Nonequilibrium Quantum Impurities: From Entropy Production to Information Theory

Pankaj Mehta
Department of Molecular Biology and Department of Physics, Princeton University, Princeton, New Jersey 08544, USA

Natan Andrei
Center for Materials Theory, Rutgers University, Piscataway, New Jersey 08854, USA

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Nonequilibrium steady-state currents, unlike their equilibrium counterparts, continuously dissipate energy into their physical surroundings leading to entropy production and time-reversal symmetry breaking. This Letter discusses these issues in the context of quantum impurity models. We use simple thermodynamic arguments to define the rate of entropy production σ and show that σ has a simple information-theoretic interpretation in terms of nonequilibrium distribution functions. This allows us to show that the entropy production is *strictly positive* for any nonequilibrium steady state. We conclude by applying these ideas to the resonance level model and the Kondo model.

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Over the last decade, it has become possible to explore experimentally quantum-impurity systems in nonequilibrium settings [1]. The quantum impurities are most often realized using quantum dots, small pools of electrons confined in space. In a typical experimental setup, a quantum dot is attached to multiple leads at different chemical potentials resulting in a nonequilibrium current across the dot. This opens up the exciting possibility of using quantum-impurity models to experimentally and theoretically explore nonequilibrium physics.

We focus in this Letter on the case when the currents across the dot are in a nonequilibrium steady state (NESS). NESSs have been extensively studied in a variety of physical systems including lattice models and fluid systems [2]. A system in a NESS continuously dissipates energy into its surrounding, resulting in a continuous production of entropy and the breaking of time-reversal symmetry. This is in contrast to an equilibrium steady-state where a persistent current can flow without producing entropy or dissipating energy.

NESSs in quantum-impurity systems are usually modeled by adiabatically turning on the interaction between the impurity and the leads in the far past, at a time $t_0 < 0$, and then evolving the system in time to the present, $t = 0$. Thus at $t < t_o$ the system is described by a density matrix ρ_o describing two noninteracting leads (reservoirs) at different chemical potentials μ_i and different temperatures T_i and the uncoupled impurity. At time $t = t_0$ the impurity is coupled to the leads and evolves adiabatically according to $H = H_o + \theta(t - t_0)e^{\eta t}H_1$. At a time t it is described by a time-evolved density operator $\rho(t) = U(t, t_0)\rho_o U^\dagger(t, t_0)$, where the evolution operator $U(t, t_0)$ corresponds to $H(t)$. The nonequilibrium density matrix is used to compute nonequilibrium expectation values $\langle \hat{O} \rangle = \mathrm{Tr}\rho(t)\hat{O}$.This expectation value becomes time independent and a steady state emerges if the leads are good thermal baths and the system is *open*. Namely, both the number of particles in the lead N_i and the size of the lead L are infinite, with the limit taken *ab initio*. The establishment of a steady state follows, in this language, from the existence of the open-system limit $v_F/L \ll 1/|t_o| \ll \eta \to 0$, with v_F the Fermi velocity. In this case, the density matrix becomes time independent [3]. We denoted it by ρ_s. Most treatments of the problem are based on calculating ρ_s in various ways. A large class of theoretical treatments uses Keldysh perturbation theory to calculate ρ_s [3,4], while others use Bethe-Ansatz based approaches [5] or a recursive algorithm [6]. The scattering framework recently proposed by the authors uses scattering theory to calculate ρ_s in a time-independent manner [7]. In all these treatments, the nonequilibrium density matrix ρ_s commutes with the Hamiltonian of the system. This raises the intriguing question of how a density matrix that commutes with the Hamiltonian captures defining characteristics of the nonequilibrium physics, such as energy and particle currents, energy dissipation, and entropy production.

