## Mesoscopic Coulomb Drag, Broken Detailed Balance, and Fluctuation Relations

Rafael Sánchez,<sup>1</sup> Rosa López,<sup>2</sup> David Sánchez,<sup>2</sup> and Markus Büttiker<sup>1</sup>

<sup>1</sup>Département de Physique Théorique, Université de Genève, CH-1211 Genève 4, Switzerland <sup>2</sup>Departament de Física, Universitat de les Illes Balears, E-07122 Palma de Mallorca, Spain (Received 16 October 2009; published 16 February 2010)

When a biased conductor is put in proximity with an unbiased conductor a drag current can be induced in the absence of detailed balance. This is known as the Coulomb drag effect. However, even in this situation far away from equilibrium where detailed balance is explicitly broken, theory predicts that fluctuation relations are satisfied. This surprising effect has, to date, not been confirmed experimentally. Here we propose a system consisting of a capacitively coupled double quantum dot where the nonlinear fluctuation relations are verified in the absence of detailed balance.

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Introduction.-Mesoscopic physics offers a unique laboratory to investigate the extension of equilibrium fluctuation-dissipation theorems into the nonlinear nonequilibrium regime [1]. The equilibrium fluctuationdissipation theorem and its closely related Onsager symmetry relations [2] are a corner stone of linear transport. It has therefore been natural to ask whether such relations exist also if the system is driven out of the linear transport regime. For steady state transport, fluctuation relations have been developed which relate higher order response functions to fluctuation properties of the system [1,3-6]. For example, the current response to second order in the voltage (the second order conductance) is related to the voltage derivative of the noise of the system and, in the presence of a magnetic field, to the third cumulant of the current fluctuations at equilibrium [1,5].

Clearly, tests of nonequilibrium fluctuation relations are of fundamental interest. From a theoretical point of view, the task is to propose tests in which crucial relations valid at equilibrium fail in the nonlinear regime and to demonstrate that, despite such a failure, fluctuation relations hold. For instance, we have suggested experiments which test fluctuation relations for systems in the presence of a magnetic field and in a regime where the Onsager relations are already known to fail [1,7]. Such experimental tests are currently under way [8]. Here we propose to test fluctuation relations in a system where away from equilibrium we have no detailed balance. We consider two quantum dots in close proximity to each other such that they interact via long-range Coulomb forces, as shown in Fig. 1. The absence of detailed balance is manifest in a Coulomb drag [9]: the charge noise of one of the systems (the driver) drives a current through the other unbiased system [10]. Therefore, the drag current is a direct indication that this fundamental symmetry is absent. Nevertheless, we demonstrate below that there exist fluctuation relations.

The interaction of two systems in close proximity to each other plays a role in many important setups in physics. We recall here only the interaction of a detector with a system to be measured [11], which also provides a test of fluctuation relations [12]. The shot noise current-current correlation in nearby quantum dots which do not exchange particles has been measured by McClure et al. [13] and discussed theoretically [14,15]. Recently, reciprocity relations of two coupled conductors were proposed by Astumian [16]. Here we emphasize that one conductor, even if unbiased, can act as a gate to the other conductor. As a consequence, the currents are not a function only of voltage differences applied to each conductor but also depend on potential differences of one conductor to the other one. In an instructive work, Levchenko and Kamenev discuss the mesoscopic Coulomb drag for two quantum point contacts in close proximity [17]. In this geometry, charging of the point contacts can be neglected and the coupling of the two conductors is *extrinsic* via the capacitance of the leads.

