupled reservoirs), the currents given by Eq. (5) are functions of the probe Fermi-Dirac distribution specified by its temperature and chemical potential

$$I_p^{(\nu)} \equiv I_p^{(\nu)}(\mu_p, T_p). \tag{14}$$

It can be seen that the currents are continuous functions of $\mu_p \in (-\infty,\infty)$ and $T_p \in (0,\infty)$ with continuous gradient vector fields defined by

$$\nabla I_p^{(\nu)} \equiv \left(\frac{\partial I_p^{(\nu)}}{\partial \mu_p}, \frac{\partial I_p^{(\nu)}}{\partial T_p} \right). \tag{15}$$

With k_B set to unity, we compute the gradients of the currents using Eq. (5). We find the gradient of the number current to be

$$\nabla I_p^{(0)} = \left(-\mathcal{L}_{ps}^{(0)}, -\frac{\mathcal{L}_{ps}^{(1)}}{T_p} \right). \tag{16}$$

The gradient of the heat current reduces to

$$\nabla I_p^{(1)} = \left(-\mathcal{L}_{ps}^{(1)} - I_p^{(0)}, -\frac{\mathcal{L}_{ps}^{(2)}}{T_p} \right), \tag{17}$$

where we define the response coefficients $\mathcal{L}_{ps}^{(\nu)}$ as

$$\mathcal{L}_{ps}^{(\nu)} \equiv \mathcal{L}_{ps}^{(\nu)}(\mu_p,T_p) = \frac{1}{h}\int_{-\infty}^{\infty} d\omega(\omega-\mu_p)^\nu \mathcal{T}_{ps}(\omega)\left(-\frac{\partial f_p}{\partial \omega}\right), \quad (18)$$

which are easily seen to be finite [61].

Although the coefficients $\mathcal{L}_{ps}^{(\nu)}$ formally resemble the Onsager linear-response coefficients [62] of an elastic quantum conductor [63], it is very important to note that we do *not* make the assumptions of linear response, time-reversal symmetry, local equilibrium, or elastic transport in the above definition of $\mathcal{L}_{ps}^{(\nu)}$: The system itself may be arbitrarily far from equilibrium with arbitrary inelastic scattering processes. The coefficients above appear naturally when we calculate the gradient fields defined by Eq. (15), and the gradient operator is of course given by the first derivatives. Our main results follow from an analysis of the properties of these gradient fields.

IV. UNIQUENESS AND THE SECOND LAW

We now turn to one of the central problems which we set out to address: $I_p^{(\nu)}(\mu_p,T_p)=0$, with $\nu=\{0,1\}$, is a system of coupled nonlinear equations in two variables that defines our local voltage and temperature measurement. There is no *a priori* reason to expect a unique solution, if a solution exists at all. We begin the section with statements of the second law of thermodynamics, and conclude by showing that the uniqueness of the measurement emerges as a consequence.

A. Statements of the second law

We note that $\forall\, \mu_p \in (-\infty,\infty)$ and $T_p \in (0,\infty)$,

$$\mathcal{L}_{ps}^{(0)}(\mu_p,T_p) > 0, \qquad \mathcal{L}_{ps}^{(2)}(\mu_p,T_p) > 0, \quad (19)$$

since $\mathcal{T}_{ps}(\omega) \geqslant 0$, and the measure of $\mathcal{T}_{ps}(\omega)$ and the Fermi-function derivative are both nonzero and strictly positive. This leads to two statements of the second law of thermodynamics, related to the Clausius statement, which are presented in the following two lemmas. The idea is to choose the correct contour for each case, and to evaluate the line integral over the current gradients in Eqs. (16) and (17). A cursory glance at the number current gradient in Eq. (16) suggests that the contour should be defined over a constant temperature, while the heat current gradient in Eq. (17) suggests a line integral over a constant voltage contour.

Lemma 1. The number current contour defined by $I_p^{(0)}(\mu_p,T_p)=0$ exists for all $T_p \in (0,\infty)$ and defines a function $M:(0,\infty)\to\mathbb{R}$ where $\mu_p = M(T_p)$, such that the second law of thermodynamics is obeyed:

$$I_p^{(0)}(\mu_p',T_p) > 0, \quad \text{if} \quad \mu_p' < M(T_p) \quad \text{and} \qquad I_p^{(0)}(\mu_p',T_p) < 0, \quad \text{if} \quad \mu_p' > M(T_p). \quad (20)$$

Proof. We first show that $I^{(0)}(\mu_p,T_p)=0$ is satisfied for all $T_p \in (0,\infty)$. For any $T_p \in (0,\infty)$, we

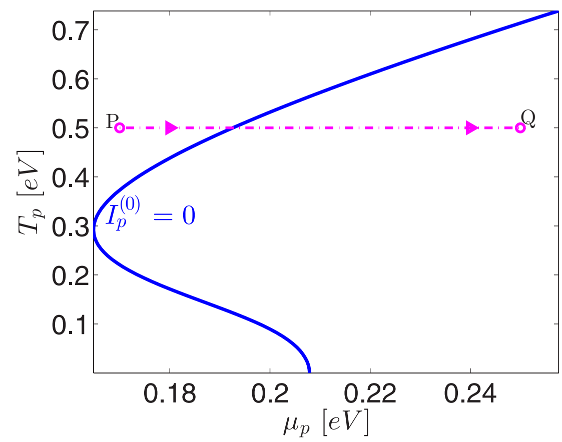

FIG. 2. Illustration of Lemma 1: The contour PQ shown in magenta cuts the number current contour $I_p^{(0)}=0$ (or any $I_p^{(0)}=$ const.) exactly once. The contour line from P to Q is at a constant temperature ($T_p=$ const.), and illustrates the Clausius statement: The number current is monotonically decreasing along PQ. The system and bias conditions are detailed in Sec. V D.

have

$$\lim_{\mu_p\to-\infty} I^{(0)}(\mu_p,T_p) = \frac{1}{h}\int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega) \times [f_s(\omega) - \lim_{\mu_p\to-\infty} f_p(\omega)] = \frac{1}{h}\int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega) f_s(\omega) > 0 \quad (21)$$

and

$$\lim_{\mu_p\to\infty} I^{(0)}(\mu_p,T_p) = \frac{1}{h}\int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega)[f_s(\omega) - \lim_{\mu_p\to\infty} f_p(\omega)] = \frac{1}{h}\int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega)[f_s(\omega)-1] < 0. \quad (22)$$

This ensures at least one solution due to the continuity of the currents, but does not ensure uniqueness.

We note that $I_p^{(0)}$ is monotonically decreasing along $\mathbf{dl} = (d\mu_p,0)$,

$$\Delta I_p^{(0)} = \int_{\mu_p}^{\mu_p'} \nabla I_p^{(0)} \cdot \mathbf{dl} = \int_{\mu_p}^{\mu_p'} -\mathcal{L}_{ps}^{(0)} d\mu_p \quad (23)$$

due to the fact that $\mathcal{L}_{ps}^{(0)}$ is positive, and more explicitly:

$$\Delta I_p^{(0)} = \frac{1}{h}\int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega)[f_p(\mu_p,T_p;\omega) - f_p(\mu_p',T_p;\omega)] \; > 0, \;\text{if}\; \mu_p' < \mu_p, \quad < 0, \;\text{if}\; \mu_p' > \mu_p. \quad (24)$$

This implies the existence of a unique solution to $I_p^{(0)}(\mu_p,T_p)=0$ for every $T_p \in (0,\infty)$ which we denote by $\mu_p = M(T_p)$, and Eq. (20) is implied by Eq. (24). ∎

We also note that the number current $[\mu_p = M(T_p)]$ contour is vertical when the temperature approaches absolute zero, as shown in Fig. 2, since $\mathcal{L}_{ps}^{(1)}/T_p \to 0$ as $T_p \to 0$, and implies a vanishing Seebeck coefficient for the probe-system junction near absolute zero.

An "ideal potentiometer" was initially proposed [37] by merely requiring $I_p^{(0)} = 0$. Subsequently, Büttiker [64,65] clarified that this definition holds only near absolute zero due to the absence of thermoelectric corrections. Such a voltage probe determines the voltage uniquely at zero temperature in the linear response regime, and is relevant for experiments in mesoscopic circuits [16–19] which are carried out at cryogenic temperatures. However, at higher temperatures and/or larger bias voltages, where the sample may be heated by both the Joule and Peltier effects, thermoelectric corrections to voltage measurements must be considered. Indeed, Bergfield and Stafford [60] argue that an *ideal voltage probe* must be required to equilibrate thermally with the system ($I_p^{(1)} = 0$), without which "a voltage will develop across the system-probe junction due to the *Seebeck effect*."

Voltage probes have been used extensively in the theoretical literature to mimic the effects of various scattering processes, such as inelastic scattering [64,66–70] and dephasing [71–73] in mesoscopic systems. A modern variation of Büttiker's voltage probe, additionally requiring that the probe exchange no heat current, has been used to model inelastic scattering in quantum transport problems at finite temperature [39,42,74–76]. The probe technique, as a model for scattering, has also been extensively studied beyond the linear response regime [77–79].

Lemma 1 implies that a "voltage probe" (defined only by $I_p^{(0)} = 0$) requires the simultaneous specification of a probe temperature T_p so that $\mu_p = M(T_p)$ is uniquely determined. Figure 2 illustrates that the measured voltage shows a large dependence on the probe temperature. Therefore, it is important to define a simultaneous temperature measurement by imposing $I_p^{(1)}(\mu_p,T_p) = 0$.