In an *open* system it is not necessary to include explicit mechanisms, such as phonons, to allow the relaxation of the high-energy electrons transferred between leads. Instead, energy dissipation is implemented by the open-system limit. Since the size of the system is much larger than the turn-on time, the high-energy electrons transferred between leads continue off to the edges of the leads and "dissipate" their energy infinitely far away. The limit also ensures that once the electrons cross the impurity they cannot return to the system, giving rise to time-reversal symmetry breaking. In the language of Green's functions, the open-system limit induces the poles in the self-energy to merge into a branch cut, leading to dissipative effects [8]. Under these circumstances the nonequilibrium physics is captured by *scattering eigenstates*—eigenstates of the Hamiltonian $H = H_0 + H_1$ defined on the open system with appropriate asymptotic boundary conditions [7]. Their existence follows in the open-system limit by the Gellman-Low theorem [9].

In the time-independent picture that emerges after taking the appropriate limits, time has been traded for space [10]. It is convenient to follow the standard steps to express the impurity problem in a one dimensional language and "unfold" the leads [11,12]. One then has only right movers, with the incoming particles in the region $x < 0$ and the outgoing particles in the region $x > 0$, while the impurity is located at $x = 0$. In this picture, the far past corresponds to the region $x \ll -l_0$ and the far future to the region $x \gg l_0$, where l_0 is the length scale characterizing the influence of the impurity. It is typically expressed as $l_0 = v_F/k_B T_0$, with T_0 being the low-energy scale, e.g., the Kondo temperature. The incoming and outgoing regions can be neatly described by introducing the corresponding asymptotic single-particle distribution functions $f_i^{\mp}$ [13,14] (Fig. 1). The distribution function of the incoming particles in lead i is given by the Fermi-Dirac function $f_i^-(p_\alpha) = f_{\mathrm{FD}}(p_\alpha; T_i, \mu_i)$ at the temperature and chemical potential appropriate to the lead. After scattering off the impurity at $x = 0$, the outgoing particles in lead i are asymptotically described by a distribution function $f_i^+(p_\alpha)$ which is no longer the Fermi distribution function but a nonequilibrium distribution function that encodes information about scattering. The distribution functions contain all the information about the nonequilibrium energy and particle currents since these are computed from local single-particle operators. Note, these distribution functions can be computed perturbatively in the Landauer-Büttiker approach [15].

We now discuss the entropy production in the system. We begin with thermodynamic considerations, then reformulate the problem in terms of nonequilibrium distributions which emerge naturally in the scattering formalism. We then relate our expressions for entropy production to information-theoretic quantities of the underlying nonequilibrium distribution functions. This allows us to show that the entropy production is due to two fundamental processes, *mixing* and *relaxation*, for which we give explicit expressions. Finally we prove that the rate of entropy production is strictly positive in the NESS. As an application of these concepts, we explore how strong correlations manifest themselves in the entropy production of the Kondo model.

The thermodynamic definition of the rate of entropy production follows naturally from the observation that a quantum impurity coupled to leads is a discontinuous system: the two reservoirs are connected to each other by a single impurity [16]. In such a system, all entropy is produced in the leads and the entropy produced at the quantum impurity itself is negligible. Recall that the differential entropy of a system, dS, is related to δQ the heat that flows into or out of the system via $TdS = \delta Q$ with T the temperature of the system. For a discontinuous system with two leads, this allows us to define the rate of entropy production σ as

$$\sigma \equiv \frac{dS}{dt} \equiv \frac{1}{T_1}\frac{\delta Q_1}{dt} + \frac{1}{T_2}\frac{\delta Q_2}{dt}, \tag{1}$$

where δQ_i is the heat that flows into lead i, and T_i is the temperature of lead i. The heat produced by a lead is related to the change in energy and particle number in the lead by $\delta Q_i = dE_i - \mu_i dN_i$. Furthermore, since neither particles nor energy can disappear at the quantum impurity in a nonequilibrium steady state we have the conservation laws $\frac{dN_1}{dt} = -\frac{dN_2}{dt}$ and $\frac{dE_1}{dt} = -\frac{dE_2}{dt}$, and therefore, $\sigma = (\frac{1}{T_1} - \frac{1}{T_2})\frac{dE_1}{dt} - (\frac{\mu_1}{T_1} - \frac{\mu_2}{T_2})\frac{dN_1}{dt}$. Note that an important ingredient in our consideration has been the inclusion of implicit relaxation mechanisms (e.g., the open-system limit or phonons) in each lead that equilibriate high-energy electrons. This allows us to characterize the lead i by a temperature T_i even in the presence of nonequilibrium currents. As a result, we shall see that the entropy production contains not only a mixing term but also a term due to relaxation processes.