General theory.—The probability P(N, t) that  $N = (N_1, \ldots, N_M)$  particles are transmitted through M leads during time t characterizes the statistical properties of



FIG. 1 (color online). Sketch of two capacitively coupled quantum dots, each one attached to two different terminals. For very large intradot charging energy, only four charge states are allowed, as depicted. Their dynamics is governed by the tunneling rates  $\Gamma_l^{\pm}$  and  $\gamma_l^{\pm}$ .

our system. It is useful to consider the generating function which is the logarithm of the "Fourier transform"  $\mathcal{F}(\chi) = \ln \sum_{N} P(N, t) e^{i\chi N}$  of the distribution function. Here  $\chi$  is the vector of the counting fields. From the generating function all cumulants can be obtained by simple differentiation. The fluctuation relations are a consequence of symmetries of the generating function [3,4]. In particular (in the absence of a magnetic field), it holds that  $\mathcal{F}(i\chi) =$  $\mathcal{F}(-i\chi + qV/kT)$ , which is equivalent to P(N) = $e^{NqV/kT}P(-N)$ . Here qV/kT is the affinity vector with components given by the applied voltages  $V_i$ . By expanding the current through lead i,  $I_i = \langle \hat{I}_i \rangle$ , where  $\hat{I}_i$  is the current operator, and the current correlations  $S_{ij} =$  $\langle \Delta I_i \Delta I_i \rangle$ , where  $\Delta I_i = \hat{I}_i - I_i$ ,

$$I_{i} = \sum_{j} G_{i,j} V_{j} + \frac{1}{2} \sum_{jk} G_{i,jk} V_{j} V_{k} + \cdots, \qquad (1)$$

$$S_{ij} = S_{ij}^{\text{eq}} + \sum_{k} S_{ij,k} V_k + \cdots, \qquad (2)$$

we can relate linear response current and equilibrium fluctuations by means of the fluctuation-dissipation theorem,  $S_{ij}^{\text{eq}} = 2kTG_{i,j}$ . The generalization to the weakly non-linear regime reads [1,4–6,18]

$$S_{\alpha\beta,\gamma} + S_{\alpha\gamma,\beta} + S_{\beta\gamma,\alpha} = kT(G_{\alpha,\beta\gamma} + G_{\beta,\alpha\gamma} + G_{\gamma,\alpha\beta}).$$
(3)

Notably, we find that these nonlinear fluctuation relations are valid even in the absence of detailed balance.

To determine the general current-voltage characteristics and the nonlinear fluctuations relations for two interacting conductors, we employ the classical treatment of the Coulomb interaction that respects charge conservation (gauge invariance). We take the interaction to be *intrinsic*, determined by the charges on the mesoscopic conductors, and assume the leads to be metals with perfect screening. Then, the dynamics of the system is determined by the sequential tunneling between states with a well defined charge occupation which obeys the master equation  $\dot{\rho}(t) =$  $\mathcal{M}\rho(t)$  for the occupation probabilities [19]. Analogously, one can write the equation of motion for the generating function,  $\zeta(t) = \sum_{N} P(N, t) e^{iN\chi}$ , given by  $\dot{\zeta}(t) =$  $\mathcal{M}(\chi)\zeta(t)$ . The cumulant generating function  $\mathcal{F}$  is given by the eigenvalue of  $\mathcal{M}(\chi)$  that develops adiabatically from zero with small  $\chi$  [20]. Generally, the explicit expression for  $\mathcal{F}$  is difficult to handle, so in practice it is more convenient to calculate the cumulants recursively order by order [21,22]. Thus, from coefficients  $c_{\{l_k\}}$  of the expansion  $\mathcal{F} = \sum_{\{l_k\}} c_{\{l_k\}} (e^{i\chi_1} - 1)^{l_1} \dots (e^{i\chi_M} - 1)^{l_M}$ , we obtain the current-current correlations up to any order [21]: e.g., the current,  $I_i = q \sum_{\{l_k\}} c_{\{l_k\}} \delta_{l_{i},1} \delta_{\Sigma l_k,1}$ , the zero frequency noise,  $S_{ii} = qI_i + 2q^2 \sum_{\{l_k\}}^{(l_k)} c_{\{l_k\}} \delta_{l_{i,2}} \delta_{\Sigma l_{k,2}}$ , and the cross correlations  $S_{ij} = q^2 \sum_{\{l_k\}} c_{\{l_k\}} \delta_{l_{i,1}} \delta_{l_{j,1}} \delta_{\Sigma l_{k,2}}$ , where  $\{l_k\} = \{l_1 \dots l_M\}$  and q is the electron charge.