Lemma 2. The heat current contour defined by $I_p^{(1)}(\mu_p,T_p) = c$, where c is some constant, obeys the second law of thermodynamics, namely,

$$\begin{aligned} I_p^{(1)}(\mu_p,T_p') &> c, \quad \text{if} \quad T_p' < T_p, \\ &< c, \quad \text{if} \quad T_p' > T_p. \end{aligned} \tag{25}$$

Proof. We follow an analogous argument to Lemma 1, and show the monotonicity of $I_p^{(1)}(\mu_p,T_p)$ along a certain contour in the μ_p-T_p plane. Naturally, the contour we choose is along a fixed μ_p [cf. Eq. (17)] since we know that $\mathcal{L}_{ps}^{(2)}$ is positive. Therefore we have $\Delta I_p^{(1)} = I_p^{(1)}(\mu_p,T_p') - I_p^{(1)}(\mu_p,T_p) = \int_{T_p}^{T_p'} \nabla I_p^{(1)} \cdot \mathbf{dl}$, where $\mathbf{dl} = (0,dT_p)$ and explicitly,

$$\begin{aligned} \Delta I_p^{(1)} &= \frac{1}{h}\int_{-\infty}^{\infty} d\omega(\omega-\mu_p)\mathcal{T}_{ps}(\omega) \\ &\quad \times [f_p(\mu_p,T_p;\omega) - f_p(\mu_p,T_p';\omega)] \\ &> 0, \quad \text{if} \quad T_p' < T_p, \\ &< 0, \quad \text{if} \quad T_p' > T_p. \end{aligned} \tag{26}$$

This implies Eq. (25). ■

We stated Lemma 2 with a constant c [80], not necessarily $c = 0$, unlike Lemma 1. This is because we do *not a priori* know whether the contour $I_p^{(1)} = 0$ exists, and we derive a necessary and sufficient condition for its existence in Sec V.

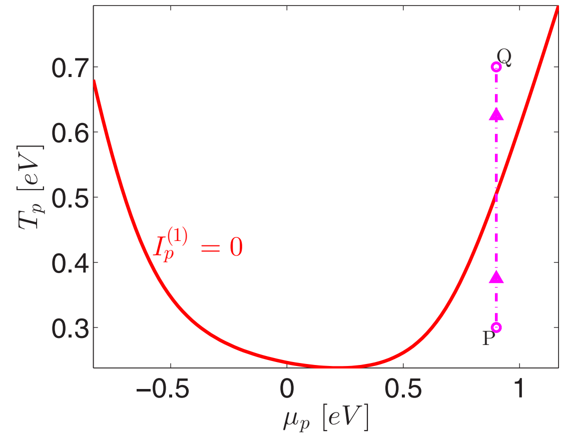

FIG. 3. Illustration of Lemma 2: The contour PQ shown in magenta cuts $I_p^{(1)} = 0$ (or any $I_p^{(1)} =$ const.) exactly once. Contour PQ is defined along constant voltage $\mu_p =$ const., and illustrates the Clausius statement: The heat current is monotonically decreasing along PQ. The system and bias conditions are detailed in Sec. V D.

Analogous to Lemma 1, Lemma 2 implies that a "temperature probe" [37] (defined only by $I_p^{(1)} = 0$) requires the simultaneous specification of a probe voltage μ_p so that the temperature $T_p = \tau_0(\mu_p)$ (cf. [80]) is uniquely determined. Figure 3 illustrates that the measured temperature shows a large dependence on the probe voltage. Therefore, it becomes important to simultaneously measure the voltage by imposing $I_p^{(0)} = 0$. If the temperature probe is not allowed to equilibrate electrically with the system, then a temperature difference will build up across the probe-system junction due to the *Peltier effect*, leading to an error in the temperature measurement.

Clearly, depending upon the probe voltage, the temperature probe could measure any of a range of values, rendering the measurement somewhat meaningless (see Fig. 3). Analogously, the "voltage probe" could measure any of a range of values depending upon the probe temperature (see Fig. 2). *Thermoelectric probes* (also referred to as dual probes and voltage-temperature probes) treat temperature and voltage measurements on an equal footing, and implicitly account for the thermoelectric corrections exactly. Only such a dual probe is in *both thermal and electrical equilibrium* with the system being measured, and therefore yields an unbiased measurement of both quantities. A mathematical proof of the uniqueness of a voltage and temperature measurement is therefore of fundamental importance.

We may also deduce that $T_p = 0$ cannot be obtained as a measurement outcome since

$$\begin{aligned} \lim_{T_p\to 0} I_p^{(1)}(\mu_p,T_p) &= \int_{-\infty}^{\infty} d\omega\,(\omega-\mu_p)\mathcal{T}_{ps}(\omega) \\ &\quad \times \Big[f_s(\omega) - \lim_{T_p\to 0} f_p(\mu_p,T_p)\Big] \\ &= \int_{-\infty}^{\mu_p} d\omega\,(\omega-\mu_p)\mathcal{T}_{ps}(\omega)[f_s(\omega)-1] \\ &\quad + \int_{\mu_p}^{\infty} d\omega\,(\omega-\mu_p)\mathcal{T}_{ps}(\omega)f_s(\omega) \\ &> 0, \end{aligned} \tag{27}$$

consistent with the third law of thermodynamics. However, temperatures arbitrarily close to absolute zero are, in principle, possible [15].

Lemmas 1 and 2 may be interpreted in terms of the Clausius statement of the second law [81]: "No process is possible whose sole effect is to transfer heat from a colder body to a warmer body." Lemma 2 gives us the direction in which heat will flow [cf. Eq. (26)] when the probe is biased away from the point of thermal equilibrium with the system, $I_p^{(1)}(\mu_p,T_p)=0$: whenever the probe is hotter than the temperature corresponding to thermal equilibrium, with the chemical potential held constant, heat flows *out* of the probe and vice versa. Similarly, Lemma 1 gives us the direction in which particle flow occurs when the probe is biased away from the point of electrical equilibrium, $I_p^{(0)}(\mu_p,T_p)=0$: whenever the probe is at a higher chemical potential than the one corresponding to electrical equilibrium, with temperature held constant, particles flow *out* of the probe and vice versa. Here we refer to electrical ($\nu=0$, Lemma 1) and thermal ($\nu=1$, Lemma 2) equilibration of the probe with the system under the local exchange of particles and energy. The system itself may be arbitrarily far from equilibrium, and may possess no local equilibrium.

The problem of a unique measurement of a voltage probe (defined only by $I_p^{(0)}=0$), or a temperature probe (defined only by $I_p^{(1)}=0$) has been attempted previously by Jacquet and Pillet [12] for transport beyond linear response, and to our knowledge is the only work in this direction. However, in Ref. [12], the bias conditions considered are quite restrictive and the result assumes noninteracting electrons. Lemmas 1 and 2, respectively, generalize the result to arbitrary bias conditions, and arbitrary interactions within a quantum electron system while also providing a useful insight via the Clausius statement of the second law of thermodynamics. However, the question we would like to answer in this article pertains to the uniqueness of a *thermoelectric probe* measurement, defined by both $I_p^{(0)}=0$ and $I_p^{(1)}=0$. A result for such dual probes has been obtained only in the linear response regime and for noninteracting electrons [39].

Theorem 1. The coefficients $\mathcal{L}_{ps}^{(\nu)}$ satisfy the inequality

$$\mathcal{L}_{ps}^{(0)}\mathcal{L}_{ps}^{(2)}-\left(\mathcal{L}_{ps}^{(1)}\right)^2>0. \tag{28}$$

Proof. We may define functions $g(\omega)$ and $h(\omega)$ as

$$g(\omega)=\sqrt{\mathcal{T}_{ps}(\omega)\left(-\frac{\partial f_p}{\partial\omega}\right)} \tag{29}$$

and

$$h(\omega)=(\omega-\mu_p)\sqrt{\mathcal{T}_{ps}(\omega)\left(-\frac{\partial f_p}{\partial\omega}\right)}. \tag{30}$$

We note that $g(\omega)$ and $h(\omega)$ belong to $\mathbf{L}^2(\mathbb{R})$ [61]. Noting that g and h are real, we apply the Cauchy-Schwarz inequality

$$\left|\int_{-\infty}^{\infty}d\omega g(\omega)h(\omega)\right|^2\leqslant\int_{-\infty}^{\infty}d\omega|g(\omega)|^2\int_{-\infty}^{\infty}d\omega|h(\omega)|^2. \tag{31}$$

The integral appearing on the left-hand side (lhs) is $\mathcal{L}_{ps}^{(1)}$, while on the right-hand side (rhs) we have the product of $\mathcal{L}_{ps}^{(0)}$ and $\mathcal{L}_{ps}^{(2)}$, respectively. We drop the absolute value on the lhs by noting that $\mathcal{L}_{ps}^{(1)}$ is real and write

$$\left(\mathcal{L}_{ps}^{(1)}\right)^2\leqslant\mathcal{L}_{ps}^{(0)}\mathcal{L}_{ps}^{(2)}. \tag{32}$$

We drop the equality case above by noting that g and h are linearly independent except for the trivial case when $\mathcal{T}_{ps}(\omega)=0\ \forall\omega$, or when the probe coupling is narrowband $[\mathcal{T}_{ps}(\omega)=\bar{\gamma}\delta(\omega-\omega_0)]$ which we discuss in Sec. V C. ■

The proof above can be easily extended to show the positive-definiteness of the linear response matrices [62] widely used for elastic transport calculations (e.g., in Refs. [63,82]). Theorem 1 implies a positive thermal conductance (see, e.g., Ref. [82]), which is necessary for positive entropy production consistent with the second law of thermodynamics.

B. Uniqueness

Theorem 2. The local temperature and voltage of a nonequilibrium quantum system, measured by a *thermoelectric probe*, is unique when it exists.

Proof. The tangent vectors $\mathbf{t}^{(\nu)}$ for $I_p^{(\nu)}$ are along

$$\mathbf{t}^{(0)}=\left(-\frac{\mathcal{L}_{ps}^{(1)}}{T_p},\mathcal{L}_{ps}^{(0)}\right) \tag{33}$$

and

$$\begin{aligned}\mathbf{t}^{(1)}&=\left(\frac{\mathcal{L}_{ps}^{(2)}}{T_p},-\mathcal{L}_{ps}^{(1)}-I_p^{(0)}\right)\\&=\left(\frac{\mathcal{L}_{ps}^{(2)}}{T_p},-\mathcal{L}_{ps}^{(1)}\right),\quad\text{if}\quad I_p^{(0)}=0,\end{aligned} \tag{34}$$

respectively, such that we have

$$\int_{s_1}^{s_2}ds\frac{\mathbf{t}^{(\nu)}\cdot\nabla I_p^{(\nu)}}{|\mathbf{t}^{(\nu)}|}=0, \tag{35}$$

where s is a scalar that labels points along the contour $I_p^{(\nu)}=$ const.