This expression can be related to the nonequilibrium currents across the dot by noting that in a NESS, the rate of change in energy of lead 1 is the expectation value of the energy current across the dot, $\frac{dE_1}{dt} = \langle I_E \rangle_s$, and the rate of change in particle number in lead 1 is the expectation value of the nonequilbrium particle current, $\frac{dN_1}{dt} = \langle I_N \rangle$. Thus, in terms of the currents across the dot, the rate of entropy production takes the form

$$\sigma = -\left(\frac{1}{T_1} - \frac{1}{T_2}\right)\langle I_E \rangle_s + \left(\frac{\mu_1}{T_1} - \frac{\mu_2}{T_2}\right)\langle I_N \rangle_s. \tag{2}$$

For the special case, when the temperatures of the two leads are equal, $T_1 = T_2 = T$, one has $\sigma = \frac{V\langle I \rangle_s}{T}$ where we have defined the voltage $V = \mu_1 - \mu_2$. The term in the numerator of the last expression is the familiar power of an electrical circuit, $P = \langle I \rangle_s V$. Thus, for any quantum-impurity model where the two leads are at the same temperature, the rate of entropy production has the simple interpretation as the power across the circuit divided by the temperature.

We now show how to define σ directly in terms of information-theoretic quantities involving the nonequilibrium distribution functions $f_i^{\pm}$ introduced above. We first express the energy and particle currents in terms of $f_i^{\pm}$. In a steady state the particle current $\langle I \rangle_N$ can be calculated in the time-independent picture using the relation $\langle I_N \rangle = \frac{dN_1}{dt} = \lim_{L\to\infty} \frac{N_1(L/2) - N_1(-L/2)}{L/v_F}$ with $N_1(\pm L/2) = \sum_{p_\alpha} f_1^{\pm}(p_\alpha)$ the asymptotic particle number of the outgoing and incoming electrons (e.g., [17] and references therein). In writing this expression, we have used the fact that the leads are noninteracting and that in the time-independent picture the far past (future) corresponds to $x \to -\infty(+\infty)$. One can also define the energy current in an analogous manner by the expression $\langle I_E \rangle = \frac{dE_1}{dt} = \lim_{L\to\infty} \frac{E_1(L/2) - E_1(-L/2)}{L/v_F}$ where $E(\pm L/2) = \sum_{p_\alpha} \epsilon_{p_\alpha} f_i^{\pm}(p_\alpha)$

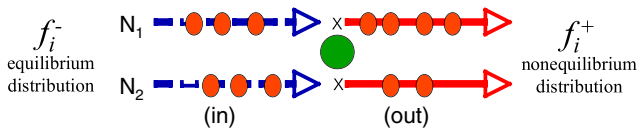

FIG. 1 (color online). Incoming and outgoing distribution functions.

measures the asymptotic energy of the incoming and outgoing particles and ϵ_{p_α} is the energy dispersion for the free electrons. Substituting these expressions into (2) and using the conservation laws relating the energy and particle number in the two leads, one can write the rate of entropy production in terms of these distribution functions as

$$\sigma = \lim_{L\to\infty} \frac{1}{L} \sum_{\alpha} \sum_{i=1,2} v_F [f_i^+(p_\alpha) - f_i^-(p_\alpha)] \frac{\epsilon_{p_\alpha} - \mu_\alpha}{T_i}. \tag{3}$$