Drag current and fluctuation relations.—In the following we explicitly show, using the previous formalism, the fulfillment of the fluctuation relations in a nonequilibrium system where detailed balance is broken. We consider two capacitively coupled two-terminal quantum dots (see Fig. 1) with large intradot charging energy. The interdot coupling is described with a capacitance C. Hence, the dynamics is characterized by four charge states: the empty state  $|0\rangle = |00\rangle$ , the singly occupied states  $|u\rangle = |10\rangle$  and  $|d\rangle = |01\rangle$ , and the doubly occupied state  $|2\rangle = |11\rangle$ . Quite generally, the tunneling amplitudes are energy dependent. Therefore, we distinguish  $\Gamma_l$ , which denotes a tunneling process through barrier l = 1, ..., 4 when the system is empty, and  $\gamma_l$ , which corresponds to tunneling when the coupled dot is already occupied. This is the *minimal* charge model that manifests violation of detailed balance leading to drag currents. Detailed balance is broken when the probability to transfer one charge from left to right differs from the reverse process (from right to left). For instance, an electron is transported from left to right in the drag system by the sequence  $|0\rangle \rightarrow |u\rangle \rightarrow |2\rangle \rightarrow |d\rangle \rightarrow |0\rangle$  with a probability  $\propto \Gamma_1 \gamma_2$ , whereas the probability to transport it from right to left is  $\propto \gamma_1 \Gamma_2$ . Clearly, both probabilities differ and a nontrivial current, the drag current  $I_{\rm drag} \propto$  $\Gamma_1 \gamma_2 - \gamma_1 \Gamma_2$ , will be generated. We need that (i) both empty and doubly occupied states are taken into account and (ii) the tunneling rates depend on the charge state. Thus, a model with three charge states only  $(|u\rangle, |d\rangle$ , and  $|0\rangle$  or  $|2\rangle$ ) cannot break the detailed balance and the drag effect is absent. The biased dot then acts merely as a fluctuating gate on the other dot.

For the system depicted in Fig. 1, with  $kT \gg \hbar\Gamma_i$ ,  $\hbar\gamma_i$ , writing  $\zeta = (\zeta_0, \zeta_u, \zeta_d, \zeta_2)$ , the equation  $\dot{\zeta} = \mathcal{M}(\chi)\zeta$  becomes

$$\mathcal{M} = \begin{pmatrix} -\Gamma_{u}^{-} - \Gamma_{d}^{-} & \tilde{\Gamma}_{u}^{+} & \tilde{\Gamma}_{d}^{+} & 0\\ \tilde{\Gamma}_{u}^{-} & -\Gamma_{u}^{+} - \gamma_{d}^{-} & 0 & \tilde{\gamma}_{d}^{+}\\ \tilde{\Gamma}_{d}^{-} & 0 & -\gamma_{u}^{-} - \Gamma_{d}^{+} & \tilde{\gamma}_{u}^{+}\\ 0 & \tilde{\gamma}_{d}^{-} & \tilde{\gamma}_{u}^{-} & -\gamma_{u}^{+} - \gamma_{d}^{+} \end{pmatrix},$$
(4)