We now compute the change in $I_p^{(1)}$ along the contour $I_p^{(0)}=0$. The points along $I_p^{(0)}=0$ are labeled by the continuous parameter ξ such that $\mu_p=\mu_p(\xi)$ and $T_p=T_p(\xi)$. ξ is chosen to be increasing with increasing temperature. The change $\Delta I_p^{(1)}$ becomes

$$\begin{aligned}\Delta I_p^{(1)}&=\int_{\xi_1}^{\xi_2}d\xi\frac{\mathbf{t}^{(0)}\cdot\nabla I_p^{(1)}}{|\mathbf{t}^{(0)}|}\\&=\int_{\xi_1}^{\xi_2}d\xi\frac{1}{|\mathbf{t}^{(0)}|T_p}\left[\left(\mathcal{L}_{ps}^{(1)}\right)^2-\mathcal{L}_{ps}^{(0)}\mathcal{L}_{ps}^{(2)}\right]\\&>0\quad\text{if}\quad\xi_2<\xi_1,\\&<0\quad\text{if}\quad\xi_2>\xi_1,\end{aligned} \tag{36}$$

due to Theorem 1. Therefore $I_p^{(1)}=0$ (or for that matter $I_p^{(1)}=c$, for any c) is satisfied at most at a single point along $I_p^{(0)}=0$. ■

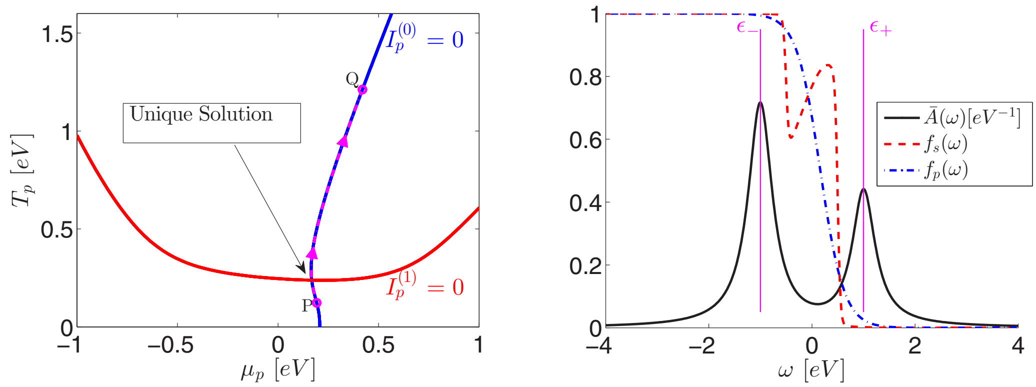

FIG. 4. Left panel: Illustration of Theorem 2 for positive temperatures. The contour PQ along $I_p^{(0)} = 0$ (shown in blue) cuts the contour $I_p^{(1)} = 0$ (shown in red) exactly once. Contour PQ illustrates a certain statement of the second law of thermodynamics: The heat current is monotonically decreasing along PQ (thus implying uniqueness). Right panel: The local spectrum sampled by the probe $\bar{A}(\omega)$ (black), the nonequilibrium distribution function $f_s(\omega)$ (red), and the probe Fermi-Dirac distribution $f_p(\omega)$ (blue) corresponding to the unique solution in the left panel. The resonances in the spectrum $\bar{A}(\omega)$ correspond to the eigenstates of the closed two-level Hamiltonian (see Sec. V D) $\epsilon_\pm = \pm 1$ shown in magenta. The Fermi-Dirac distribution is monotonically decreasing with energy, and corresponds to a situation with positive temperature (no net population inversion). The necessary and sufficient condition for the existence of a positive temperature solution is stated in Theorem 3.

Theorem 1 is a form of the second law of thermodynamics that gives us the direction in which the heat current flows along the contour $I_p^{(0)} = 0$ [cf. Eq. (36)]. The heat current $I_p^{(1)}$ decreases monotonically along the contour $I_p^{(0)} = 0$. Therefore we may find only one point along $I_p^{(0)} = 0$ that also satisfies $I_p^{(1)} = 0$, which implies a unique solution to Eq. (13) when it exists.

Indeed, Onsager points out in his 1931 paper [62] that for positive entropy production, the linear response matrix will have to be positive-definite (which translates to our condition in Theorem 1). However, that analysis rests upon the assumption of linear response near equilibrium. Our result in Theorem 1 does not require such a condition for the nonequilibrium state of the system, but instead emerges out of the analysis of the currents flowing into a weakly coupled probe. In addition, we obtain a strict mathematical proof of Theorem 1. We point out that Theorem 1 holds even when the physically expected Postulate 1 fails, making the uniqueness result in Theorem 2 very general [61].

V. EXISTENCE

A unique local measurement of temperature and voltage is only part of our main problem. An equally important part is to derive the conditions for the existence of a solution. The main idea behind this analysis is to follow the number current contour $I_p^{(0)} = 0$ and ask what happens to the heat current $I_p^{(1)}$ as we traverse towards higher and higher temperatures $T_p \to \infty$. We noted that near $T_p = 0$, the heat current into the probe must be positive, consistent with the third law of thermodynamics [cf. Eq. (27)]. Since we know that the heat current is monotonically decreasing along the number current contour (Theorem 2), we could guess whether or not a solution occurs depending upon the asymptotic value of the heat current along that contour as $T_p \to \infty$. In this way, we find a necessary and sufficient condition for the existence of a solution while analyzing the problem for positive temperatures (see Fig. 4 for an illustration of this case). On the other hand, when this condition is not met, one can immediately prove that a negative temperature must satisfy the measurement condition $I_p^{(\nu)} = 0,\ \nu = \{0,1\}$. This latter condition corresponds to a system exhibiting local population inversion which leads to negative temperature [83] solutions, as illustrated in Fig. 5.

Our results here are again completely general and are valid for electron systems with arbitrary interactions, arbitrary steady state bias conditions, and for any weakly coupled probe. However, our analysis here leads us to demarcate between two extremes of the probe-system coupling. We conclude that an *ideal probe* is one which operates in the *broadband limit*. A measurement by such a probe depends only on the properties of the system that it couples to, and is independent of the spectral properties of the probe itself. The broadband limit lends itself to an easier physical interpretation of the population inversion condition as well, and we discuss this important limit in Sec. V B. The other extreme is that of a *narrowband probe* which is capable of probing the system at just one value of energy, leading to a nonunique measurement (see also the proof of Theorem 1), and is discussed in Sec. V C. Only this pathological case leads to an exception to Theorem 2.

The simplest system which could, in principle, exhibit population inversion is a two-level system. Therefore, our results, including that of the previous section, have been illustrated by using a two-level system. The details of the nonequilibrium two-level system and its coupling to the thermoelectric probe are given in Sec. V D.

Our analysis starts with a rearrangement of the currents given by Eq. (5) and a restatement of the measurement condition [cf. Eq. (13)] in terms of energy currents, and we also define some useful quantities along the way. We may rewrite the number current in Eq. (5) as

$$I_p^{(0)} = \langle \dot{N} \rangle|_{f_s} - \langle \dot{N} \rangle|_{f_p}, \tag{37}$$

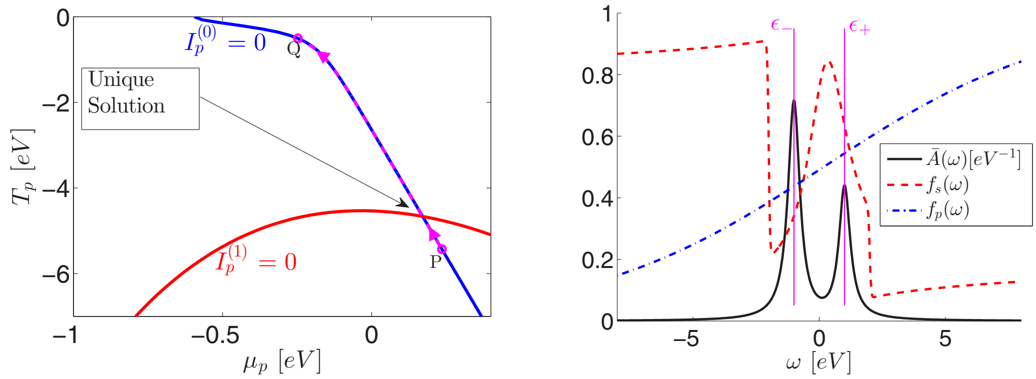

FIG. 5. Left panel: Illustration of Theorem 2 for negative temperatures. The contour PQ along $I_p^{(0)} = 0$ (shown in blue) cuts the contour $I_p^{(1)} = 0$ (shown in red) exactly once. Contour PQ illustrates a certain statement of the second law of thermodynamics: The heat current is monotonically decreasing along PQ (thus implying uniqueness). Right panel: The local spectrum sampled by the probe $\bar{A}(\omega)$ (black, and nearly unchanged from Fig. 4), the nonequilibrium distribution function $f_s(\omega)$ (red), and the probe Fermi-Dirac distribution $f_p(\omega)$ (blue) which corresponds to the unique solution (shown in the left panel). The resonances in the spectrum $\bar{A}(\omega)$ correspond to the eigenstates of the closed two-level Hamiltonian (see Sec. V D) $\epsilon_\pm = \pm 1$ shown in magenta. The system has a net population inversion, satisfying the conditions of Corollary 3.1, and the probe Fermi-Dirac distribution is monotonically increasing with energy, corresponding to a negative temperature.

where

$$\langle \dot{N} \rangle|_{f_s} \equiv \frac{1}{h} \int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega) f_s(\omega), \tag{38}$$

and similarly

$$\langle \dot{N} \rangle|_{f_p} \equiv \frac{1}{h} \int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega) f_p(\omega). \tag{39}$$

The quantity $\langle \dot{N} \rangle|_{f_s}$ is the rate of particle flow into the probe from the system, while $\langle \dot{N} \rangle|_{f_p}$ gives the rate of particle flow out of the probe and into the system.

