Heat Flux and Entropy Produced by Thermal Fluctuations

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We report an experimental and theoretical analysis of the energy exchanged between two conductors kept at different temperature and coupled by the electric thermal noise. Experimentally we determine, as functions of the temperature difference, the heat flux, the out-of-equilibrium variance, and a conservation law for the fluctuating entropy, which we justify theoretically. The system is ruled by the same equations as two Brownian particles kept at different temperatures and coupled by an elastic force. Our results set strong constraints on the energy exchanged between coupled nanosystems held at different temperatures.

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The fluctuations of thermodynamics variables play an important role in understanding the out-of-equilibrium dynamics of small systems [1,2], such as Brownian particles [3–7], molecular motors [8], and other small devices [9]. The statistical properties of work, heat, and entropy have been analyzed within the context of the fluctuation theorem [10] and stochastic thermodynamics [1,2] in several experiments on systems in contact with a single heat bath and driven out of equilibrium by external forces or fields [3–9]. In contrast, the important case in which the system is driven out of equilibrium by a temperature difference and energy exchange is produced only by the thermal noise has been analyzed only theoretically on model systems [11–19] but never in an experiment because of the intrinsic difficulties of dealing with large temperature differences in small systems.

We report here an experimental and theoretical analysis of the statistical properties of the energy exchanged between two conductors kept at different temperature and coupled by the electric thermal noise, as depicted in Fig. 1(a). This system is inspired by the proof developed by Nyquist [20] in order to give a theoretical explanation of the measurements of Johnson [21] on the thermal noise voltage in conductors. In his proof, assuming thermal equilibrium between the two conductors, he deduces the Nyquist noise spectral density. At that time, well before the fluctuation dissipation theorem, this was the second example, after the Einstein relation for Brownian motion, relating the dissipation of a system to the amplitude of the thermal noise. In this Letter we analyze the consequences of removing the Nyquist's equilibrium conditions, and we study the statistical properties of the energy exchanged between the two conductors kept at different temperature. This system is probably among the simplest examples where recent ideas of stochastic thermodynamics can be tested, but in spite of its simplicity the explanation of the observations is far from trivial. We measure experimentally the heat flowing between the two heath baths and show that the fluctuating entropy exhibits a conservation law. This system is very general because it is ruled by the same equations of two Brownian particles kept at different temperatures and coupled by an elastic force [13,19]. Thus it gives more insight into the properties of the heat flux produced by mechanical coupling, in the famous Feymann ratchet [22–24] widely studied theoretically [13] but never in an experiment. Therefore, our results have implications well beyond the simple system we consider here.

Such a system is sketched in Fig. 1(a). It is constituted by two resistances R_1 and R_2, which are kept at different temperature T_1 and T_2, respectively. These temperatures are controlled by thermal baths and T_2 is kept fixed at 296 K, whereas T_1 can be set at a value between 296 and 88 K using liquid nitrogen vapor as a circulating coolant. In the figure, the two resistances have been drawn with their associated thermal noise generators η_1 and η_2, whose power spectral densities are given by the Nyquist formula $|\tilde{\eta}_m|^2 = 4k_B R_m T_m$, with $m = 1, 2$ (see Eqs. (2) and (3) and Supplemental Material [25]). The coupling capacitance C controls the electrical power exchanged between the

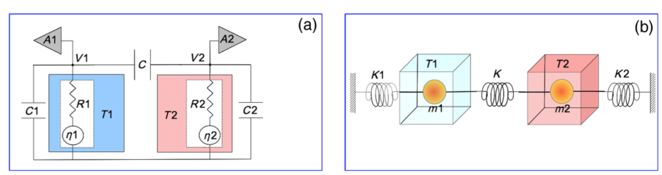

FIG. 1 (color online). (a) Diagram of the circuit. The resistances R_1 and R_2 are kept at temperature T_1 and $T_2 = 296$ K, respectively. They are coupled via the capacitance C. The capacitances C_1 and C_2 schematize the capacitance of the cables and of the amplifier inputs. The voltages V_1 and V_2 are amplified by the two low noise amplifiers A_1 and A_2 [26]. (b) The circuit in (a) is equivalent to two Brownian particles (m_1 and m_2) moving inside two different heat baths at T_1 and T_2. The two particles are trapped by two elastic potentials of stiffness K_1 and K_2 and coupled by a spring of stiffness K [see text and Eqs. (3) and (4)] The analogy with the Feymann ratchet can be made by assuming, as in Ref. [13], that the particle $m1$ has an asymmetric shape and on average moves faster in one direction than in the other.