We proceed to interpret our results in information-theoretic terms and rewrite σ in terms of the Shannon entropy current and the Kullback-Leibler distance of the probability distributions $f_i^\pm$. The Shannon entropy, $S_{\mathrm{IT}}[q(x)]$, of a probability distribution $q(x)$ measures the uncertainty of a random variable X that takes discrete values in the set $\{x_\beta\}$ with probability $q(x)$ and is defined as $S_{\mathrm{IT}}[q] \equiv -\sum_\beta q(x_\beta) \ln q(x_\beta)$ [18,19]. The relative entropy or Kullback-Leibler (KL) distance between the distribution $q(x)$ and $s(x)$,

$$D_{\mathrm{KL}}[s(x) \,\|\, q(x)] = \sum_{x_\beta} s(x_\beta) \ln \frac{s(x_\beta)}{q(x_\beta)}, \tag{4}$$

measures the inefficiency in assuming that the distribution is $q(x)$ when the true distribution is $s(x)$ [19].

The entropy of a free electron bath, usually defined thermodynamically using the partition function, can also be defined by means of information-theoretic ideas as the Shannon entropy of the Fermi-Dirac distribution function $f_{\mathrm{FD}}(p_\alpha)$ [20],

$$S_{\mathrm{IT}}[f_{\mathrm{FD}}] = -\sum_\alpha [1 - f_{\mathrm{FD}}(p_\alpha)] \ln [1 - f_{\mathrm{FD}}(p_\alpha)] - \sum_\alpha f_{\mathrm{FD}}(p_\alpha) \ln f_{\mathrm{FD}}(p_\alpha). \tag{5}$$

The advantage of the Shannon entropy as compared to thermodynamic entropy is that it has a natural generalization to nonequilibrium systems, namely, the Shannon entropy of the nonequilibrium distribution function.

To rewrite the rate of entropy production in terms of the information-theoretic quantities defined above we make use of the fact that the incoming particles from lead i are distributed according the Fermi-Dirac distribution function, $f_i^-(p_\alpha) = f_{i\mathrm{FD}}(p_\alpha)$. As a result, we know that we can write $(\epsilon_p - \mu_i)/T_i = \ln\{[1 - f_i^-(p_\alpha)]/f_i^-(p_\alpha)\}$. Substituting this expression into (3) and using the definitions (5) and (4) one gets

$$\sigma = \lim_{L\to\infty} \frac{1}{L} \sum_i v_F [S_{\mathrm{IT}}(f_i^+) - S_{\mathrm{IT}}(f_i^-)] + v_F D_{\mathrm{KL}}[f_i^+ \,\|\, f_i^-] \equiv \sum_i \Delta\sigma_i + v_F \lim_{L\to\infty} \frac{1}{L} \sum_i D_{\mathrm{KL}}[f_i^+ \,\|\, f_{i\mathrm{FD}}], \tag{6}$$

where in the second line we defined σ_i, the Shannon entropy produced per unit time at lead i. The only assumption we used is that the leads are noninteracting. Thus, this expression is valid for *all* nonequilibrium quantum-impurity models with noninteracting leads where no entropy is produced at the impurity.

The two terms in (6) have clear physical meanings. $\Delta\sigma_i$ measures the increase in the entropy per unit time of the system due to the mixing of electrons between leads. In information-theoretic terms, it measures the uncertainty about electrons as they are being transferred between leads. This term does not, however, take into account the extra entropy produced in the physical system by the relaxation to equilibrium of high-energy electrons transferred between leads. This is captured by the second term in (6), the KL distance between the nonequilibrium distribution function of the outgoing electrons of lead i and the thermal equilibrium Fermi-Dirac distribution function. Thus, in thermodynamic terms, we can interpret $T_i D_{\mathrm{KL}}[f_i^+ \,\|\, f_{i\mathrm{FD}}]$ as the work that can be extracted when a system with the nonequilibrium distribution function $f_i^+(p)$ relaxes to equilibrium at temperature T_i. A similar interpretation of the KL distance has been suggested in the theory of nonequilibrium chemical reaction [21]. In information-theoretic terms, $D_{\mathrm{KL}}[f_i^+ \,\|\, f_{i\mathrm{FD}}]$ measures the uncertainty introduced by relaxation processes. Thus, even though we have not included physical energy dissipation mechanisms such as phonons in our system, we can still extract the entropic effects produced by relaxation of high-energy electrons to equilibrium. The separation between mixing and relaxation, clear in the information-theoretic formalism, is obscured in the thermodynamic language since the energy and particle currents contribute to both relaxation and mixing.