Similarly, the rate of energy flow into the probe from the system is

$$\langle \dot{E} \rangle|_{f_s} \equiv \frac{1}{h} \int_{-\infty}^{\infty} d\omega\, \omega \mathcal{T}_{ps}(\omega) f_s(\omega), \tag{40}$$

while

$$\langle \dot{E} \rangle|_{f_p} \equiv \frac{1}{h} \int_{-\infty}^{\infty} d\omega\, \omega \mathcal{T}_{ps}(\omega) f_p(\omega) \tag{41}$$

gives the rate of energy outflux from the probe back into the system. The net energy current flowing into the probe is given by $I_p^E = \langle \dot{E} \rangle|_{f_s} - \langle \dot{E} \rangle|_{f_p}$.

The local equilibration conditions in Eq. (13) now become

$$\begin{aligned} \langle \dot{N} \rangle|_{f_p} &= \langle \dot{N} \rangle|_{f_s}, \\ \langle \dot{E} \rangle|_{f_p} &= \langle \dot{E} \rangle|_{f_s}. \end{aligned} \tag{42}$$

The equation for the rate of energy flow above is equivalent to the condition $I_p^{(1)} = 0$ when $I_p^{(0)} = 0$ since

$$I_p^E(\mu_p, T_p) \equiv \langle \dot{E} \rangle|_{f_s} - \langle \dot{E} \rangle|_{f_p} = I_p^{(1)} + \mu_p I_p^{(0)}. \tag{43}$$

The lhs in Eq. (42) depends upon the probe parameters (temperature and voltage) while the rhs is fixed for a given nonequilibrium system with a given local distribution function $f_s(\omega)$. The probe measures the appropriate voltage and temperature when it exchanges no net charge and energy with the system.

We may introduce a characteristic rate of particle flow [cf. Eq. (9)] as

$$\langle \dot{N} \rangle|_{f\equiv 1} = \frac{1}{h} \int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega) \equiv \frac{\gamma_p}{\hbar}. \tag{44}$$

This leads to the following inequalities:

$$\begin{aligned} 0 < \langle \dot{N} \rangle|_{f_s} &< \frac{\gamma_p}{\hbar}, \\ 0 < \langle \dot{N} \rangle|_{f_p} &< \frac{\gamma_p}{\hbar}. \end{aligned} \tag{45}$$

The lhs in the inequality for $\langle \dot{N} \rangle|_{f_s}$ above excludes $f_s(\omega) \equiv 0$ while the rhs excludes $f_s(\omega) = 1\ \forall \omega \in \mathbb{R}$, and we retain the strict inequalities imposed by Eq. (45) [see also Eqs. (11) and (12) and the preceding discussion].

We similarly introduce a characteristic rate for the energy flow between the system and probe:

$$\begin{aligned} \langle \dot{E} \rangle|_{f\equiv 1} &= \frac{1}{h} \int_{-\infty}^{\infty} d\omega\, \omega \mathcal{T}_{ps}(\omega) \\ &\equiv \frac{\gamma_p}{\hbar} \omega_c, \end{aligned} \tag{46}$$

where $\omega_c < \infty$ (due to Postulate 1) can be interpreted as the centroid of the probe-sample transmission function. We find that $\omega_c \to \infty$ necessarily implies a positive temperature solution. We remind the reader that $\omega_c \to -\infty$ is physically impossible due to the principle that any physical system must have a lower bound for the energy ($\langle H \rangle \geqslant -c$ for some finite $c \in \mathbb{R}$).

The quantities $\langle \dot{N} \rangle|_{f_s}$, $\langle \dot{N} \rangle|_{f_p}$, $\langle \dot{N} \rangle|_{f\equiv 1}$, $\langle \dot{E} \rangle|_{f_s}$, $\langle \dot{E} \rangle|_{f_p}$, $\langle \dot{E} \rangle|_{f\equiv 1}$ are all finite due to Postulate 1 [cf. Eqs. (9)–(12)].

A. Asymptotic properties, and conditions for the existence of a solution

Traversing along $I_p^{(0)} = 0$ results in a monotonically *decreasing* heat current $I_p^{(1)}$ (Theorem 2). Here we traverse the contour from low temperatures ($T_p \to 0$) to higher temperatures ($T_p \to \infty$) as discussed in Theorem 2. This implies a monotonically *increasing* $\langle \dot{E}\rangle|_{f_p}$ due to Eq. (43). We proceed to calculate the asymptotic value of $\langle \dot{E}\rangle|_{f_p}$ along the number current contour.

Let the asymptotic scaling of $\mu_p = M(T_p)$ defined by the contour $I_p^{(0)}(\mu_p,T_p) = 0$ (Lemma 1) be

$$\lim_{T_p\to\infty} \frac{M(T_p)}{T_p} = \Lambda. \quad (47)$$

We use the above limiting value to calculate $\langle \dot{N}\rangle|_{f_p}$ along the contour $\mu_p = M(T_p)$:

$$\begin{aligned}\lim_{T_p\to\infty}\langle \dot{N}\rangle|_{f_p} &= \frac{1}{h}\int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega) \lim_{T_p\to\infty}\frac{1}{1+\exp\left(\frac{\omega-M(T_p)}{T_p}\right)}\\ &= \frac{1}{h}\int_{-\infty}^{\infty} d\omega \mathcal{T}_{ps}(\omega)\frac{1}{1+\exp(-\Lambda)}\\ &= \frac{1}{1+\exp(-\Lambda)}\frac{\gamma_p}{\hbar}. \end{aligned} \quad (48)$$

The above limiting value satisfies the inequality in Eq. (45) for any $\Lambda \in \mathbb{R}$. The points on the contour satisfy $\langle \dot{N}\rangle|_{f_p} = \langle \dot{N}\rangle|_{f_s}$ by construction, therefore Λ is computed from the equation

$$\frac{1}{1+\exp(-\Lambda)}\frac{\gamma_p}{\hbar} = \langle \dot{N}\rangle|_{f_s}. \quad (49)$$

It is important to note that the asymptotic scaling defined by Eq. (47) does not mean that the scaling is linear. For example, a sublinear scaling $M(T_p) = \alpha T_p^n$ with $n < 1$ merely corresponds to $\Lambda = 0$ which could satisfy Eq. (49) if the nonequilibrium system is prepared in that way. However, $\Lambda \to \pm\infty$ do not obey the strict inequality in Eq. (45). $\Lambda \to \infty$ corresponds to a trivial and unphysical nonequilibrium distribution $f_s(\omega) \equiv 1$, and likewise, $\Lambda \to -\infty$ corresponds to $f_s(\omega) \equiv 0\ \forall\omega$.

The asymptotic value of $\langle \dot{E}\rangle|_{f_p}$ along the $I_p^{(0)} = 0$ contour is simply

$$\begin{aligned}\lim_{T_p\to\infty}\langle \dot{E}\rangle|_{f_p} &= \frac{1}{h}\int_{-\infty}^{\infty} d\omega\, \omega\mathcal{T}_{ps}(\omega) \lim_{T_p\to\infty}\frac{1}{1+\exp\left(\frac{\omega-M(T_p)}{T_p}\right)}\\ &= \frac{1}{h}\int_{-\infty}^{\infty} d\omega\, \omega\mathcal{T}_{ps}(\omega)\frac{1}{1+\exp(-\Lambda)}\\ &= \frac{1}{1+\exp(-\Lambda)}\frac{\gamma_p}{\hbar}\omega_c\\ &= \omega_c\langle \dot{N}\rangle|_{f_s}. \end{aligned} \quad (50)$$

Theorem 3. A positive temperature solution exists if and only if there is no net population inversion, i.e., when

$$\frac{\langle \dot{E}\rangle|_{f_s}}{\langle \dot{N}\rangle|_{f_s}} < \omega_c. \quad (51)$$

Proof. $\langle \dot{E}\rangle|_{f_p}/\langle \dot{N}\rangle|_{f_s} < \langle \dot{E}\rangle|_{f_s}/\langle \dot{N}\rangle|_{f_s}$ when $T_p \to 0$ along the contour $I_p^{(0)} = 0$ [cf. Eqs. (27) and (43)]. The asymptotic limit of $\langle \dot{E}\rangle|_{f_p}/\langle \dot{N}\rangle|_{f_s}$ is ω_c [cf. Eq. (50)]. $\langle \dot{E}\rangle|_{f_p}$ is continuous $\forall\ \ \mu_p \in (-\infty,\infty), T_p \in (0,\infty)$ and is monotonically increasing along $I_p^{(0)} = 0$ (Theorem 2). We use the intermediate value theorem. ■

Corollary 3.1. There exists a negative temperature solution for a nonequilibrium system with net population inversion, i.e., when

$$\frac{\langle \dot{E}\rangle|_{f_s}}{\langle \dot{N}\rangle|_{f_s}} > \omega_c. \quad (52)$$

Proof. Let $f_p(\mu_p,T_p)$ be the Fermi-Dirac distribution with $T_p > 0$; we define the Fermi-Dirac distribution $f_p^- \equiv f_p(\mu_p,-T_p) = 1 - f_p$:

$$\begin{aligned} &I_p^{(\nu)}(\mu_p,-T_p)\\ &= \frac{1}{h}\int_{-\infty}^{\infty} d\omega(\omega-\mu_p)^\nu \mathcal{T}_{ps}(\omega)\{f_s(\omega)-[1-f_p(\omega)]\}\\ &= \frac{1}{h}\int_{-\infty}^{\infty} d\omega(\omega-\mu_p)^\nu \mathcal{T}_{ps}(\omega)\{f_p(\omega)-[1-f_s(\omega)]\}\\ &= \frac{1}{h}\int_{-\infty}^{\infty} d\omega(\omega-\mu_p)^\nu \mathcal{T}_{ps}(\omega)[f_p(\omega)-f_s^-(\omega)]\\ &\equiv -I_p^{(\nu)-}, \end{aligned} \quad (53)$$

$I_p^{(\nu)-} = 0$ with $\nu = \{0,1\}$ is now understood to solve the complementary nonequilibrium system with $f_s^-(\omega) \equiv 1 - f_s(\omega)$.