resistances and, as a consequence, the energy exchanged between the two baths. No other coupling exists between the two resistances, which are inside two separated screened boxes. The quantities C_1 and C_2 are the capacitances of the circuits and the cables. Two extremely low noise amplifiers A_1 and A_2 [26] measure the voltage V_1 and V_2 across the resistances R_1 and R_2, respectively. All the relevant quantities considered in this Letter can be derived by the measurements of V_1 and V_2, as discussed below. In the following, we will take $C = 100$ pF, $C_1 = 680$ pF, $C_2 = 420$ pF, and $R_1 = R_2 = 10\ \mathrm{M}\Omega$, if not differently stated. When $T_1 = T_2$, the system is in equilibrium and exhibits no net energy flux between the two reservoirs. This is indeed the condition imposed by Nyquist to prove his formula, and we use it to check all the values of the circuit parameters. Applying the fluctuation dissipation theorem to the circuit, one finds the Nyquist's expression for the variance of V_1 and V_2 at equilibrium, which reads $\sigma^2_{m,\mathrm{eq}}(T_m) = k_B T_m (C + C'_m)/X$ with $X = C_2 C_1 + C(C_1 + C_2)$, $m' = 2$ if $m = 1$, and $m' = 1$ if $m = 2$. For example, one can check that at $T_1 = T_2 = 296$ K, using the above-mentioned values of the capacitances and resistances, the predicted equilibrium standard deviations of V_1 and V_2 are 2.33 μV and 8.16 μV, respectively. These are indeed the measured values with an accuracy better than 1%; see Supplemental Material [25] for further details on the system calibration.

The important quantity to consider here is the joint probability $P(V_1, V_2)$, which is plotted in Fig. 2(a) at $T_1 = T_2$ and at Fig. 2(b) at $T_1 = 88$ K. The fact that the axis of the ellipses defining the contour lines of $P(V_1, V_2)$ are inclined with respect to the x and y axis indicates that there is a certain correlation between V_1 and V_2. This correlation, produced by the electric coupling, plays a major role in determining the mean heat flux between the two reservoirs, as we discuss below. The interesting new features occur of course when $T_1 \neq T_2$. Following are the questions that we address for such a system: (1) What are the heat flux and the entropy production rate? (2) How is the variance of V_1 and V_2 modified because of the heat flux? (3) What is the role of correlation between V_1 and V_2? We will see that these questions are quite relevant and have no obvious answers because of the statistical nature of the energy transfer.

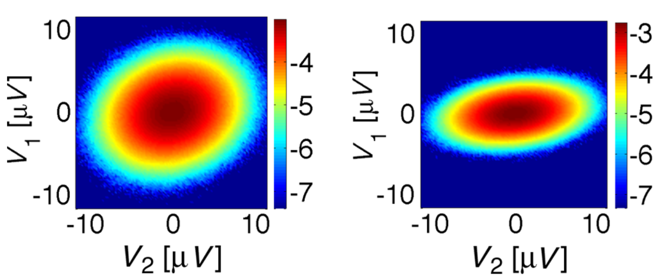

FIG. 2 (color online). The joint probability $\log_{10} P(V_1, V_2)$ measured at $T_1 = 296$ K equilibrium (a) and out of equilibrium $T_1 = 88$ K(b). The color scale is indicated on the color bar on the right side.

We consider the electric power dissipated in the resistance R_m with $m = 1, 2$, which reads $\dot{Q}_m = V_m i_m$ where i_m is the current flowing in the resistance m. The integral of the power over a time τ is the total energy Q_m, dissipated by the resistance in this time interval, i.e., $Q_{m,\tau} = \int_t^{t+\tau} i_m V_m dt$. All the voltages V_m and currents i_m can be measured; indeed, we have $i_m = i_C - i_{C_m}$ where $i_C = C\frac{d(V_2 - V_1)}{dt}$ is the current flowing in the capacitance C and $i_{C_m} = C_m dV_m/dt$ is the current flowing in C_m. Thus rearranging the terms one finds that $Q_{m,\tau} = W_{m,\tau} - \Delta U_{m,\tau}$, where $W_{1,\tau} = \int_t^{t+\tau} C V_1 (dV_2/dt) dt$, $W_{2,\tau} = \int_t^{t+\tau} C V_2 (dV_1/dt) dt$, and $\Delta U_{m,\tau} = [(C_m + C)/2] \times [V_m(t+\tau)^2 - V_m(t)^2]$ is the potential energy change of the circuit m in the time τ. Notice that W_m are the terms responsible for the energy exchange since they couple the fluctuations of the two circuits. The quantities $W_{1,\tau}$ and $W_{2,\tau}$ can be identified as the work performed by the circuit 2 on 1 and vice versa [27–29], respectively. Thus, the quantity $Q_{1,\tau}$ ($Q_{2,\tau}$) can be interpreted as the heat flowing from the reservoir 2 to the reservoir 1 (from 1 to 2), in the time interval τ, as an effect of the temperature difference. As the two variables V_m are fluctuating voltages, all the other quantities also fluctuate. In Fig. 3(a) we show the

