

Fluctuation Relations Without Microreversibility in Nonlinear Transport

H. Förster and M. Büttiker

Département de Physique Théorique, Université de Genève, CH-1211 Genève 4, Switzerland

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In linear transport, the fluctuation-dissipation theorem relates equilibrium current correlations to the linear conductance coefficient. For nonlinear transport, there exist fluctuation relations that rely on Onsager's principle of microscopic reversibility away from equilibrium. However, both theory and experiments have shown deviations from microreversibility in the form of magnetic field asymmetric current-voltage relations. We present novel fluctuation relations for nonlinear transport in the presence of magnetic fields that relate current correlation functions at any order at equilibrium to response coefficients of current cumulants of lower order. We illustrate our results with the example of an electrical Mach-Zehnder interferometer.

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Introduction.—Onsager derived the symmetry of transport coefficients of irreversible processes using the principle of microscopic reversibility for the fluctuations of the equilibrium system [1,2]. Thus the symmetry of transport coefficients in the linear transport regime is directly related to the fluctuation-dissipation theorem of Einstein, Johnson, Nyquist, and Kubo [3–6]. Naturally, the question arises whether there are fluctuation relations beyond the linear response regime. In statistical mechanics, fluctuation relations were derived [7,8] as an extension of Onsager's relation to systems far from equilibrium. These fluctuation relations make statements on the distribution function of observables conjugate to thermodynamic forces for a wide variety of nonequilibrium systems [9,10]. In electrical transport the variable of interest is the transferred charge. The theory is known as full counting statistics [11,12], and fluctuation relations for conductors have been discussed in the absence [12–15] and the presence of a magnetic field [16].

At equilibrium, in the presence of a magnetic field, Onsager reciprocity still holds. However, away from equilibrium, the potential landscape inside the conductor is neither an odd nor an even function of magnetic field. As a consequence, electrical conductors exhibit manifest deviations from symmetries based on microreversibility and fluctuation relations derived from this principle [16] are not valid. Surprisingly, and this is a central point of our work, we obtain novel fluctuation relations even without invoking the principle of microreversibility. Importantly, the novel fluctuation relations are general and independent of a specific model for interactions.

Full counting statistics and fluctuation theorem.—The full counting statistics of a conductor with M terminals is the probability distribution $P(\mathbf{Q})$ that $\mathbf{Q} = (Q_1, Q_2, \dots, Q_M)$ charges are transmitted into the reservoirs during the measurement time t . The distribution function $P(\mathbf{Q})$ is expressed by the generating function $F(i\mathbf{\Lambda}) = \ln \sum_{\mathbf{Q}} P(\mathbf{Q}) e^{i\mathbf{\Lambda}\mathbf{Q}}$, where $\mathbf{\Lambda} = (\lambda_1, \lambda_2, \dots, \lambda_M)$ are called counting fields. In the long time limit, all irreducible

current cumulants at zero frequency are obtained by consecutive derivatives of the generating function, in contact α this is $\langle (\Delta I_\alpha)^k \rangle = (-ie)^k [\partial^k F / \partial \lambda_\alpha^k]_{\mathbf{\Lambda}=\mathbf{0}} / t$. The magnetic field B perpendicular to the conductor and the affinities $\mathbf{A} = (eV_1/k_B T, eV_2/k_B T, \dots, eV_M/k_B T)$ are externally controlled. Here eV_α is the potential at terminal α and T the temperature, assumed to be equal and nonzero in all terminals.

The fluctuation relation for the full counting statistics gives a simple relation for the probability that \mathbf{Q} or—at reversed magnetic field— $-\mathbf{Q}$ charges are transmitted. Derivations [12–16] rely fundamentally on microscopic reversibility: a process from terminal α to β has the same probability as the reversed process, from terminal β to α at inversed magnetic field. References [12–16] assume that this is valid also far from equilibrium and find

$$P(\mathbf{Q}, B) = e^{\mathbf{A}\mathbf{Q}} P(-\mathbf{Q}, -B), \quad (1)$$

$$F_\pm(i\mathbf{\Lambda}, \mathbf{A}) = \pm F_\pm(-i\mathbf{\Lambda} - \mathbf{A}, \mathbf{A}). \quad (2)$$

