Bidirectional single-electron counting and the fluctuation theorem

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(Received 24 December 2009; revised manuscript received 9 March 2010; published 29 March 2010)

We investigate the direction-resolved full counting statistics of single-electron tunneling through a double quantum-dot system and compare with predictions of the fluctuation theorem (FT) for Markovian stochastic processes. Experimental data obtained for GaAs/GaAlAs heterostructures appear to violate the FT. After analyzing various potential sources for the discrepancy we conclude that the nonequilibrium shot noise of the quantum point contact electrometer, which is used to study the transport, induces strong dot-level fluctuations which significantly influence the tunneling statistics. Taking these modifications into account we find consistency with the FT.

DOI: 10.1103/PhysRevB.81.125331 PACS number(s): 73.23.Hk, 72.70.+m, 05.70.Ln

According to the second law of thermodynamics, the entropy of a macroscopic system driven out of equilibrium increases with time until equilibrium is reached. Thus the dynamics of such a system is irreversible. In contrast, for a mesoscopic system performing a random trajectory in phase space and measured during a sufficiently short time, the entropy may either increase or decrease. The "fluctuation theorem" (FT), which relies only on the microreversiblity of the underlying equation of motion, states that the probability distribution $P_\tau(\Delta S)$ for processes increasing or decreasing the entropy by $\pm\Delta S$ during a time interval τ obeys the relation

$$\frac{P_\tau(-\Delta S)}{P_\tau(\Delta S)} = \exp(-\Delta S). \quad (1)$$

From Eq. (1) one can also derive a relation between the total probabilities of negative and positive entropy changes

$$\frac{\sum_{\Delta S<0} P_\tau(\Delta S)}{\sum_{\Delta S>0} P_\tau(\Delta S)} = \frac{\sum_{\Delta S>0} P_\tau(\Delta S)e^{-\Delta S}}{\sum_{\Delta S>0} P_\tau(\Delta S)} = \langle \exp(-\Delta S)\rangle_{\Delta S>0}. \quad (2)$$

Here the summation runs over all possible processes resulting in positive or negative ΔS.

The entropy in Eqs. (1) and (2) is defined as the work W, done by external forces for a given realization of a random nonequilibrium process, divided by temperature T, i.e., $\Delta S = W/T$. It differs from Gibbs entropy used in statistical mechanics or the Shannon entropy considered in the information theory. The latter two entropies are uniquely defined and therefore do not fluctuate.

The FT remains valid even far from equilibrium. It has been proven for thermalized Hamiltonian systems,[1] Markovian stochastic processes,[2,3] and mesoscopic conductors.[4–8] The FT is fundamentally important for transport theory, one of its consequences being the Jarzynski equality,[9,10] which, in turn, leads to the second law of thermodynamics. It also leads to the fluctuation-dissipation theorem and Onsager symmetry relations,[11] including the extensions to nonlinear transport.[4–8]

In electron transport experiments the entropy production is related to Joule heating, and for a system isolated from electromagnetic environment reads $\Delta S = qeV_S/T$. Here q is the number of electron charges e transferred across a voltage drop V_S and T is the temperature (we put $k_B=\hbar=1$). Hence the FT can be formulated in terms of the distribution of transferred charge $P_\tau(q)$ at sufficiently long times, $\tau \gtrsim e/I$ (I is the electric current), as follows:

$$\frac{P_\tau(-q)}{P_\tau(q)} = \exp\left(-\frac{qeV_S}{T}\right), \quad (3)$$

or, corresponding to Eq. (2), in the integrated form as

$$\frac{\sum_{q=-\infty}^{0} P_\tau(q)}{\sum_{q=0}^{\infty} P_\tau(q)} = \frac{\sum_{q=0}^{\infty} P_\tau(q)e^{-qeV_S/T}}{\sum_{q=0}^{\infty} P_\tau(q)}. \quad (4)$$

The FT has been experimentally verified in chemical physics at room temperature[12] while tests of the FT in mesoscopic electron transport experiments in the low-temperature regime have been lacking so far. On the other hand, recent advances in time-resolved charge detection by quantum point contacts (QPCs) (Refs. 13–16) made it possible to measure the distribution $P_\tau(q)$ for single-electron tunneling through quantum dots. This opens the possibility to test the FT in mesoscopic transport.