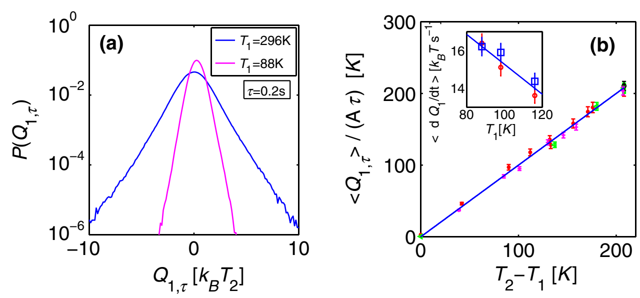

FIG. 3 (color online). (a) The probability $P(Q_{1,\tau})$ measured at $T_1 = 296$ K (blue line) equilibrium and $T_1 = 88$ K (magenta line) out of equilibrium. Notice that the peak of the $P(Q_{1,\tau})$ is centered at zero at equilibrium and shifted towards a positive value out of equilibrium. The amount of the shift is very small and is $\sim k_B(T_2 - T_1)$. (b) The measured mean value of $\langle Q_{1,\tau}\rangle$ is a linear function of $(T_2 - T_1)$. The red points correspond to measurements performed with the values of the capacitance C_1, C_2, C given in the text and $\tau = 0.2s$. The other symbols and colors pertain to different values of these capacitance and other τ: (black circles) $\tau = 0.4s$, $C = 1000$ pF, (green left triangles) $\tau = 0.1s$, $C = 100$ pF, (magenta plus) $\tau = 0.5s$, $C = 100$ pF. The values of $\langle Q_{1,\tau}\rangle$ have been rescaled by the parameter-dependent theoretical prefactor A, which allows the comparison of different experimental configurations. The continuous blue line with slope 1 is the theoretical prediction of Eq. (7). In the inset the values of $\langle \dot{Q}_1\rangle$ (at $C = 1000$ pF) directly measured using $P(Q_1)$ (blue square) are compared with those (red circles) obtained from the equality $\langle \dot{Q}_1\rangle = (\sigma_1^2 - \sigma^2_{1,\mathrm{eq}})/R_1$, as discussed in the text.

probability density function $P(Q_{1,\tau})$ at various temperatures: we see that $Q_{1,\tau}$ is a strongly fluctuating quantity, whose $P(Q_{1,\tau})$ has long exponential tails.

Notice that although for $T_1 < T_2$ the mean value of $Q_{1,\tau}$ is positive, instantaneous negative fluctuations can occur; i.e., sometimes the heat flux is reversed. The mean values of the dissipated heats are expected to be linear functions of the temperature difference $\Delta T = T_2 - T_1$, i.e., $\langle Q_{1,\tau}\rangle = A\tau\Delta T$, where A is a parameter-dependent quantity that can be obtained explicitly from Eqs. (3) and (4) below. This relation is confirmed by our experimental results, as shown in Fig. 3(b). Furthermore, the mean values of the dissipated heat satisfy the equality $\langle Q_2\rangle = -\langle Q_1\rangle$, corresponding to an energy conservation principle: the power extracted from the bath 2 is dissipated into the bath 1 because of the electric coupling. This mean flow produces a change of the variances $\sigma_m^2(T_m)$ of V_m with respect to the equilibrium value $\sigma_{m,\text{eq}}^2(T_m)$, i.e., the equilibrium value measured when the two baths are at the same temperature T_m. Specifically, we find $\sigma_m^2(T_m) = \sigma_{m,\text{eq}}^2(T_m) + \langle\dot{Q}_m\rangle R_1$, which is an extension to two temperatures of the Harada-Sasa relation [30] (see also Supplemental Material [25] for a theoretical proof of this experimental result). This result is shown in the inset of Fig. 3(b), where the values of $\langle\dot{Q}_m\rangle$ directly estimated from the experimental data [using the steady state $P(Q_m)$] are compared with those obtained from the difference of the variances of V_1 measured in equilibrium and out of equilibrium. The values are comparable within error bars and show that the out-of-equilibrium variances are modified only by the heat flux. It is now important to analyze the entropy produced by the total system, circuit plus heat reservoirs. We consider first the entropy $\Delta S_{r,\tau}$ due to the heat exchanged with the reservoirs, which reads $\Delta S_{r,\tau} = Q_{1,\tau}/T_1 + Q_{2,\tau}/T_2$. This entropy is a fluctuating quantity as both Q_1 and Q_2 fluctuate, and its average in a time τ is $\langle\Delta S_{r,\tau}\rangle = \langle Q_{r,\tau}\rangle(1/T_1 - 1/T_2) = A\tau(T_2 - T_1)^2/(T_2 T_1)$. However, the reservoir entropy $\Delta S_{r,\tau}$ is not the only component of the total entropy production: one has to take into account the entropy variation of the system, due to its dynamical evolution. Indeed, the state variables V_m also fluctuate as an effect of the thermal noise and, thus, if one measures their values at regular time intervals, one obtains a "trajectory" in the phase space $(V_1(t), V_2(t))$. Thus, following Seifert [31], who developed this concept for a single heat bath, one can introduce a trajectory entropy for the evolving system $S_s(t) = -k_B \log P(V_1(t), V_2(t))$, which extends to nonequilibrium systems the standard Gibbs entropy concept. Therefore, when evaluating the total entropy production, one has to take into account the contribution over the time interval τ,