Equation (2) is the Fourier transform of Eq. (1) and deter-

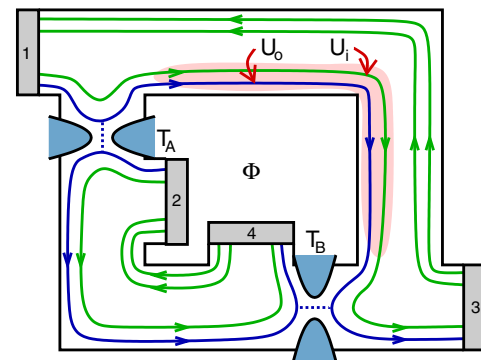


FIG. 1 (color online). Mach-Zehnder interferometer at filling factor 2. Only the outer edge state enters the interferometer. Coulomb interactions between neighboring edges—as indicated by shading—lead to internal potentials U_i and U_o in the inner and outer edge. For reversed magnetic field, all arrows point in the opposite direction.

mines the symmetry of the generating function. For convenience, the (anti)symmetrized generating function $F_{\pm}(i\Lambda) = F(i\Lambda, B) \pm F(i\Lambda, -B)$ is used, and the notation $F_{\pm}(i\Lambda, \mathbf{A})$ emphasizes that the generating function depends explicitly on the affinities \mathbf{A} .

Mach-Zehnder interferometer (MZI).—As an instructive example, we present a MZI (see Fig. 1) and show how interaction (screening) effects lead to deviations from reversibility. It is a four-terminal conductor with two quantum point contacts acting as beam splitters as shown in Fig. 1. The two interferometer arms enclose a magnetic flux Φ , such that interference arises due to the Aharonov-Bohm effect. In experiments [17–20], the MZI is realized using edge states in the quantum Hall regime, and it is often operated at filling factor 2. Only carriers in the outer edge enter the interferometer and are able to interfere. Here, the inner edge state moves in vicinity in both interferometer arms and carries current from terminal 1 to 3 and from 2 to 4 [20]. Although a four-terminal conductor, the MZI is characterized by only a single transmission probability T_{31} , due to the separation of left and right movers. T_{31} is the probability for a particle to be transmitted in the outer edge state from terminal 1 to 3. In the linear transport regime, reciprocity means that [21] $T_{31}(+B) = T_{13}(-B)$. We next demonstrate that already Hartree interactions lead to a violation of Eqs. (1) and (2).

Breakdown.—Interactions can lead to magnetic field asymmetry in nonlinear transport, as was shown theoretically [22–24] as well as experimentally [25–29]: Every particle is moving in a local potential $U(\vec{r})$ generated by all the other particles. The internal potential has to be determined self-consistently and depends on all potentials V_{γ} applied in the external contacts, $U(\vec{r}) = U(\vec{r}, \{V_{\gamma}\})$. The scattering matrix depends on the energy of the particle and is a functional of the internal potential, $\mathcal{S} = \mathcal{S}(E, U(\vec{r}))$. Indeed the functional dependence of the scattering matrix is required by gauge invariance: The generating function has to be invariant under a shift of all potentials by the same amount, $V_{\gamma} \rightarrow V_{\gamma} + U_0$. This condition can be expressed as $\frac{dF}{dU_0} = 0$. For long times and neglecting interactions, the generating function in the scattering approach is [11]

$$F(i\Lambda) = \frac{i}{h} \int dE \operatorname{tr}[\ln(\mathbf{1} - \tilde{f}K)]. \quad (3)$$

Here, $K = (\mathbf{1} - \tilde{\lambda}^{\dagger} \mathcal{S}^{\dagger} \tilde{\lambda} \mathcal{S})$ is composed of the scattering matrix \mathcal{S} , the unit matrix $\mathbf{1}$, and the matrix $\tilde{\lambda}$ introducing the counting fields, $\tilde{\lambda} = \operatorname{diag}(e^{-i\lambda_1}, e^{-i\lambda_2}, \dots, e^{-i\lambda_M})$. The diagonal matrix \tilde{f} contains the Fermi functions of the different terminals with $\tilde{f} = \operatorname{diag}(f_1, f_2, \dots, f_M)$. With this we can show that gauge invariance requires

$$\sum_{\gamma} \frac{\partial K}{\partial V_{\gamma}} + e \frac{\partial K}{\partial E} = 0. \quad (4)$$