In this paper we report on experiments on the direction-resolved full counting statistics (FCS) of single-electron tunneling in a double quantum dot (DQD) system, which is probed by an asymmetrically coupled QPC.[14] We analyze the experimental data in the frame of the FT and find that the forms (3) and (4) appear to be violated. However, they are

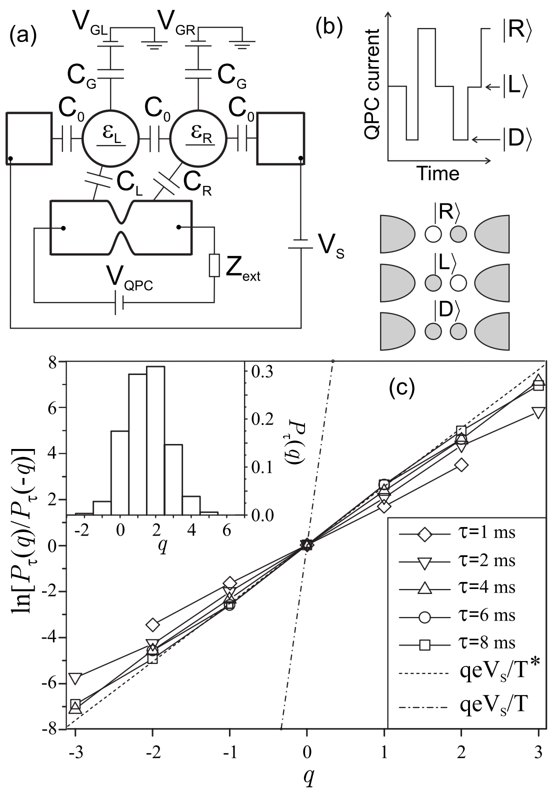

FIG. 1. (a) Setup of the system with two quantum dots (DQD) with single-level energies ε_L and ε_R coupled to a QPC. (b) The QPC current switches between three values corresponding to the three charge states of the DQD. (c) Test of FT, Eq. (3), at several times. Lines with symbols: logarithm of left-hand side (lhs) of Eq. (3) at several times; dashed line: qeV_S/T^* with $T^*=1.37$ K; dot-dashed line: qeV_S/T. Inset: the distribution $P_\tau(q)$ at $\tau=4$ ms.