where  $\tilde{\Gamma}_{\alpha}^{\pm} = \sum_{l \in \alpha} e^{\pm i\chi_l} \Gamma_l^{\pm}$ , and  $\tilde{\gamma}_{\alpha}^{\pm} = \sum_{l \in \alpha} e^{\pm i\chi_l} \gamma_l^{\pm}$ ,  $u = \{1, 2\}$  and  $d = \{3, 4\}$ . The tunneling rates in(-) and out(+) of the dot read  $\Gamma_l^{\pm} = \Gamma_l f_{l0}^{\pm}$  and  $\gamma_l^{\pm} = \gamma_l f_{l1}^{\pm}$  with  $f_{ln}^{\pm} = f^{\pm}(\mu_{ln} - qV_l)$  (n = 0, 1). Here,  $f^+(\varepsilon) = 1 - f(\varepsilon)$  and  $f^-(\varepsilon) = f(\varepsilon)$  denote the hole and electron Fermi functions, respectively. The effective level of dot  $\alpha$  with bare level  $\varepsilon_{\alpha}$  when dot  $\beta \neq \alpha$  is uncharged (n = 0) is  $\mu_{\alpha 0} = \varepsilon_{\alpha} + [q^2 C_{\Sigma\alpha}/2 + q(C_{\Sigma\beta} \sum_{l \in \alpha} C_l V_l + C \sum_{l \in \beta} C_l V_l)]/C\tilde{C}$ , where  $C_l$  is the capacitance of the *l*th barrier,  $C_{\Sigma\alpha} = \sum_{l \in \alpha} C_l + C$ , and  $\tilde{C} = (C_{\Sigma u} C_{\Sigma d} - C^2)/C$ . In the charged case (n = 1), we find  $\mu_{\alpha 1} = \mu_{\alpha 0} + E_C$  with  $E_C = 2q^2/\tilde{C}$  the energy needed to add a second electron. Note that the tunneling rates through each dot do not depend on the position of the level in the other dot, but by its charge occupation.

We now investigate the drag current, for which we take the upper subsystem as the drag circuit  $(V_1 = V_2)$  and the lower one as the driver. Then,  $I_1 = -I_2 = I_{drag}$  and we find

$$I_{\text{drag}} = \frac{q(\gamma_1 \Gamma_2 - \gamma_2 \Gamma_1) \Gamma_d \gamma_d \sum_{\sigma = \{+, -\}} \sigma f_{10}^{\sigma} f_{11}^{-\sigma} g_0^{-\sigma} g_1^{\sigma}}{\Gamma_u \Gamma_d (\gamma_u h^+ + \gamma_d k^+) + \gamma_u \gamma_d (\Gamma_u h^- + \Gamma_d k^-)},$$
(5)

where  $\Gamma_{\alpha} = \sum_{l \in \alpha} \Gamma_l$ ,  $\gamma_{\alpha} = \sum_{l \in \alpha} \gamma_l$ ,  $h^{\pm} = f_{11}^{\pm} \pm g_{0(1)}^-(f_{11}^- - f_{10}^-)$ , and  $k^{\pm} = g_1^{\pm} \pm f_{10(11)}^-(g_1^- - g_0^-)$ .  $g_0^{\pm} = (\Gamma_3 f_{30}^{\pm} + \Gamma_4 f_{40}^{\pm}) / \Gamma_d$  and  $g_1^{\pm} = (\gamma_3 f_{31}^{\pm} + \gamma_4 f_{41}^{\pm}) / \gamma_d$  are nonequilibrium distribution functions.

When the drive voltage  $V_3 - V_4$  is small, detailed balance must be broken also in the drive circuit in order to have a linear  $I_{\text{drag}}$ :  $G_{2,4} \propto (\gamma_1 \Gamma_2 - \gamma_2 \Gamma_1)(\gamma_4 \Gamma_3 - \gamma_3 \Gamma_4)$ . Therefore, asymmetry in both the drag and the drive systems is required for a nonzero linear drag current. Moreover, we get  $G_{2,4} = G_{4,2}$ , satisfying the Onsager-Casimir reciprocity relations [2]. Note that if the drive conductor is also unbiased ( $V_3 = V_4$ ), equilibrium fluctuations are expectedly not enough to induce a net current. This can be seen in Fig. 2(a). For low voltages there is a Coulomb gap where transport is not allowed. This result also demonstrates that the voltage difference between the two subsystems  $V_1 - V_3$  plays a crucial role, affecting the dynamics: In this case, one of the conductors acts as a gate on the other one. The gate effect of the drag circuit onto the driver is shown in Fig. 2(b), where we obtain a typical Coulomb blockade stability diagram for the drive current  $I_3 = -I_4 = I_{drive}$ . The linear drag current is exponentially suppressed at low temperatures (compared to the charging energy) and decays as 1/T for higher ones. This differs from the most usual  $T^2$  behavior [9,17].