Here we used that the derivative of the Fermi functions with respect to U_0 can be expressed as an energy derivative

and that $\partial K / \partial U_0 = \sum_{\gamma} \partial K / \partial V_{\gamma}$. Therefore, the scattering matrix depends also via the internal potential landscape on the external voltages, $\mathcal{S} = \mathcal{S}(E, \{V_{\gamma}\})$. As mentioned above, the local internal potential has to be determined self-consistently and it is not necessarily an even function of magnetic field [22]. As a consequence, for nonlinear transport, the scattering matrix is not reversible, $\mathcal{S}_{\alpha\beta}(B, \{V_{\gamma}\}) \neq \mathcal{S}_{\beta\alpha}(-B, \{V_{\gamma}\})$. This implies immediately the breakdown of the fluctuation theorem (1) and (2) for nonlinear transport, since any derivation is based upon reciprocity.

The lack of reversibility can be shown explicitly for the MZI. Coulomb interactions between the two edge states moving through the interferometer lead to internal potentials U_o, U_i in the outer and inner edge. In this respect the inner edge acts as a gate on the outer edge. For the interference, this gives rise to an additional phase difference $\varphi(B) = e\Delta U_o \tau / h$ between the two interferometer arms. Here, τ is the time an electron needs to traverse the interferometer, and ΔU_o is the difference of the internal potential in the outer edge between the two arms.

It is easy to see that the internal screening potential U_o is not an even function of magnetic field: For positive magnetic field as shown in Fig. 1, only processes from left to right contribute, and the potential will depend on the reflection $R_A = 1 - T_A$ of the left beam splitter and on the voltages V_1 and V_2 . For inverted magnetic field, processes from right to left are important, which depend on $R_B = 1 - T_B$ and voltages V_3 and V_4 . To be explicit, we determine the potential self-consistently within a Hartree approximation [22,24,30]. The average charges q_o and q_i in the edges of the upper interferometer arm are, on the one hand, expressed as the difference between injected and screened charge, and are, on the other hand, determined by Coulomb interaction. For positive magnetic field, this determines U_o and U_i in the upper arm through

$$q_i = e^2 D(V_1 - U_i) = C(U_i - U_o), \quad (5)$$

$$q_o = e^2 D(R_A V_1 + T_A V_2 - U_o) = C(U_o - U_i). \quad (6)$$

Here, C is the geometric capacitance between the two edges, and D is the density of states of an edge state. Similar equations hold for the lower interferometer arm and for reversed magnetic field. To first order in external voltage, the potential difference $\Delta U_o = \sum_{\alpha} u_{\alpha} V_{\alpha}$ is determined by the characteristic potentials $u_{\alpha} = [\partial \Delta U_o / \partial V_{\alpha}]_{\text{eq}}$. We find $u_3(B) = u_1(-B) = 0$, $u_1(B) = -u_2(B) = R_A - e^2 D T_A / (2C + e^2 D)$, and $u_3(-B) = -u_4(-B) = R_B - e^2 D T_B / (2C + e^2 D)$.

Using the characteristic potentials, the self-consistent transmission probability $T_{31} = T_{31}(+B, V_1 - V_2)$ for a particle in the interfering edge to transmit from terminal 1 to 3 for positive magnetic field is $T_{31} = R_A R_B + T_A T_B - 2\sqrt{R_A R_B T_A T_B} \cos(\Phi - \varphi)$ with $\varphi(+B) = e u_1(+B) \times \tau(V_1 - V_2) / h$. For $T_{13} = T_{13}(-B, V_3 - V_4)$ at negative magnetic field, the additional phase is $\varphi(-B) = e u_3(-B) \tau(V_3 - V_4) / h$. The lack of reversibility out of

equilibrium is evident:

$$T_{31}(+B, V_1 - V_2) \neq T_{13}(-B, V_3 - V_4). \quad (7)$$

This means that the fluctuation relation (2) is, strictly speaking, valid only at equilibrium but has corrections for finite voltages. In general, taking into account interactions beyond the Hartree-level will not reestablish reversibility.