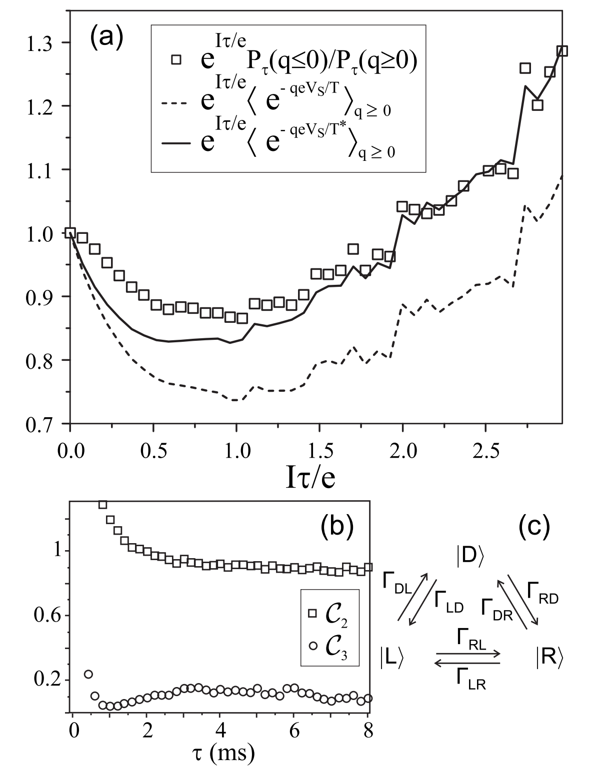

FIG. 2. (a) Test of the integrated FT, Eq. (4), in the time interval $0<\tau<8$ ms. The time scale is set by the frequency of electron tunneling $I/e\approx370$ Hz. Squares: lhs of Eq. (4) [denoted as $P_\tau(q\leq0)/P_\tau(q\geq0)$]; dashed line: rhs of Eq. (4) [denoted as $\langle\exp(-qeV_S/T)\rangle_{q\geq0}$]; solid line: rhs of Eq. (4) with T replaced by $T^*=1.37$ K. All three curves are multiplied by $\exp(I\tau/e)$ for clarity. (b) Normalized second, $\mathcal{C}_2=\langle(q-\langle q\rangle)^2\rangle/\langle q\rangle$, and third cumulants, $\mathcal{C}_3=\langle(q-\langle q\rangle)^3\rangle/\langle q\rangle$, of the charge distribution $P_\tau(q)$. (c) The six transitions with rates Γ_{ij} between three charge states.

satisfied, if we replace the temperature by an enhanced value T^*. After exploring various potential sources of this apparent heating we conclude that it arises mostly due to the shot noise of the QPC detector,[17] for which we provide quantitative estimates consistent with experimental parameters. Moreover, we note that for Markovian stochastic processes defined by general rates, the forms (1) and (2) of the FT are satisfied if we relate the entropy change to a ratio of the relevant nonequilibrium tunneling rates.

I. EXPERIMENTAL TEST OF FT

The setup of our experiment[14] consisting of a DQD coupled to a QPC detector is shown in Fig. 1(a). The left and right gate voltages, V_{GL} and V_{GR}, applied to the quantum dots are tuned in such a way that only three charge states of the DQD, $|L\rangle$, $|R\rangle$, and $|D\rangle$, need to be considered. They denote states where the left or right dot is occupied with a single electron, or where both dots are occupied, respectively. Accordingly, the current through the QPC, which is coupled asymmetrically to the DQD, switches between three different values [Fig. 1(b)]. This setup allows distinguishing electron tunneling in different directions. For example, the switching $|L\rangle\rightarrow|R\rangle$ corresponds to the transfer of one electron from the left to the right dot while $|R\rangle\rightarrow|L\rangle$ signals a transfer in opposite direction. From the time trace of the current taken during 67 s one obtains the distribution of charges transferred during shorter time intervals τ, $P_\tau(q)$, an example being shown in the inset of Fig. 1(c).

In Fig. 1(c) we perform a direct test of the FT, Eq. (3). The combination $\ln[P_\tau(q)/P_\tau(-q)]$ depends indeed linearly on the transferred charge q, approaching for long time τ a slope eV_S/T^*. Here $V_S=300$ μV is the applied DQD bias voltage but the effective temperature $T^*=1.37$ K fitting the data (dashed line) strongly exceeds the bath temperature of the leads of $T=130$ mK (dot-dashed line). To further test the time dependence contained in the FT we check in Fig. 2(a) the integrated form (4). Plotting both sides assuming the value of the electron temperature, $T=130$ mK, we observe a clear discrepancy between both (open squares and dashed line). However, by adjusting the temperature in the right-hand side (rhs) of Eq. (4) to the effective temperature $T^*=1.37$ K (solid line), we get a good fit for $\tau\gtrsim e/I$.

The experimental distribution of the transferred charge [inset of Fig. 1(c)] deviates strongly from a Gaussian shape. For illustration the second and the third cumulants of the charge distribution are plotted in Fig. 2(b). The normalized third cumulant remains close to 0.1 at all times. This observation additionally confirms a nontrivial nature of our result