FIG. 2 (color online). Voltage dependence of (a) the drag current  $I_{\text{drag}}$  through the upper dot  $(V_1 = V_2)$ , (b) the drive current  $I_{\text{drive}}$  for the lower dot, (c) the current-current correlations in the drive system,  $S_{22} = \langle \Delta I_2 \Delta I_2 \rangle$ , and (d) the cross correlation between currents at different conductors,  $S_{24} = \langle \Delta I_2 \Delta I_4 \rangle$ , for the drag configuration,  $V_1 = V_2$ . Parameters:  $\Gamma_i = \gamma_i = \Gamma$ , except  $\gamma_1 = 0.1\Gamma$ ,  $kT = 5\hbar\Gamma$ ,  $q^2/C_i = 20\hbar\Gamma$ ,  $q^2/C = 50\hbar\Gamma$ ,  $\varepsilon_u = \varepsilon_d = 0$ .

It is worth noticing that, at high enough drive bias,  $I_{drag}$  is suppressed since the interdot capacitance brings the dot states outside the transport window. Then, the drag current peaks at an optimal value of  $V_1 - V_3$  [10] and vanishes away from it. On the other hand, at very low temperature  $I_{drag}$  is finite only within a voltage range defined by  $\mu_{10} < qV_1 < \mu_{11}$  and  $\min\{qV_3, qV_4\} < \mu_{10}, \mu_{11} < \max\{qV_3, qV_4\}$ . As expected, the drag current increases with *C*, but the voltage window where  $I_{drag}$  is observable becomes narrower. Then, for large coupling the drag circuit effectively induces dynamical channel blockade [23] in the driver and, eventually, the drive current shows electron bunching.

If the drive system is symmetric, the sign of  $I_{\text{drag}}$  depends on the asymmetry factor  $(\Gamma_1 \gamma_2 - \gamma_1 \Gamma_2)$  due to the competition of processes transferring an electron in each direction, independently of the direction of  $I_{\text{drive}}$ . These two contributions have been detected separately in coupled double dot systems in the cotunneling regime giving rise to bidirectional drag [24]. Note that the asymmetry of the drag system can be enough to get a *negative* drag.

We now investigate the nonlinear fluctuation relations for our system. We first analyze the occurrence of  $I_{drag}$  and the current cross correlations  $S_{ii}$  for different conductors (e.g.,  $i = \{1, 2\}$  and  $j = \{3, 4\}$ ). The observation of drag current in one conductor requires the occurrence of correlated tunneling events between the two dots involving the states  $|0\rangle$  and  $|2\rangle$ . These correlated events lead to finite cross correlations. This would not be the case for a model that includes only three charge states. Our minimal model of four charge states does generate correlations between the currents through the two dots. For example, at equilibrium, the fluctuation-dissipation theorem relates the linear drag current to the equilibrium cross correlations for different conductors,  $G_{2,4} = S_{24}^{eq}/2kT$ . Similarly to  $I_{drag}$ , if both conductors are symmetric, i.e.,  $\gamma_1\Gamma_2 = \gamma_2\Gamma_1$  and  $\gamma_3\Gamma_4 =$  $\gamma_4\Gamma_3$ ,  $S_{ii}$  vanishes to first order in a voltage expansion. Figure 2(d) shows that the cross correlation between the drag and drive currents is finite only when there is a drag current flowing in the upper conductor. In general, the sign of the cross correlations is not determined by the direction of the averaged currents [13]. However, in our case, the cross correlations are positive whenever the two currents flow in the same direction, and negative when they are opposite. Interestingly,  $I_{drag}$  can present negative excess noise; i.e., the noise  $S_{22}$  decreases in the presence of drag, as shown in Fig. 2(c).  $S_{22}$  reaches its maximal value when the effective upper dot level is aligned with the Fermi level [25].