Fluctuation relations for correlation functions.— Including interactions, the fluctuation relation (2) for the counting statistics is not valid anymore, as shown above explicitly within a Hartree model. Nevertheless, we can derive fluctuation relations for current correlation functions. We emphasize that the following section is general, no specific model for interactions is needed. It is useful to expand the first few cumulants for $eV \ll k_B T$,

$$I_\alpha = \sum_\beta G_{\alpha,\beta}^{(1)} V_\beta + \sum_{\beta\gamma} G_{\alpha,\beta\gamma}^{(2)} \frac{V_\beta V_\gamma}{2} + \mathcal{O}(V^3), \quad (8)$$

$$S_{\alpha\beta} = S_{\alpha\beta}^{(0)} + \sum_\gamma S_{\alpha\beta,\gamma}^{(1)} V_\gamma + \mathcal{O}(V^2), \quad (9)$$

$$C_{\alpha\beta\gamma} = C_{\alpha\beta\gamma}^{(0)} + \mathcal{O}(V). \quad (10)$$

Up to second order in voltage, the mean current I_α in terminal α is determined by the linear and nonlinear conductance coefficients, $G_{\alpha,\beta}^{(1)}$ and $G_{\alpha,\beta\gamma}^{(2)}$. The zero-frequency current correlations $S_{\alpha\beta} = \langle \Delta I_\alpha \Delta I_\beta \rangle$ contain equilibrium Nyquist noise $S_{\alpha\beta}^{(0)}$ and the noise susceptibility [31] $S_{\alpha\beta,\gamma}^{(1)}$ which includes the emergent shot noise. Of the third cumulant $C_{\alpha\beta\gamma} = \langle \Delta I_\alpha \Delta I_\beta \Delta I_\gamma \rangle$, only the equilibrium value $C_{\alpha\beta\gamma}^{(0)}$ is used in the following. All response coefficients are obtained from the generating function, e.g., $G_{\alpha,\beta\gamma}^{(2)} = -ie[\partial^3 F / \partial \lambda_\alpha \partial V_\beta \partial V_\gamma]_0 / t$, where the index 0 means setting Λ and \mathbf{A} to zero.

(Anti)symmetrizing the above definitions, both the fluctuation-dissipation theorem (for +) and the Onsager-Casimir relations (for -) can be formulated concisely as

$$S_{\alpha\beta\pm}^{(0)} = k_B T (G_{\alpha,\beta\pm}^{(1)} + G_{\beta,\alpha\pm}^{(1)}) = \pm S_{\alpha\beta\pm}^{(0)}. \quad (11)$$

The next order fluctuation relation connects the third cumulant at equilibrium which is odd in magnetic field with combinations of the noise susceptibility and nonlinear conductance coefficients,

$$\begin{aligned} C_{\alpha\beta\gamma,\pm}^{(0)} &= k_B T [S_{\alpha\beta,\gamma\pm}^{(1)} + S_{\alpha\gamma,\beta\pm}^{(1)} + S_{\beta\gamma,\alpha\pm}^{(1)} \\ &\quad - k_B T (G_{\alpha,\beta\gamma\pm}^{(2)} + G_{\beta,\alpha\gamma\pm}^{(2)} + G_{\gamma,\alpha\beta\pm}^{(2)})] \\ &= \mp C_{\alpha\beta\gamma,\pm}^{(0)}. \end{aligned} \quad (12)$$

These universal fluctuation relations can be extended to any order: A current correlation function at equilibrium is expressed by combinations of successive response coefficients of lower order current cumulants. They are graphically represented in Fig. 2. The first two lines of the figure

correspond to Eqs. (11) and (12), higher order relations can easily be constructed.