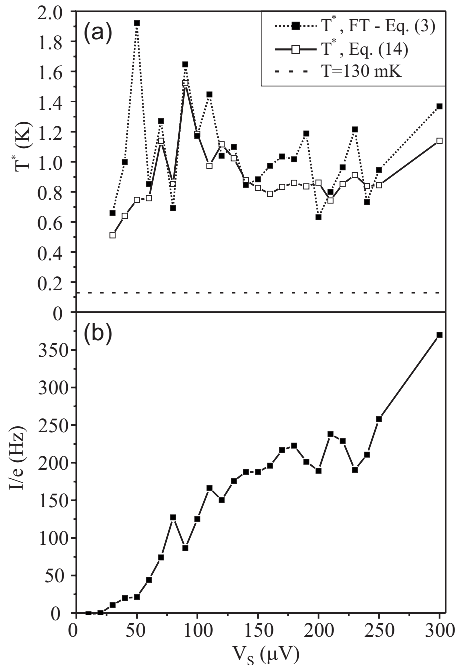

FIG. 3. (a) Effective temperature T^* versus bias voltage V_S. Black squares and dotted line: T^* needed to satisfy FT, Eq. (3); empty squares and solid line: T^* obtained from Eq. (14); dashed line: temperature of the leads $T=130$ mK. (b) Frequency of electron tunneling I/e versus bias voltage.

since for a Gaussian distribution $P_\tau(q)$ one can always tune an effective temperature T^* to satisfy Eq. (3).

In order to test the dependence of the effective temperature T^* on the bias voltage V_S we have repeated this analysis for a set of 23 bias voltages in the range from 30 to 250 μV. The result is shown in Fig. 3(a). For every value of V_S the QPC current data have been acquired for a relatively short period of 1.3 s. This limited the total number of electron tunneling events and, hence, the accuracy with which T^* could be determined. The lack of statistics was especially evident at low bias voltages where the electron tunneling frequency, shown in Fig. 3(b), was very low. The statistical uncertainty of T^* in this limit exceeded 50%. In spite of these uncertainties, Fig. 3(a) clearly demonstrates that T^* has only a rather weak, and nonmonotonous, dependence on V_S.

II. VIOLATION AND RECOVERY OF FT

Why does the FT appear to be violated? In a systematic approach, the FT for the system composed of a DQD and the QPC, depicted in Fig. 1(a), should be formulated in terms of the joint probability distribution $P_\tau(q,q')$ for charges q and q' transmitted through the DQD and the QPC, respectively.[7] It satisfies

$$P_\tau(q,q') = \exp\left(\frac{qeV_S + q'eV_{\mathrm{QPC}}}{T}\right)P_\tau(-q,-q'), \tag{5}$$

where T is the temperature of the leads. Since only the number of charges q is measured, Eq. (5) should be summed over q'. Performing this operation at $V_{\mathrm{QPC}} \neq 0$ and introducing the probability distribution $P_\tau(q) \equiv \Sigma_{q'} P_\tau(q,q')$, we observe that

$$P_\tau(q) \neq \exp\left(\frac{qeV_S}{T}\right)P_\tau(-q), \tag{6}$$

which appears to violate the FT. In Sec. III we provide a microscopic analysis of this violation.

Then why is the FT recovered by introducing one single fit parameter, the effective temperature T^*? In the experiment, the DQD is in the sequential tunneling regime and the probability distribution $P_\tau(q)$ can be found within the master equation approach to FCS.[3,18,19] It builds up on a "modified" master equation

$$\partial_t \boldsymbol{p}(t) = \boldsymbol{\Gamma}(\lambda)\boldsymbol{p}(t), \tag{7}$$

where $\boldsymbol{p}^T=(p_L,p_R,p_D)$, denotes the occupation probabilities of the three charge states (L, R, and D) of the two quantum dots. The transition matrix $\boldsymbol{\Gamma}(\lambda)$ between these three states

$$\boldsymbol{\Gamma}(\lambda) = \begin{bmatrix} -\Gamma_{RL}-\Gamma_{DL} & \Gamma_{LR}e^{-i\lambda} & \Gamma_{LD} \\ \Gamma_{RL}e^{i\lambda} & -\Gamma_{LR}-\Gamma_{DR} & \Gamma_{RD} \\ \Gamma_{DL} & \Gamma_{DR} & -\Gamma_{LD}-\Gamma_{RD} \end{bmatrix} \tag{8}$$

is modified to include the counting field λ, which allows deriving the statistics of the electron transfer. The characteristic function (CF) then takes the form[18]

$$\mathcal{Z}_\tau(\lambda) = \sum_q P_\tau(q)e^{iq\lambda} = \boldsymbol{e}^T e^{\tau\boldsymbol{\Gamma}(\lambda)}\boldsymbol{p}^{st}, \tag{9}$$