$f_s^-(\omega)$ is of course a completely valid nonequilibrium distribution function and satisfies Eq. (8). We apply Theorem 3 and find that

$$\begin{aligned} \langle \dot{E}\rangle|_{f_s^-} &< \omega_c\langle \dot{N}\rangle|_{f_s^-},\\ \frac{\gamma_p}{\hbar}\omega_c - \langle \dot{E}\rangle|_{f_s} &< \omega_c\Big(\frac{\gamma_p}{\hbar} - \langle \dot{N}\rangle|_{f_s}\Big),\\ -\langle \dot{E}\rangle|_{f_s} &< -\omega_c\langle \dot{N}\rangle|_{f_s},\\ \langle \dot{E}\rangle|_{f_s} &> \omega_c\langle \dot{N}\rangle|_{f_s}. \end{aligned} \quad (54)$$

For the case that $\langle \dot{E}\rangle|_{f_s} = \omega_c\langle \dot{N}\rangle|_{f_s}$, $T_p = \pm\infty$, corresponding to $f_p = 1/2$, independent of energy. ■

B. Ideal probes: The broadband limit

In the broadband limit, the probe-system coupling becomes energy independent, and we may write $\Gamma^p(\omega) = \Gamma^p(\mu_0)$. The spectrum of the system, sampled locally by the probe, is given by

$$\begin{aligned}\bar{A}(\omega) &\equiv \frac{\mathrm{Tr}\{\Gamma^p(\omega)A(\omega)\}}{\mathrm{Tr}\{\Gamma^p(\omega)\}}\\ &= \frac{\mathrm{Tr}\{\Gamma^p(\mu_0)A(\omega)\}}{\mathrm{Tr}\{\Gamma^p(\mu_0)\}}. \end{aligned} \quad (55)$$

The occupancy and energy of the system, respectively, are given by

$$\begin{aligned}\langle N\rangle|_{f_s} &= \int_{-\infty}^{\infty} d\omega \bar{A}(\omega) f_s(\omega),\\ \langle E\rangle|_{f_s} &= \int_{-\infty}^{\infty} d\omega\, \omega\bar{A}(\omega) f_s(\omega). \end{aligned} \quad (56)$$

The measurement conditions in Eq. (13) become simply [13]

$$\begin{aligned}\langle N\rangle|_{f_p} &= \langle N\rangle|_{f_s},\\ \langle E\rangle|_{f_p} &= \langle E\rangle|_{f_s}. \end{aligned} \quad (57)$$

The above equations imply that an *ideal measurement* of voltage and temperature constitutes a measurement of the zeroth and first moments of the local energy distribution of the system. That is to say, when the probe is in local equilibrium with the nonequilibrium system, the local occupancy and energy of the system are the same as they would be if the system's local spectrum were populated by the equilibrium Fermi-Dirac distribution $f_p \equiv f_p(\mu_p, T_p)$ of the probe.

We may now write the condition for the existence of a positive temperature solution (Theorem 3) simply as

$$\frac{\langle E\rangle|_{f_s}}{\langle N\rangle|_{f_s}} < \omega_c, \tag{58}$$

where ω_c is the centroid of the spectrum given by

$$\omega_c = \int_{-\infty}^{\infty} d\omega\, \omega \bar{A}(\omega). \tag{59}$$

The condition in Eq. (58) implies the following: Given some nonequilibrium distribution function f_s, one can have a positive temperature solution if and only if the average energy per particle is smaller than the centroid of the spectrum. In other words, a positive temperature solution exists if and only if there is no net population inversion. Similarly, Corollary 3.1 states that there exists a negative temperature solution for a system exhibiting population inversion:

$$\frac{\langle E\rangle|_{f_s}}{\langle N\rangle|_{f_s}} > \omega_c. \tag{60}$$

The advantage of the broadband limit is that one may write the measurement conditions, as well as the condition for the existence of a solution, in terms of the local expectation values of the energy and occupancy directly, instead of using the rate of particle and energy flow into the probe. We also do not need to introduce a "characteristic tunneling rate." We note that ω_c in Eq. (59) is the centroid since the local spectrum $\bar{A}$ normalizes to unity within the broadband limit (see Appendix A 1).

A local measurement by a weakly coupled broadband thermoelectric probe is *ideal* in the sense that the result is independent of the properties of the probe, and depends only on the nonequilibrium state of the system and the subsystem thereof sampled by the probe. Such a measurement provides more than just an operational definition of the local temperature and voltage of a nonequilibrium quantum system, since the thermodynamic variables are determined directly by the moments (56) of the local (nonequilibrium) energy distribution.

C. Nonunique measurements: The narrowband limit

A narrowband probe is one that samples the system only within a very narrow window of energy. The extreme case of such a probe-system coupling would be a Dirac-delta function:

$$\Gamma_p(\omega) = 2\pi V_p^\dagger V_p \delta(\omega - \omega_0), \tag{61}$$

which gives $\mathcal{T}_{ps}(\omega) = 2\pi\, \mathrm{Tr}\{V_p A(\omega) V_p^\dagger\}\delta(\omega-\omega_0)$ which we write simply as

$$\mathcal{T}_{ps}(\omega) = \gamma(\omega)\, \delta(\omega - \omega_0), \tag{62}$$

where $\gamma(\omega) = 2\pi\, \mathrm{Tr}\{V_p A(\omega) V_p^\dagger\}$ has dimensions of energy.

We previously noted that Theorem 1 does not hold for $\mathcal{T}_{ps}$ given by Eq. (62). One can verify straightforwardly that, for a probe-sample transmission that is extremely narrow, we will have

$$\mathcal{L}_{ps}^{(0)}\mathcal{L}_{ps}^{(2)} - \left(\mathcal{L}_{ps}^{(1)}\right)^2 = 0. \tag{63}$$

This results in a nonunique solution since following the proof of Theorem 2 would give us [cf. Eq. (36)] $\Delta I_p^{(1)} = 0$. In fact, it would lead to a family of solutions.

We may solve for the solution explicitly. The number current reduces to

$$I_p^{(0)} = \frac{\gamma(\omega_0)}{h}[f_p(\omega_0) - f_s(\omega_0)], \tag{64}$$

while the heat current is given by

$$I_p^{(1)} = (\omega_0 - \mu_p)\frac{\gamma(\omega_0)}{h}[f_p(\omega_0) - f_s(\omega_0)], \tag{65}$$

which trivially vanishes for vanishing number current. Therefore, the family of solutions to the measurement is simply given by

$$f_p(\omega_0; \mu_p, T_p) = f_s(\omega_0), \tag{66}$$

which is linear in the μ_p-T_p plane and is given by

$$\mu_p = \omega_0 - T_p \log\left(\frac{1 - f_s(\omega_0)}{f_s(\omega_0)}\right). \tag{67}$$

$f_s(\omega)$ has the following explicit form:

$$f_s(\omega) = \frac{\mathrm{Tr}\{V_p G^<(\omega) V_p^\dagger\}}{2\pi i\, \mathrm{Tr}\{V_p A(\omega) V_p^\dagger\}}. \tag{68}$$

A *narrowband probe* is therefore unsuitable for thermoelectric measurements. Even if a probe were to sample the system at just two distinct energies ω_1 and ω_2, Theorem 1 would hold and the thermoelectric measurement would be unique. Indeed, the narrowband probe is a pathological case whose only function is to highlight a certain theoretical limitation for the measurement of the temperature and voltage.

D. Example: Two-level system

Net population inversion is essentially a quantum phenomenon, since classical Hamiltonians are generally unbounded above due to the kinetic energy term, i.e., there does not exist a finite $c \in \mathbb{R}$ that satisfies $\langle H\rangle < c$. In other words, $\omega_c \to \infty$ generally holds for classical systems and negative temperatures are not possible. The simplest quantum system where a net population inversion can be achieved is a two-level system (see Fig. 6). We therefore illustrated our results for a two-level system in Figs. 2–5.

The system Hamiltonian here was taken to be

$$H = \begin{bmatrix} \epsilon_1 & V \\ V^* & \epsilon_2 \end{bmatrix}, \tag{69}$$

whose values were set as $V = \frac{2(1-i)}{3}$, $\epsilon_1 = 1/3$, and $\epsilon_2 = -1/3$, such that the eigenvalues are $\epsilon_\pm = \pm 1$ and units are taken as eV. We introduce two reservoirs that are strongly coupled locally to each site with $\Gamma_1 = \mathrm{diag}(0.5,0)$ and $\Gamma_2 = \mathrm{diag}(0,0.5)$, while the probe coupling is taken as

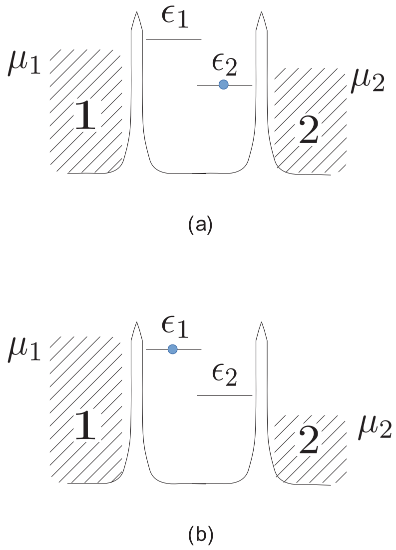

FIG. 6. Schematic diagram of a two-level system coupled to two electron reservoirs under bias. (a) Bias condition not leading to population inversion. (b) Bias condition leading to population inversion due to direct injection into excited state.

$\Gamma_p = \mathrm{diag}(0.01,0.1)$, which is about five times weaker than the coupling to the reservoirs that bias the system.

We used two different bias conditions: (a) To illustrate the case without a net population inversion in Figs. 2–4, the reservoirs had a symmetric ($\mu_1 + \mu_2 = 0$) voltage bias $\mu_1 - \mu_2 = 1$ eV; (b) to illustrate the case with a net population inversion in Fig. 5, the reservoirs had a symmetric voltage bias of $\mu_1 - \mu_2 = 4$ eV. The two reservoirs are held at $T = 300$ K for both cases.