$$\Delta S_{s,\tau} = -k_B \log\left[\frac{P(V_1(t+\tau), V_2(t+\tau))}{P(V_1(t), V_2(t))}\right]. \qquad (1)$$

It is worth noting that the system we consider is in a nonequilibrium steady state, with a constant external driving ΔT. Therefore the probability distribution $P(V_1, V_2)$ [as shown in Fig. 2(b)] does not depend explicitly on the time, and $\Delta S_{s,\tau}$ is nonvanishing whenever the final point of the trajectory is different from the initial one, $(V_1(t+\tau), V_2(t+\tau)) \neq (V_1(t), V_2(t))$. Thus, the total entropy change reads $\Delta S_{\text{tot},\tau} = \Delta S_{r,\tau} + \Delta S_{s,\tau}$, where we omit the explicit dependence on t, as the system is in a steady state as discussed above. This entropy has several interesting features. The first one is that $\langle\Delta S_{s,\tau}\rangle = 0$, and as a consequence $\langle\Delta S_{\text{tot}}\rangle = \langle\Delta S_r\rangle$, which grows with increasing ΔT. The second and most interesting result is that independently of ΔT and of τ, the following equality always holds:

$$\langle\exp(-\Delta S_{\text{tot}}/k_B)\rangle = 1, \qquad (2)$$

for which we both find experimental evidence, as discussed in the following, and provide a theoretical proof in Supplemental Material [25]. Equation (2) represents an extension to two temperature sources of the result obtained for a system in a single heat bath driven out of equilibrium by a time-dependent mechanical force [6,31], and our results provide the first experimental verification of the expression in a system driven by a temperature difference. Equation (2) implies that $\langle\Delta S_{\text{tot}}\rangle \geq 0$, as prescribed by the second law. From symmetry considerations, it follows immediately that, at equilibrium ($T_1 = T_2$), the probability distribution of ΔS_{tot} is symmetric, $P_{\text{eq}}(\Delta S_{\text{tot}}) = P_{\text{eq}}(-\Delta S_{\text{tot}})$. Thus Eq. (2) implies that the probability density function of ΔS_{tot} is a Dirac δ function when $T_1 = T_2$, i.e., the quantity ΔS_{tot} is rigorously zero in equilibrium, both in average and fluctuations, and so its mean value and variance provide a measure of the entropy production. The measured probabilities $P(\Delta S_r)$ and $P(\Delta S_{\text{tot}})$ are shown in Fig. 4(a). We see that $P(\Delta S_r)$ and $P(\Delta S_{\text{tot}})$ are quite different and that the latter is close to a Gaussian and reduces to a