where $\boldsymbol{e}^T=(1,1,1)$ and $\boldsymbol{p}^{st}$ is the stationary solution of $\boldsymbol{\Gamma}(0)\boldsymbol{p}^{st}=0$. In the long-time limit, $\tau \gg e/I$, the CF assumes an exponential form

$$\mathcal{Z}_\tau(\lambda) \approx e^{\tau\mathcal{F}(\lambda)}, \tag{10}$$

where $\mathcal{F}(\lambda)$ is the eigenvalue of the matrix $\boldsymbol{\Gamma}(\lambda)$ with largest real part. It satisfies

$$0 = \det[\boldsymbol{\Gamma}(\lambda) - \mathcal{F}\mathbf{I}] = \mathcal{F}^3 + K\mathcal{F}^2 + K'\mathcal{F} - \Gamma_{DR}\Gamma_{RL}\Gamma_{LD}(e^{i\lambda}-1) - \Gamma_{DL}\Gamma_{LR}\Gamma_{RD}(e^{-i\lambda}-1), \tag{11}$$

where $\mathbf{I}$ is the identity matrix. where K and K' are parameters independent of the counting field,

$$K = \sum_{i,j=L,R,D} \Gamma_{ij},$$

$$K' = \Gamma_{LD}\Gamma_{DR} + \Gamma_{LR}\Gamma_{RD} + \Gamma_{RD}\Gamma_{DL} + \Gamma_{DL}\Gamma_{LR} + \Gamma_{DR}\Gamma_{RL} + \Gamma_{RL}\Gamma_{LD} + \Gamma_{LD}\Gamma_{LR} + \Gamma_{DL}\Gamma_{DR} + \Gamma_{RD}\Gamma_{RL}. \tag{12}$$

Since the parameters K and K' are independent of λ, we observe without solving Eq. (11), that the CF satisfies the identity,

$$\mathcal{Z}(\lambda) = \mathcal{Z}(-\lambda + ieV_S/T^*), \tag{13}$$

with effective temperature

$$T^* = eV_S/\mathcal{A}, \quad \mathcal{A} = \ln\frac{\Gamma_{DR}\Gamma_{RL}\Gamma_{LD}}{\Gamma_{DL}\Gamma_{LR}\Gamma_{RD}}. \tag{14}$$

Performing the inverse Fourier transform of Eq. (13), we arrive at the FT in the form (3) with T replaced by T^*.

As we will see later, the effective temperature T^*, Eq. (14), becomes equal to the base temperature T if the local detailed balance holds. At finite QPC bias voltage the detailed balance is violated, but even in this case the FT in the form (1) formally remains valid if we identify the parameter $\mathcal{A}$ with the entropy production per unit transferred electron, $\mathcal{A} \to \Delta S/q$.

The argument of the logarithm $\mathcal{A}$, Eq. (14), is the ratio of products of the tunneling rates corresponding to forward and backward cycles [i.e., the counterclockwise and clockwise cycles shown in Fig. 2(c)]. While in our simple system with three states only one such cycle exists, in a general system many different cycles are possible. For every cycle one can construct a logarithm, or affinity,[3] $\mathcal{A}_\alpha$ in the same way as we did in Eq. (14). These affinities are not necessarily all the same and it is in general not possible to define a unique effective temperature.[3]

In the experiments the tunneling rates at $V_S=300$ μV are estimated as $\Gamma_{DR}=4$ kHz, $\Gamma_{RD}=0.3$ kHz, $\Gamma_{DL}=1$ kHz, $\Gamma_{LD}=1.5$ kHz, $\Gamma_{LR}=1.7$ kHz, and $\Gamma_{RL}=1.8$ kHz. The values lie in a regime where they do not suffer from the finite bandwidth of the QPC detector ($\gtrsim 10$ kHz). Using these rates we obtain from Eq. (14) the effective temperature $T^*=1.14$ K, which agrees well with the value 1.37 K obtained from the fit of the FT. Within the statistical uncertainty this agreement actually persists at all bias voltages as shown in Fig. 3, where the comparison between the effective temperatures derived from FT, Eq. (3), and from Eq. (14) is made.