It has been previously noted that the probe-system coupling strength does not strongly affect the measured temperature and voltage even when varied over several orders of magnitude [52], but we remind the reader that our theoretical results depend upon the assumption of a weakly coupled probe (noninvasive measurements). How *weak* is *weak enough* is a different, and more subtle, theoretical question addressed in Appendix B. Numerically, however, we do find that the probe measurements are not much altered even when the probe coupling strength is comparable to that of the strongly coupled reservoirs. We also show in Appendix B that, for noninteracting electrons, all our results hold for strongly-coupled probes with arbitrary bias conditions.

VI. CONCLUSIONS

The local temperature and voltage of a nonequilibrium quantum system are defined in terms of the equilibration of a noninvasive thermoelectric probe, locally coupled to the system. The simultaneous temperature and voltage measurement is shown to be unique for any system of fermions in steady state, arbitrarily far from equilibrium, with arbitrary interactions within the system, and the conditions for the existence of a solution are derived. In particular, it is shown that a positive temperature solution exists provided the system does not have a net local population inversion; in the case of population inversion, a unique negative temperature solution is shown to exist. Our results hold for arbitrarily strong probe couplings for noninteracting systems. These results provide a firm mathematical foundation for temperature and voltage measurements in quantum systems far from equilibrium.

Our analysis reveals that a simultaneous temperature and voltage measurement is uniquely determined by the local spectrum and nonequilibrium distribution of the system [cf. Eq. (42)], and is independent of the properties of the probe for broadband coupling (ideal probe). Such a measurement therefore provides a *fundamental definition* of local temperature and voltage, which is not merely operational.

In contrast, prior theoretical work relied almost exclusively on operational definitions [12,37–46], leading to a competing panoply of often contradictory predictions for the measurement of such basic observables as temperature and voltage. Measurements of temperature or voltage, taken separately (see, e.g., Refs. [12,37]), are shown to be ill-posed: a thermometer out of electrical equilibrium with a system produces an error due to the Peltier effect across the probe-sample junction, while a potentiometer out of thermal equilibrium with a system produces an error due to the Seebeck effect.

Our results put the local thermodynamic variables temperature and voltage on a mathematically rigorous footing for fermion systems under very general nonequilibrium steady-state conditions, a necessary first step toward the construction of nonequilibrium thermodynamics [8–15]. Our analysis includes the effect of interactions with bosonic degrees of freedom (e.g., photons, phonons, etc.) on the fermions. However, the temperatures of the bosons themselves [53,54] were not addressed in the present analysis. Moreover, we did not explicitly consider magnetic systems, which require separate consideration of the spin degree of freedom, and its polarization. Future investigation of probes that exchange bosonic or spin excitations may enable similarly rigorous analysis of local thermodynamic variables in bosonic and magnetic systems, respectively.

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APPENDIX A: THE NONEQUILIBRIUM STEADY STATE

We consider a system whose Hamiltonian $\hat{H}$ is independent of time, but is driven out of equilibrium, e.g., by electrical and/or thermal bias. The nonequilibrium steady state is described by a density matrix $\hat{\rho}$ that is time independent. The expectation values of observables are given by their usual prescription in statistical physics

$$\langle \hat{Q} \rangle = \mathrm{Tr}\{\hat{\rho}\hat{Q}\} = \sum_{\mu,\nu} \rho_{\mu\nu} \langle \nu | \hat{Q} | \mu \rangle. \tag{A1}$$

The "lesser" and "greater" Green's functions [58] used in the paper are defined as follows:

$$G^{<}_{\alpha\beta}(t) \equiv i \langle d^{\dagger}_{\beta}(0) d_{\alpha}(t) \rangle, \tag{A2}$$

while its Hermitian conjugate is

$$G^{>}_{\alpha\beta}(t) \equiv -i\langle d_\alpha(t) d^\dagger_\beta(0)\rangle, \tag{A3}$$

where

$$d_\alpha(t) = e^{i\frac{\hat{H}}{\hbar}t} d_\alpha(0) e^{-i\frac{\hat{H}}{\hbar}t} \tag{A4}$$

evolves according to the Heisenberg equation of motion for a system with Hamiltonian $\hat{H}$. Here α, β denote basis states in the one-body Hilbert space of the system.

The spectral representation uses the eigenbasis of the Hamiltonian $\hat{H}|\nu\rangle = E_\nu|\nu\rangle$, where ν denotes a many-body energy eigenstate. One may write the lesser Green's function as

$$G^{<}_{\alpha\beta}(\omega) = 2\pi i \sum_{\mu,\mu',\nu} \rho_{\mu\nu}\langle\nu|d^\dagger_\beta|\mu'\rangle\langle\mu'|d_\alpha|\mu\rangle \times \delta\left(\omega - \frac{E_\mu - E_{\mu'}}{\hbar}\right), \tag{A5}$$

while the greater Green's function becomes

$$G^{>}_{\alpha\beta}(\omega) = -2\pi i \sum_{\mu,\mu',\nu} \rho_{\mu\nu}\langle\nu|d_\alpha|\mu'\rangle\langle\mu'|d^\dagger_\beta|\mu\rangle \times \delta\left(\omega - \frac{E_{\mu'} - E_\nu}{\hbar}\right). \tag{A6}$$

The spectral function $A(\omega)$ is given by

$$A(\omega) \equiv \frac{1}{2\pi i}(G^<(\omega) - G^>(\omega)), \tag{A7}$$

and can be expressed in the spectral representation as

$$A_{\alpha\beta}(\omega) = \sum_{\mu,\mu',\nu} [\rho_{\mu\nu}\langle\nu|d^\dagger_\beta|\mu'\rangle\langle\mu'|d_\alpha|\mu\rangle + \rho_{\nu\mu'}\langle\mu'|d_\alpha|\mu\rangle\langle\mu|d^\dagger_\beta|\nu\rangle]\delta\left(\omega - \frac{E_\mu - E_{\mu'}}{\hbar}\right). \tag{A8}$$

1. Sum rule for the spectral function

Equation (A8) leads to the following sum rule for the spectral function:

$$\begin{aligned}\int_{-\infty}^{\infty} d\omega A_{\alpha\beta}(\omega) &= \sum_{\mu,\nu} \rho_{\mu\nu}\langle\nu|d^\dagger_\beta d_\alpha|\mu\rangle + \sum_{\mu',\nu} \rho_{\nu\mu'}\langle\mu'|d_\alpha d^\dagger_\beta|\nu\rangle \\ &= \sum_{\mu,\nu} \rho_{\mu\nu}\langle\nu|d^\dagger_\beta d_\alpha + d_\alpha d^\dagger_\beta|\mu\rangle \\ &= \sum_{\mu,\nu} \rho_{\mu\nu}\langle\nu|\delta_{\alpha\beta}|\mu\rangle \\ &= \sum_{\mu,\nu} \rho_{\mu\nu}\delta_{\mu\nu}\delta_{\alpha\beta} \\ &= \delta_{\alpha\beta}\,\mathrm{Tr}\,\{\hat{\rho}\} \\ &= \delta_{\alpha\beta}.\end{aligned} \tag{A9}$$

In our theory of local thermodynamic measurements, the quantity of interest is the local spectrum of the system sampled by the probe $\bar{A}(\omega)$, defined in Eq. (55). This obeys a further sum rule in the broadband limit (*ideal probe*), discussed below.

Local spectrum in the broadband limit

The probe-system coupling is energy independent in the broadband limit, $\Gamma^p(\omega) = \text{const}$, and we write $\mathrm{Tr}\,\{\Gamma^p\} = \bar{\Gamma}^p$ for its trace. The local spectrum sampled by the probe $\bar{A}(\omega)$ defined in Eq. (55) can be written in the broadband limit as

$$\bar{A}(\omega) = \frac{1}{\bar{\Gamma}^p}\sum_{\alpha,\beta}\langle\beta|\Gamma^p|\alpha\rangle A_{\alpha\beta}(\omega). \tag{A10}$$

In this limit it obeys a further sum rule:

$$\begin{aligned}\int_{-\infty}^{\infty} d\omega \bar{A}(\omega) &= \frac{1}{\bar{\Gamma}^p}\sum_{\alpha,\beta}\langle\beta|\Gamma^p|\alpha\rangle \int_{-\infty}^{\infty} d\omega A_{\alpha\beta}(\omega) \\ &= \frac{1}{\bar{\Gamma}^p}\sum_{\alpha,\beta}\langle\beta|\Gamma^p|\alpha\rangle\delta_{\alpha\beta} \\ &= 1.\end{aligned} \tag{A11}$$

The broadband limit is special in that the measurement is determined by the local properties of the system itself, and is not influenced by the spectrum of the probe. In this limit, the local spectrum $\bar{A}(\omega)$ obeys the sum rule (A11) since the probe samples the same subsystem at all energies. One should not expect such a local sum rule to hold outside the broadband limit, since the probe samples different subsystems at different energies.

2. Diagonality of $\hat{\rho}$

We have, for any observable $\hat{Q}$,

$$\begin{aligned}\langle\hat{Q}(t)\rangle &= \sum_{\mu,\nu}\rho_{\mu\nu}\langle\nu|\hat{Q}(t)|\mu\rangle \\ &= \sum_{\mu,\nu}\rho_{\mu\nu}\langle\nu|e^{i\frac{\hat{H}}{\hbar}t}\hat{Q}e^{-i\frac{\hat{H}}{\hbar}t}|\mu\rangle \\ &= \sum_{\mu,\nu}\rho_{\mu\nu}e^{-i\frac{E_\mu - E_\nu}{\hbar}t}\langle\nu|\hat{Q}|\mu\rangle.\end{aligned} \tag{A12}$$

The system observables must be independent of time in steady state. Therefore $\hat{\rho}$ must be diagonal in the energy basis, as seen from the above equation. The nondiagonal parts of $\hat{\rho}$ in the energy basis, when they exist, must be in a degenerate subspace so that $E_\mu = E_\nu$ in the above equation.

For states degenerate in energy, the boundary conditions determining the nonequilibrium steady state will determine the basis in which $\hat{\rho}$ is diagonal. Henceforth, we work in that basis.