Finally, we explicitly check that these fluctuation relations [1,4–6] hold even for our system in which detailed balance is violated. Charge conservation in each subsystem implies  $I_{\alpha} = -I_{\bar{\alpha}}$  and  $S_{\alpha\alpha} = S_{\bar{\alpha}\bar{\alpha}} = -S_{\alpha\bar{\alpha}}$  for two different terminals in the same conductor. Then, from Eq. (3) we derive the nonlinear fluctuation relations involving terminals of the same conductor, and rewrite them as





FIG. 3 (color online). Verification of the nonequilibrium fluctuation relations. Left-hand panels: Nonlinear conductances for (a) Eq. (6), (c),(e) Eq. (7). Right-hand panels: Noise susceptibilities for the corresponding relations in the left. Same parameters as in Fig. 2, except  $\gamma_4 = 0.1\Gamma$ .

$$S_{\alpha\alpha,\alpha} = kTG_{\alpha,\alpha\alpha},$$
  

$$S_{\alpha\alpha,\bar{\alpha}} = -kTG_{\alpha,\bar{\alpha}\,\bar{\alpha}} = kT(2G_{\alpha,\alpha\bar{\alpha}} + G_{\alpha,\alpha\alpha}),$$
(6)

with  $G_{\alpha,\alpha\alpha} + G_{\alpha,\bar{\alpha}\bar{\alpha}} + 2G_{\alpha,\alpha\bar{\alpha}} = 0$ . In Figs. 3(a) and 3(b) we explicitly check that these fluctuation relations hold despite broken detailed balance. The relations including derivatives of current cross correlations at different conductors,  $\alpha$  and  $\beta$ , read

$$2S_{\alpha\beta,\beta} + S_{\beta\beta,\alpha} = kT(G_{\alpha,\beta\beta} + 2G_{\beta,\beta\alpha}),$$
  

$$S_{\alpha\beta,\bar{\alpha}} - S_{\alpha\beta,\alpha} + S_{\bar{\alpha}\alpha,\beta} = kT(G_{\beta,\alpha\bar{\alpha}} + G_{\alpha,\bar{\alpha}\beta} - G_{\alpha,\alpha\beta}),$$
(7)

with  $\sum_{\alpha,\beta} G_{\gamma,\alpha\beta} = 0$ . Equation (7) is verified in Figs. 3(c)– 3(f). It is important to realize here that full access to the fluctuation relations is *only* possible in the presence of drag current [see Figs. 2(a) and 2(c)]. In other words, only when detailed balance is broken and a drag current appears are all fluctuation relations nontrivially verified. In contrast, the absence of drag, i.e.,  $G_{\alpha,\beta\beta} = G_{\alpha,\beta\bar{\beta}} = 0$ , implies  $S_{\alpha\beta,\gamma} = 0$ , for any terminal  $\gamma$ , in which case the fluctuation relations (7) are simply reduced to the relation  $S_{\alpha\alpha,\beta} = 2kTG_{\alpha,\alpha\beta}$ , with  $G_{\alpha,\alpha\beta} = -G_{\alpha,\bar{\alpha}\beta}$ .

*Conclusions.*—In summary, we have proposed a geometry of two conductors put in proximity interacting via longrange Coulomb forces to test fluctuation relations in the nonlinear transport regime. This system exhibits a drag current as a direct consequence of the absence of detailed balance. Our main findings are (i) the general expression for the current-voltage characteristic of two interacting conductors and (ii) the verification of the fluctuation relations in a nonequilibrium system when detailed balance is broken. Our proposal motivates new experiments to test the fluctuation relations away from equilibrium when detailed balance does not hold. The influence of coherent transport is a challenging problem that we hope will be encouraged by our work.

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