III. QPC BACKACTION

The tunnel rates are influenced by environmental effects. In particular, the DQD is coupled to the QPC via the capacitors C_L and C_R indicated in Fig. 1(a). The nonequilibrium current noise of the QPC produces fluctuations of the potentials of the quantum dots $e\delta V_{L,R}$, which in turn influence the tunnel rates[15,20] as known from the so-called $P(E)$ theory.[21] Introducing three such functions $P_j(E)$ with $j=L,R,dd$ we find

$$\Gamma_{RL} = 2\pi |t_{dd}|^2 P_{dd}(E_L - E_R), \tag{15}$$

$$\Gamma_{DL} = \Gamma_R \int dE f(E_D - E_L - \mu_R - E) P_R(E), \tag{16}$$

$$\Gamma_{DR} = \Gamma_L \int dE f(E_D - E_R - \mu_L + E) P_L(E). \tag{17}$$

Here t_{dd} is the matrix element describing tunneling between the two dots, the rates Γ_L and Γ_R characterize the coupling between the dots and the leads. $f(E)$ is the Fermi function, where we fix the chemical potentials of the leads as $\mu_L = -\mu_R = eV_S/2$. The energies of the charge states $E_{L,R,D}$ include the electrostatic energy of the electric field stored in the capacitors. The rates Γ_{LR}, Γ_{LD}, and Γ_{RD} are given by the same expressions where the Fermi function f should be replaced by $1-f$ and the argument of the functions P_j should change sign.

The spectral functions $P_{L/R/dd}$ can be expressed in terms of the phase operators $\hat{\varphi}_{L/R}(t)=\int^t dt' \, \delta\hat{V}_{L/R}(t')$ and $\hat{\varphi}_{dd}=\hat{\varphi}_R - \hat{\varphi}_L$ as follows:

$$P_j(E) = \int \frac{dt}{2\pi} e^{iEt} \langle e^{i\hat{\varphi}_j(t)} e^{-i\hat{\varphi}_j(0)} \rangle \approx \int \frac{dt}{2\pi} e^{iEt} \times \exp\left[\int \frac{d\omega}{2\pi} \frac{e^2[S_{0,j}(\omega) + S_{V,j}^{\mathrm{QPC}}(\omega)](e^{-i\omega t} - 1)}{\omega^2} \right]. \tag{18}$$

Here $S_{0,j}(\omega)$ is the spectral density of thermal fluctuations of equilibrium environments, including the impedance of the external circuit Z_{ext} [Fig. 1(a)], equilibrium phonons, etc. Within the Gaussian approximation the part $S_{V,j}^{\mathrm{QPC}}$ is proportional to the nonequilibrium and nonsymmetrized current noise of the QPC as $S_{V,j}^{\mathrm{QPC}}(\omega) = \kappa_j^2 |Z_t(\omega)|^2 S_I^{\mathrm{QPC}}(\omega)$,[20] where

$$S_I^{\mathrm{QPC}} = \frac{e^2}{\pi} \left[\sum_{\pm} \frac{\mathcal{T}(1-\mathcal{T})(\omega \pm eV_{\mathrm{QPC}})}{1 - e^{-(\omega \pm eV_{\mathrm{QPC}})/T}} + \frac{2\mathcal{T}^2 \omega}{1 - e^{-\omega/T}} \right].$$

Here $\mathcal{T}$ is the QPC transparency and $Z_t(\omega) = [-i\omega\bar{C} + 1/\bar{R}]^{-1}$ is the impedance of the electromagnetic environment seen by the QPC with resistance $\bar{R} = 1/(R_{\mathrm{QPC}}^{-1} + Z_{\mathrm{ext}}^{-1})$ and capacitance $\bar{C} = (3C_0 + C_G)/2 + C_L C_R/(C_L + C_R)$. The factors κ_j characterizing the coupling between QPC and quantum dots are given by ratios of capacitances.