3. Positivity of $-iG^<(\omega)$ and $iG^>(\omega)$

Working in the energy eigenbasis in which $\hat{\rho}$ is diagonal,

$$\begin{aligned}&-i\langle\alpha|G^<(\omega)|\alpha\rangle \\ &\equiv -iG^<_{\alpha\alpha}(\omega) \\ &= 2\pi\sum_{\mu,\mu'}\rho_{\mu\mu}|\langle\mu|d^\dagger_\alpha|\mu'\rangle|^2\delta\left(\omega - \frac{E_\mu - E_{\mu'}}{\hbar}\right) \\ &\geqslant 0.\end{aligned} \tag{A13}$$

Similarly,

$$i\langle\alpha|G^>(\omega)|\alpha\rangle \equiv iG^>_{\alpha\alpha}(\omega) = 2\pi\sum_{\mu,\mu'}\rho_{\mu\mu}|\langle\mu|d^\dagger_\alpha|\mu'\rangle|^2\delta\left(\omega-\frac{E_{\mu'}-E_\mu}{\hbar}\right) \geqslant 0. \tag{A14}$$

It follows that

$$\langle\alpha|A(\omega)|\alpha\rangle = \frac{1}{2\pi}\langle\alpha|-iG^<(\omega)+iG^>(\omega)|\alpha\rangle \geqslant 0. \tag{A15}$$

Therefore, all three operators $-iG^<(\omega)$, $iG^>(\omega)$, and $A(\omega)$ are positive-semidefinite.

4. $0 \leqslant f_s(\omega) \leqslant 1$

The nonequilibrium distribution function $f_s(\omega)$ was defined in Eq. (3) as

$$f_s(\omega) \equiv \frac{\mathrm{Tr}\{\Gamma^p(\omega)G^<(\omega)\}}{2\pi i\,\mathrm{Tr}\{\Gamma^p(\omega)A(\omega)\}}. \tag{A16}$$

We have $\Gamma^p(\omega) > 0$ by causality [58]:

$$\mathrm{Im}\ \Sigma^r_p(\omega) = -\tfrac{1}{2}\Gamma^p(\omega) < 0. \tag{A17}$$

Let $\Gamma^p|\gamma_p\rangle = \gamma_p|\gamma_p\rangle$, where $\gamma_p \geqslant 0$ and some γ_p satisfy $\gamma_p > 0$. The energy dependence is taken to be implicit. The traces in Eq. (A16) may be evaluated in the eigenbasis of Γ^p, yielding

$$f_s(\omega) = \frac{\sum_{\gamma_p}\gamma_p\langle\gamma_p|G^<(\omega)|\gamma_p\rangle}{2\pi i\sum_{\gamma_p}\gamma_p\langle\gamma_p|A(\omega)|\gamma_p\rangle} = \frac{\sum_{\gamma_p}\gamma_p\langle\gamma_p|-iG^<(\omega)|\gamma_p\rangle}{\sum_{\gamma_p}\gamma_p\langle\gamma_p|-iG^<(\omega)+iG^>(\omega)|\gamma_p\rangle}. \tag{A18}$$

Therefore

$$0 \leqslant f_s(\omega) \leqslant 1. \tag{A19}$$

APPENDIX B: NONINVASIVE PROBES

Our main results in this paper relied upon the assumption of a noninvasive probe. We explained the physical basis for this assumption in Sec. III A, and we understood it to mean that the local probe-system transmission function $\mathcal{T}_{ps}$ and the local nonequilibrium distribution function f_s are independent of the probe bias parameters (μ_p,T_p). In this Appendix we clarify the implicit mathematical details that have gone into this assumption of a noninvasive probe.

f_s and $\mathcal{T}_{ps}$ have been defined in Eqs. (3) and (4), respectively, and they depend upon the Green's functions of the nonequilibrium quantum system. The Green's functions of the system do depend upon the probe parameters (μ_p,T_p) and we clarify this dependence. We label the probe parameter simply as $x_p \in \{\mu_p,T_p\}$, which can be taken to mean either the chemical potential or the temperature of the probe.

In order to characterize the noninvasive-probe limit, we introduce a dimensionless parameter λ, and write the probe-system coupling as $\Gamma^p(\omega) = \lambda\tilde{\Gamma}^p(\omega)$. Without loss of generality, we may set $\mathrm{Tr}\{\tilde{\Gamma}^p(\mu_0)\} = \sum_{\alpha\neq p}\mathrm{Tr}\{\Gamma^\alpha(\mu_0)\}$, where $\Gamma^\alpha(\omega)$ is the tunneling-width matrix describing the coupling of lead α (e.g., source, drain, etc.) to the system, and μ_0 is the equilibrium chemical potential of the system (or some other convenient reference value). The parameter

$$\lambda \equiv \frac{\mathrm{Tr}\{\Gamma^p(\mu_0)\}}{\sum_{\alpha\neq p}\mathrm{Tr}\{\Gamma^\alpha(\mu_0)\}} \ll 1 \tag{B1}$$

thus gives the condition for a weakly coupled probe.

The currents flowing into the probe from the system are given by Eq. (5) as

$$I^{(\nu)}_p = \frac{1}{h}\int_{-\infty}^{\infty} d\omega(\omega-\mu_p)^\nu\mathcal{T}_{ps}(\omega)[f_s(\omega)-f_p(\omega)], \tag{B2}$$

where

$$\mathcal{T}_{ps} = \lambda 2\pi\,\mathrm{Tr}\{\tilde{\Gamma}^p A\} = \lambda 2\pi\,\mathrm{Tr}\{\tilde{\Gamma}^p A|_{\lambda=0}\} + \lambda^2 2\pi\,\mathrm{Tr}\left\{\tilde{\Gamma}^p\frac{\partial A}{\partial\lambda}\bigg|_{\lambda=0}\right\} + O(\lambda^3) \tag{B3}$$

and

$$\mathcal{T}_{ps}f_s = -i\lambda\,\mathrm{Tr}\{\tilde{\Gamma}^p G^<\} = -i\lambda\,\mathrm{Tr}\{\tilde{\Gamma}^p G^<|_{\lambda=0}\} - i\lambda^2\,\mathrm{Tr}\left\{\tilde{\Gamma}^p\frac{\partial G^<}{\partial\lambda}\bigg|_{\lambda=0}\right\}+O(\lambda^3) \tag{B4}$$

[cf. Eqs. (3) and (4)]. From Eqs. (B2)–(B4) we see that $I^{(\nu)}_p \sim O(\lambda)$. Similarly, it can be shown that

$$\frac{\partial I^{(\nu)}_p}{\partial x_p} = -I^{(0)}_p\delta_{\nu,1}\delta_{x_p,\mu_p} - \frac{\lambda}{\hbar}\int_{-\infty}^{\infty} d\omega(\omega-\mu_p)^\nu \times \mathrm{Tr}\{\tilde{\Gamma}^p A|_{\lambda=0}\}\frac{\partial f_p}{\partial x_p} + O(\lambda^2). \tag{B5}$$

The leading-order results for the gradients are also $O(\lambda)$, and agree with Eqs. (16) and (17). The noninvasive probe limit consists in keeping only the terms $O(\lambda)$ in Eqs. (B3), (B4), and (B5), and underlies the analysis presented in the body of the article. Deviations from the noninvasive probe limit appear as terms $O(\lambda^2)$ and higher, which we now proceed to derive.

1. Dependence of G on λ and x_p

Standard NEGF arguments can be used to elucidate the dependence of the system Green's functions on λ and x_p. Let G_0 denote the Green's function of the isolated quantum system without two-body interactions, and let Σ denote the self-energy describing two-body interactions and coupling to various reservoirs, including the probe. Dyson's equation for the retarded (advanced) Green's function is [58]

$$G^{r,a} = G_0^{r,a} + G_0^{r,a}\Sigma^{r,a}G^{r,a}. \tag{B6}$$

The Keldysh equation for $G^<$ is [58]

$$G^<(\omega) = G^r(\omega)\Sigma^<(\omega)G^a(\omega), \tag{B7}$$

where the lesser self-energy is

$$\Sigma^< = i\lambda\tilde{\Gamma}^p(\omega)f_p(\omega) + i\sum_{\alpha\neq p}\Gamma^\alpha(\omega)f_\alpha(\omega) + \Sigma^<_{\mathrm{int}}, \tag{B8}$$

and $\Sigma^<_{\mathrm{int}}$ is the self-energy contribution due to electron-electron, electron-phonon, electron-photon interactions, etc.

Similarly, the spectral function A may be expressed as

$$2\pi A(\omega) = G^r(\omega)\Gamma(\omega)G^a(\omega), \tag{B9}$$

where

$$\Gamma(\omega) = \lambda\tilde{\Gamma}^p(\omega) + \sum_{\alpha\neq p}\Gamma^\alpha(\omega) + \Gamma_{\rm int}(\omega), \tag{B10}$$

and $\Gamma_{\rm int} = i(\Sigma^r_{\rm int} - \Sigma^a_{\rm int})$ is the contribution due to two-body interactions. Note that all the terms appearing on the rhs of Eq. (B10) are positive definite due to causality.

Differentiating the self-energies with respect to x_p gives

$$\frac{\partial\Sigma^<}{\partial x_p} = i\lambda\tilde{\Gamma}^p\frac{\partial f_p}{\partial x_p} + \frac{\partial\Sigma^<_{\rm int}}{\partial x_p} \tag{B11}$$

and

$$\frac{\partial\Sigma^{r,a}}{\partial x_p} = \frac{\partial\Sigma^{r,a}_{\rm int}}{\partial x_p}. \tag{B12}$$

Using Eqs. (B6), (B7), (B11), and (B12), it can be shown that

$$\frac{\partial G^{r,a}}{\partial x_p} = G^{r,a}\frac{\partial\Sigma^{r,a}_{\rm int}}{\partial x_p}G^{r,a}, \tag{B13}$$

$$\frac{\partial G^<}{\partial x_p} = i\lambda G^r\tilde{\Gamma}^pG^a\frac{\partial f_p}{\partial x_p} + G^r\frac{\partial\Sigma^<_{\rm int}}{\partial x_p}G^a + G^r\frac{\partial\Sigma^r_{\rm int}}{\partial x_p}G^< + G^<\frac{\partial\Sigma^a_{\rm int}}{\partial x_p}G^a. \tag{B14}$$

Using $2\pi iA = G^a - G^r$, the derivative of the spectral function may be written as

$$2\pi i\frac{\partial A(\omega)}{\partial x_p} = G^a\frac{\partial\Sigma^a_{\rm int}}{\partial x_p}G^a - G^r\frac{\partial\Sigma^r_{\rm int}}{\partial x_p}G^r. \tag{B15}$$

Finally, the derivatives of $\Sigma_{\rm int}$ are given by

$$\frac{\partial\Sigma^\gamma_{\rm int}(\omega)}{\partial x_p} = \sum_{\eta=r,a,<}\int_{-\infty}^{\infty}d\omega' K^{\gamma\eta}(\omega,\omega')\frac{\partial G^\eta(\omega')}{\partial x_p}, \tag{B16}$$

where

$$K^{\gamma\eta}(\omega,\omega') \equiv \frac{\delta\Sigma^\gamma_{\rm int}(\omega)}{\delta G^\eta(\omega')} \tag{B17}$$

is the irreducible kernel for the two-particle Green's function [58].