Expression (6) allows us to adapt Shannon's original proof that a communication device always increases the entropy of a message [19] to prove that for a NESS, $\sigma > 0$. To do so, we view the scattering problem as a *classical* communication device where a transmitter at $x \to -\infty$ prepares a message—in this case the incoming particles in each lead that are to be scattered—which is then transmitted down the communication channel—in this case the scattering of the incoming electrons off the impurity resulting into outgoing particles. The receiver, at $x \to \infty$, then receives the output of this message—the outgoing electrons. Note that the messages sent by our classical device utilize only the distribution functions and not the full many-body aspects of the problem. Hence, we can apply Shannon's proof to conclude that $\Delta\sigma \geq 0$ in (6),

with equality if and only if $f_i^+ = f_i^-$. Furthermore, by the definition of the KL distance, $D_{\mathrm{KL}}[f_i^+ \| f_{i\mathrm{FD}}] \geq 0$ with equality if and only if $f_i^+ = f_{i\mathrm{FD}}$ [19]. Thus it follows that for a NESS, $\sigma > 0$. Though evident on physical grounds, mathematical proofs of positivity of entropy production are generally rare and difficult (see [2]).

Quantum impurities typically develop strong correlations on site. These correlations enter the rate of entropy production through their effect on the nonequilibrium currents, (2). As an illustration we consider the entropy production in the Kondo model and compare it to the entropy produced in the quadratic resonance level model (RLM) describing its strong coupling regime. In the experimentally relevant case where the temperatures of the two leads are identical the particle current in the high voltage regime $V \gg T_k$ is found to be $\langle I \rangle_s \sim V/\log^2[(\sqrt{V^2+T^2})/T_k]$ [3,4]. Thus, $\sigma \sim V^2/\log^2[(\sqrt{V^2+T^2})/T_k]$. At very small voltages $T \ll V \ll T_k$ the problem can be treated by linear response around the strong-coupling fixed point. The current is [22], $\langle I \rangle_s \sim V - \frac{3V^3}{2T_k^2}$ and the entropy production rate $\sigma \equiv V^2/T(1 - \frac{3V^2}{2T_k^2})$. The RLM $-i \int \psi_i^\dagger(x)\partial_x\psi_i(x) + t[d^\dagger\psi_i(0) + \mathrm{H.c.}] + \epsilon_d d^\dagger d$, consists of a local level attached to two leads of spinless electrons at different chemical potentials. The entropy production rate follows from (2): $T_1T_2\sigma = \int \frac{dp}{2\pi}[(p-\mu_2)(T_1-T_2) + VT_2] \times \frac{\Gamma^2}{(p-\epsilon_d)^2+\Gamma^2}[f_1(p) - f_2(p)]$ with $\Gamma = (1/2)t^2$ [we set $v_F = 1$, $\nu_F = 1/(2\pi)$]. The entropy production rate reduces to $\sigma \sim V^2/T(1 - \frac{V^2}{8\pi\Gamma^2})$ (taking $\mu_{1,2} = \epsilon_d \pm V/2$) at low temperatures $T_1 = T_2 = T \ll \Gamma$. Thus from the point of view of entropy production, the Kondo model at low temperatures behaves essentially as a local level with hybridization proportional to the Kondo temperature at low voltages and temperatures. This observation extends considerations, familiar in equilibrium theory, to a new nonequilibrium context. At high voltages, however, the correlations weaken, leading to an *increase* in the rate of entropy production in both the mixing and relaxation terms.