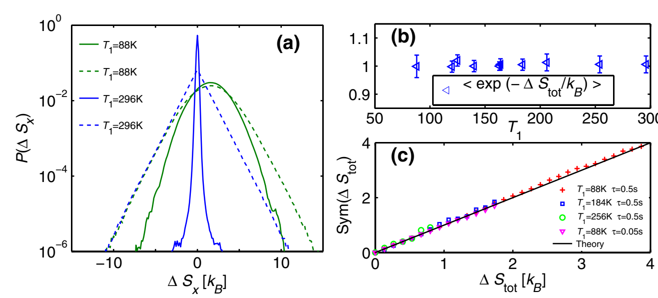

FIG. 4 (color online). (a) The probability $P(\Delta S_r)$ (dashed lines) and $P(\Delta S_{\text{tot}})$ (continuous lines) measured at $T_1 = 296$ K (blue line), which corresponds to equilibrium and $T_1 = 88$ K (green lines) out of equilibrium. Notice that both distributions are centered at zero at equilibrium and shifted towards positive value in the out of equilibrium. (b) $\langle\exp(-\Delta S_{\text{tot}})\rangle$ as a function of T_1 at two different $\tau = 0.5s$ and $\tau = 0.1s$. (c) Symmetry function $\text{Sym}(\Delta S_{\text{tot}}) = \log[P(\Delta S_{\text{tot}})/P(-\Delta S_{\text{tot}})]$ as a function of ΔS_{tot}. The black straight line of slope 1 corresponds to the theoretical prediction.

Dirac δ function in equilibrium, i.e., $T_1 = T_2 = 296$ K (notice that in Fig. 4(a), the small broadening of the equilibrium $P(\Delta S_{\rm tot})$ is just due to unavoidable experimental noise and discretization of the experimental probability density functions). The experimental measurements satisfy Eq. (2) as it is shown in Fig. 4(b). It is worth noting that Eq. (2) implies that $P(\Delta S_{\rm tot})$ should satisfy a fluctuation theorem of the form $\log[P(\Delta S_{\rm tot})/P(-\Delta S_{\rm tot})] = \Delta S_{\rm tot}/k_B$, $\forall \tau$, ΔT, as discussed extensively in Refs. [1,32]. We clearly see in Fig. 4(c) that this relation holds for different values of the temperature gradient. Thus, this experiment clearly establishes a relationship between the mean and the variance of the entropy production rate in a system driven out of equilibrium by the temperature difference between two thermal baths coupled by electrical noise. Because of the formal analogy with Brownian motion, the results also apply to mechanical coupling as discussed in the following.

We will now give a theoretical interpretation of the experimental observations. This will allow us to show the analogy of our system with two interacting Brownian particles coupled to two different temperatures; see Fig. 1(b). Let q_m ($m = 1, 2$) be the charges that have flowed through the resistances R_m, so the instantaneous current flowing through them is $i_m = \dot{q}_m$. A circuit analysis shows that the equations for the charges are

$$R_1\dot{q}_1 = -q_1\frac{C_2}{X} + (q_2 - q_1)\frac{C}{X} + \eta_1, \tag{3}$$

$$R_2\dot{q}_2 = -q_2\frac{C_1}{X} + (q_1 - q_2)\frac{C}{X} + \eta_2, \tag{4}$$

where η_m is the usual white noise, $\langle\eta_i(t)\eta_j(t')\rangle = 2\delta_{ij}k_B T_i R_j \delta(t - t')$. The relationships between the measured voltages and the charges are

$$q_1 = (V_1 - V_2)C + V_1 C_1, \tag{5}$$

$$q_2 = (V_1 - V_2)C - V_2 C_2. \tag{6}$$

Equations (3) and (4) are the same as those for the two coupled Brownian particles sketched in Fig. 1(b) by considering q_m the displacement of the particle m, i_m its velocity, $K_m = 1/C_m$ the stiffness of the spring m, and $K = 1/C$ the coupling spring. With this analogy we see that our definition of the heat flow Q_m corresponds exactly to the work performed by the viscous forces and by the bath on the particle m, and it is consistent with the stochastic thermodynamics definition [1,27,29,33,34].

Thus our theoretical analysis and the experimental results apply to both interacting mechanical and electrical systems coupled to baths at different temperatures. Starting from Eqs. (3) and (4), we can prove (see Supplemental Material [25]) that Eq. (2) is an exact result and that the average dissipated heat rate is

$$\langle\dot{Q}_1\rangle = A(T_2 - T_1) = \frac{C^2\Delta T}{XY}, \tag{7}$$

with $Y = [(C_1 + C)R_1 + (C_2 + C)R_2]$, and $A = C^2/(XY)$ is the parameter used to rescale the data in Fig. 3(b).

To conclude, we have studied experimentally the statistical properties of the energy exchanged between two heat baths at different temperature that are coupled by electric thermal noise. We have measured the heat flux and the entropy production rate, and we have shown the existence of a conservation law for entropy which imposes the existence of a fluctuation theorem which is not asymptotic in time. Our results, which are theoretically proved, are very general since the electric system considered here is ruled by the same equations as for two Brownian particles, held at different temperatures and mechanically coupled. Therefore, these results set precise constraints on the energy exchanged between coupled nano- and microsystems held at different temperatures. We finally mention that for the quantity W_i, an asymptotic fluctuation theorem can be proved both experimentally and theoretically, and this will be the subject of a Letter in preparation.

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