Equations (B13), (B14), and (B16) are three coupled linear (integral) equations for $\partial G/\partial x_p$ and $\partial\Sigma_{\rm int}/\partial x_p$. The only inhomogeneous term [first term on the rhs of Eq. (B14)] is $O(\lambda)$. Let

$$\frac{\partial G^\gamma(\omega)}{\partial x_p} \equiv \lambda F^\gamma_{x_p}(\omega), \tag{B18}$$

$$\frac{\partial\Sigma^\gamma_{\rm int}(\omega)}{\partial x_p} \equiv \lambda S^\gamma_{x_p}(\omega). \tag{B19}$$

F and S satisfy the equations

$$F^{r,a}_{x_p} = G^{r,a}S^{r,a}_{x_p}G^{r,a}, \tag{B20}$$

$$F^<_{x_p} = iG^r\tilde{\Gamma}^pG^a\frac{\partial f_p}{\partial x_p} + G^rS^<_{x_p}G^a + G^rS^r_{x_p}G^< + G^<S^a_{x_p}G^a, \tag{B21}$$

and

$$S^\gamma_{x_p} = \sum_{\eta=r,a,<}K^{\gamma\eta}F^\eta_{x_p}, \tag{B22}$$

where the energy integral on the rhs of Eq. (B22) is implicit. The leading-order solution is obtained by setting $G^\gamma = G^\gamma|_{\lambda=0}$ in Eqs. (B20) and (B21), so we see that $\partial G/\partial x_p$, $\partial\Sigma_{\rm int}/\partial x_p \sim O(\lambda)$, and can be neglected in the noninvasive probe limit. There exist a number of additional terms in $\partial G/\partial\lambda|_{\lambda=0}$ that are independent of x_p, but these do not affect the proofs of Theorems 1, 2, and 3.

2. Proof of uniqueness

We are now in a position to evaluate the dependence of the currents $I^{(\nu)}_p$ on $x_p \in \{\mu_p, T_p\}$. Taking the derivative of Eq. (B2) using the results of Appendix B 1, one obtains the exact expression

$$\begin{aligned}\frac{\partial I^{(\nu)}_p}{\partial x_p} = &-I^{(0)}_p\delta_{\nu,1}\delta_{x_p,\mu_p}\\ &-\frac{\lambda}{h}\int_{-\infty}^{\infty}d\omega(\omega-\mu_p)^\nu\,{\rm Tr}\{\tilde{\Gamma}^pG^r(\Gamma-\Gamma^p)G^a\}\frac{\partial f_p}{\partial x_p}\\ &-\frac{i\lambda^2}{h}\int_{-\infty}^{\infty}d\omega(\omega-\mu_p)^\nu\,{\rm Tr}\left\{\tilde{\Gamma}^p\left(G^rS^<_{x_p}G^a + G^rS^r_{x_p}G^< + G^<S^a_{x_p}G^a\right)\right\}\\ &+\frac{i\lambda^2}{h}\int_{-\infty}^{\infty}d\omega(\omega-\mu_p)^\nu f_p(\omega)\\ &\times{\rm Tr}\left\{\tilde{\Gamma}^p\left(G^aS^a_{x_p}G^a - G^rS^r_{x_p}G^r\right)\right\}.\end{aligned} \tag{B23}$$

To leading order in λ, Eq. (B23) reduces to the result given in Eqs. (16) and (17), while the corrections are $O(\lambda^2)$ or higher. Thus Theorems 1 and 2 hold to leading order in λ for systems with arbitrary two-body interactions, and the noninvasive-probe limit may be precisely defined as the limit $\lambda \ll 1$.

a. Special case: Noninteracting system

Without two-body interactions, only the first two terms in Eq. (B23) survive. The current gradients therefore have the same form as in Eqs. (16) and (17), while the $\mathcal{L}^{(\nu)}_{ps}$ coefficients have the same form [cf. Eq. (18)] but with the transmission function replaced by

$$\tilde{\mathcal{T}}_{ps}(\omega) = {\rm Tr}\{\Gamma^p(\omega)G^r(\omega)[\Gamma(\omega)-\Gamma^p(\omega)]G^a(\omega)\}, \tag{B24}$$

which is positive due to causality [see Eq. (B10)]. Theorem 1 therefore still holds. The uniqueness result as stated in Theorem 2 holds also, since the argument only makes use of current gradients. We note that Theorems 1 and 2 hold for arbitrarily strong probe couplings when two-body interactions are absent.

b. Example: Hartree-Fock approximation

In the Hartree-Fock approximation, the irreducible kernel defined in Eq. (B17) has the form

$$K^{r<} \equiv \frac{\delta\left(\Sigma^r_{\mathrm{HF}}\right)_{nm}}{\delta(G^<)_{ij}} = U_{nj}\delta_{nm}\delta_{ij} - U_{nm}\delta_{ni}\delta_{mj}, \tag{B25}$$

where U_{nm} is the Coulomb integral between orthonormal basis orbitals n and m of the system. Furthermore, $K^{a<} = K^{r<}$ and $K^{<<} = K^{><} = 0$. Equation (B22) therefore reduces to $S^<_{x_p} = 0$ and

$$\begin{aligned}\left(S^{r,a}_{x_p}\right)_{nm} = \delta_{nm}\sum_j U_{nj}\int_{-\infty}^{\infty} d\omega'[F^<_{x_p}(\omega')]_{jj} \\ - U_{nm}\int_{-\infty}^{\infty} d\omega'[F^<_{x_p}(\omega')]_{nm}.\end{aligned} \tag{B26}$$

3. Proof of existence

The proof of Theorem 3 is based on an analysis of the quantities $\langle\dot N\rangle|_{f_s}$, $\langle\dot N\rangle|_{f_p}$, $\langle\dot E\rangle|_{f_s}$, and $\langle\dot E\rangle|_{f_p}$ defined in Eqs. (38)–(41), respectively. These quantities are simply energy integrals of $\omega^\nu\mathcal{T}_{ps}f_s$ and $\omega^\nu\mathcal{T}_{ps}f_p$, with $\nu = 0,1$, whose dependence on the small parameter λ is given in Eqs. (B3) and (B4). Keeping only terms $O(\lambda)$ (noninvasive-probe limit), these quantities reduce to the form considered in Sec. V, so that Theorem 3 and Corollary 3.1 hold as before. Deviations from the noninvasive-probe limit involve corrections $O(\lambda^2)$ and higher, and it is an open question whether a unique solution to the probe equilibration conditions (42) exists for arbitrarily strong probe-system coupling in the presence of interactions.

Special case: Noninteracting system

For systems without two-body interactions, the proof of Theorem 3 can be straightforwardly extended to the case of arbitrarily strong probe-system coupling. Using Eqs. (B3) and (B10) with $\Gamma_{\mathrm{int}} = 0$, one can write

$$\mathcal{T}_{ps} = \sum_\alpha \mathrm{Tr}\{\Gamma^p G^r \Gamma^\alpha G^a\}. \tag{B27}$$

Similarly, using Eqs. (B4) and (B8) with $\Sigma^<_{\mathrm{int}} = 0$, one has

$$\mathcal{T}_{ps}f_s = \sum_\alpha \mathrm{Tr}\{\Gamma^p G^r \Gamma^\alpha G^a\}f_\alpha. \tag{B28}$$

The probe equilibration conditions (42) whose solution we seek may be rewritten

$$\begin{aligned}\langle\dot N\rangle|_{f_s} - \langle\dot N\rangle|_{f_p} = 0,\\ \langle\dot E\rangle|_{f_s} - \langle\dot E\rangle|_{f_p} = 0.\end{aligned} \tag{B29}$$

The integrands in both conditions involve

$$\begin{aligned}\mathcal{T}_{ps}[f_s - f_p] &= \sum_{\alpha\neq p}\mathrm{Tr}\{\Gamma^p G^r \Gamma^\alpha G^a\}[f_\alpha - f_p]\\ &= \tilde{\mathcal{T}}_{ps}[\tilde f_s - f_p],\end{aligned} \tag{B30}$$

where $\tilde{\mathcal{T}}_{ps}$ is given by Eq. (B24) and

$$\tilde f_s = \frac{\sum_{\alpha\neq p}\mathrm{Tr}\{\Gamma^p G^r \Gamma^\alpha G^a\}f_\alpha}{\sum_{\alpha\neq p}\mathrm{Tr}\{\Gamma^p G^r \Gamma^\alpha G^a\}}. \tag{B31}$$

$\tilde f_s$ and $\tilde{\mathcal{T}}_{ps}$ are both independent of x_p for the noninteracting system, and $\langle\dot N\rangle|_{f_s}$, $\langle\dot N\rangle|_{f_p}$, $\langle\dot E\rangle|_{f_s}$, and $\langle\dot E\rangle|_{f_p}$ can be redefined using $\tilde f_s$ and $\tilde{\mathcal{T}}_{ps}$ without affecting the conditions (B29). Therefore the proofs of Theorem 3 and Corollary 3.1 hold for arbitrarily strong probe-system coupling in systems without two-body interactions.

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