Nonequilibrium physics remains poorly understood. It is our belief that thinking about nonequilibrium physics in quantum-impurity models is likely to yield general concepts applicable to a variety of systems.

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[1] D. Goldhaber-Gordon *et al.*, Nature (London) **391**, 156 (1998); S. M. Cronenwett *et al.*, Science **281**, 540 (1998); J. Schmid *et al.*, Physica (Amsterdam) **258B**, 182 (1998).
[2] H. Spohn and J. L. Lebowitz, Commun. Math. Phys. **54**, 97 (1977); L. E. Reichl, *A Modern Course in Statistical Physics* (John-Wiley and Sons, Inc., New York, 1998).
[3] B. Doyon and N. Andrei, Phys. Rev. B **73**, 245326 (2006).
[4] A.-P. Jauho, N. S. Wingreen, and Y. Meir, Phys. Rev. B **50**, 5528 (1994); A. Rosch, J. Paaske, J. Kroha, and P. Wolfle, Phys. Rev. Lett. **90**, 076804 (2003); O. Parcollet and C. Hooley, Phys. Rev. B **66**, 085315 (2002); L. Borda, K. Vladar, and A. Zawadowski, Phys. Rev. B **75**, 125107 (2007); B. Doyon, Phys. Rev. Lett. **99**, 076806 (2007).
[5] P. Fendley, A. W. W. Ludwig, and H. Saleur, Phys. Rev. Lett. **74**, 3005 (1995); V. Bazhanov, S. Lukyanov, and A. Zamolodchikov Nucl. Phys. **B549**, 529 (1999); E. Boulat and H. Saleur (to be published).
[6] S. Hershfield, Phys. Rev. Lett. **70**, 2134 (1993); A. Schiller and S. Hershfield, Phys. Rev. Lett. **77**, 1821 (1996); J. E. Han, Phys. Rev. B **73**, 125319 (2006).
[7] P. Mehta and N. Andrei, Phys. Rev. Lett. **96**, 216802 (2006).
[8] S. Doniach and E. H. Sondheimer, *Green's Functions for Solid State Physicists* (World Scientific, Singapore, 1998).
[9] M. Gell-Mann and F. Low, Phys. Rev. **84**, 350 (1951).
[10] E. Merzbacher *Quantum Mechanics* (Wiley, New York, 1997).
[11] I. Affleck, A. Ludwig, and B. Jones, Phys. Rev. B **52**, 9528 (1995).
[12] We follow the standard practice of linearizing the spectrum so that all particles move with same Fermi velocity v_F which is used to convert times to distances.
[13] P. Mehta and N. Andrei, arXiv:cond-mat/0702612v2.
[14] As is usual in scattering theory, we quantize the momenta $\{p_\alpha\}$ by placing the system in a box of length L.
[15] S. Datta, *Electronic Transport in Mesoscopic Systems* (Cambridge University Press, Cambridge, U.K., 1995).
[16] S. R. de Groot and P. Mazur, *Nonequilibrium Thermodynamics* (North-Holland, Amsterdam, 1962).
[17] H. van Houten *et al.*, Semicond. Sci. Technol. **7**, B215 (1992).
[18] E. T. Jaynes, Phys. Rev. **106**, 620 (1957); E. T. Jaynes, Phys. Rev. **108**, 171 (1957).
[19] W. Weaver and C. E. Shannon, *The Mathematical Theory of Communication* (University of Illinois Press, Urbana, 1949); T. Cover and J. Thomas, *Elements of Information Theory* (Wiley & Sons, New York, 2006).
[20] N. Saito *et al.*, *Statistical Physics I* (Springer, New York, 1992).
[21] I. Procaccia and R. Levine, J. Chem. Phys. **65**, 3357 (1976).
[22] M. Pustilnik and L. I. Glazman, J. Phys. Condens. Matter **16**, R513 (2004).