When the QPC is in equilibrium, $V_{\mathrm{QPC}}=0$, the functions P_j obey the detailed balance

$$P_j(E)/P_j(-E) = e^{E/T}. \tag{19}$$

From this relation one can derive the local detailed balance[2] for the tunneling rates Γ_{RD}, Γ_{DR},

$$\Gamma_{RD}/\Gamma_{DR} = e^{(E_D - E_R - \mu_L)/T}, \tag{20}$$

and similar relations for the remaining rates. Under these conditions the effective temperature, Eq. (14), coincides with the equilibrium one, i.e., $T^* = T$.

If the QPC electrometer is turned on, $V_{\mathrm{QPC}} \neq 0$ and the detailed balance, Eq. (19), is no longer satisfied. As a consequence, the local detailed balance, Eq. (20), is violated as well.[15,20] This provides a microscopic mechanism of the apparent violation of the FT discussed above.

In the experiment, the QPC is biased at a rather high voltage $eV_{\mathrm{QPC}}=800$ $\mu\mathrm{V} \gg T$. To estimate the strength of the QPC backaction in this case we have measured the tunneling rate through the central barrier Γ_{RL} at various values of $V_S, V_{\mathrm{GL}}, V_{\mathrm{GR}}$ and plotted it as a function of the energy difference $E_R - E_L$ [see, e.g., Figure 2G in Ref. 14]. The result can be well fitted by a simple Lorentzian

$$\Gamma_{RL} \approx \frac{\Gamma_{\max}}{1 + (E_R - E_L)^2/\Gamma_0^2} \tag{21}$$

with $\Gamma_{\max} \approx 7$ kHz and $\Gamma_0 \approx 30$ μeV. The theory, Eqs. (15) and (18), predicts

$$\Gamma_0 = (e^2\kappa_{dd}\bar{R})^2 \frac{eV_{\mathrm{QPC}}}{2\pi}\mathcal{T}(1-\mathcal{T}), \quad \Gamma_{\max} = \frac{2|t_{dd}|^2}{\Gamma_0}. \quad (22)$$

Comparing the theory with the experiment and taking the experimental value of $\mathcal{T}$=0.19, we find $|t_{dd}| \approx 30$ MHz, $\kappa_{dd}\bar{R} \approx 5$ kΩ. The latter value is in agreement with other experiments.[15] From Eq. (14) at V_S=300 μV we roughly estimate $T^* \sim eV_S/\ln(\pi eV_S/\Gamma_0) \approx 1$ K, in good agreement with our previous findings, thus further supporting the QPC backaction model.

Other environmental effects introduce only weak corrections to our findings. In GaAs nanostructures acoustic phonons modify the tunneling properties via piezoelectric and deformation coupling.[16,22] At experimental values of the QPC current $I_{\mathrm{QPC}} \approx 12$ nA the phonons should stay almost in equilibrium,[16] their effect is absorbed in the equilibrium part of the spectral density $S_{0,j}$ and does not affect the FT. The effect of spontaneously emitted phonons[23] can be also included this term provided that the phonon bath is big and kept at equilibrium, which is expected for GaAs nanostructures. An intrinsic backaction of the QPC, often analyzed in the context of the quantum measurement process,[24] is also estimated to be negligible. A detailed discussion of these effects within the framework of real-time diagrammatic technique,[25,26] will be published elsewhere.[27]

IV. SUMMARY

We have investigated experimentally and theoretically the fluctuation theorem for Markovian stochastic processes by studying the direction-resolved full counting statistics in a double quantum-dot system via a nearby quantum point contact electrometer. We found an apparent violation of the fluctuation theorem, which we attribute to the nonequilibrium electromagnetic fluctuations generated by the shot noise of the quantum point-contact electrometer. We demonstrated that the FT is recovered if we adopt an effective temperature which depends on the entropy production associated with the transfer of one electron through the double quantum dot. It can be expressed by a ratio of forward to backward tunneling rates. In our experiment the effective temperature turns out to be ten times higher than the bath temperature. We hope our experimental test as well as the experimental verification of the FT in Aharonov-Bohm ring,[28] would stimulate further developments of the nonequilibrium statistical physics in mesoscopic quantum transport.

ACKNOWLEDGMENTS

We thank Mattias Hettler and Kensuke Kobayashi for helpful discussions. This work has been supported by Strategic International Cooperative Program of the Japan Science and Technology Agency (JST) and by the German Science Foundation (DFG).

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