The derivation of the fluctuation relations is based on the following properties of the generating function:

$$F_\pm(-\mathbf{A}, \mathbf{A}) = F_\pm(\mathbf{0}, \mathbf{A}) = 0, \quad (13)$$

$$F_\pm(i\Lambda, \mathbf{0}) = \pm F_\pm(-i\Lambda, \mathbf{0}). \quad (14)$$

The first equation defines a special symmetry point at $i\Lambda = -\mathbf{A}$ (for all A) for which the generating function vanishes, just as for $\Lambda = \mathbf{0}$ which originates from probability conservation. To demonstrate it for a system with arbitrary electron-electron interactions, we start from the definition of the generating function $F(i\Lambda) = \ln \langle e^{-i\Lambda \hat{Q}_t} e^{i\Lambda \hat{Q}_0} \rangle_0$. Here, \hat{Q}_0 and \hat{Q}_t denote the charge operators at time 0 and time t , and the expectation value is taken with respect to the initial state, described by a grand-canonical density matrix. At time 0 the conductor is decoupled from the reservoirs, and the initial Hamiltonian \hat{H}_0 commutes with the charge \hat{Q}_0 . To derive Eq. (13), we use that the total energy in the system “conductor + reservoirs” is conserved at all times. We emphasize that the identity Eq. (13) is valid without micro-reversibility. Special care should be taken of the case when (i) the problem is time-dependent, (ii) the temperature is not equal in all reservoirs, and (iii) a bath allows energy exchange, e.g., via electron-phonon interactions. Then, we would have to consider energy currents as well and introduce additional counting fields that account for the transferred energy. In this case, a similar relation can be derived. In terms of distribution functions, Eq. (13) defines a global detailed balance relation,

$$\sum_{\mathbf{Q}} P(\mathbf{Q}) = \sum_{\mathbf{Q}} P(\mathbf{Q}) e^{-\mathbf{A}\mathbf{Q}} = \langle e^{-\mathbf{A}\mathbf{Q}} \rangle = 1, \quad (15)$$

valid even if Eq. (1) is not true. The second equation, Eq. (14), represents the fluctuation relation (2) at $\mathbf{A} = \mathbf{0}$ and is a consequence of microscopic reversibility at equi-

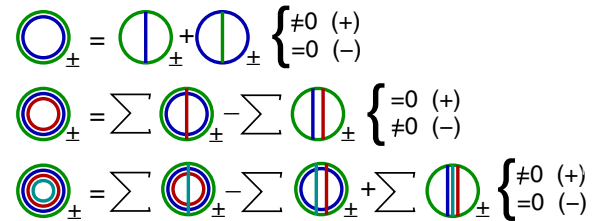


FIG. 2 (color online). A graphical representation of the relations between response coefficients, generalizing Eq. (17) to multiple terminals. The number of circles stands for the order of the cumulant, and the number of vertical lines means the order of the derivative with respect to voltage as well as the power of the factor ($k_B T$) with which the response coefficient has to be multiplied. The different heavy lines (different colors) represent derivatives with respect to quantities of different terminals. The summations go over all possible permutations $\binom{k}{l}$, with k the number of circles and l the number of vertical lines. Higher order correlations follow the same rules.

librium. It follows that even (odd) cumulants at equilibrium are even (odd) in magnetic field as expressed by

$$\langle(\Delta I_\alpha)^k\rangle_\pm^{\text{eq}} = \pm(-1)^k\langle(\Delta I_\alpha)^k\rangle_\pm^{\text{eq}}. \quad (16)$$

Both functions $F_\pm(i\mathbf{\Lambda}, \mathbf{A})$ and $F_\pm(-i\mathbf{\Lambda} - \mathbf{A}, \mathbf{A})$ can be expanded as Taylor series around $\mathbf{A} = \mathbf{0}$ and $\mathbf{\Lambda} = \mathbf{0}$. This defines general relations for specific Taylor coefficients,

$$\left. \frac{\partial^k F_\pm(-i\mathbf{\Lambda} - \mathbf{A}, \mathbf{A})}{\partial A_\alpha^k} \right|_0 = \sum_{l=0}^k \binom{k}{l} i^l \left. \frac{\partial^k F_\pm(i\mathbf{\Lambda}, \mathbf{A})}{\partial A_\alpha^{k-l} \partial \lambda_\alpha^l} \right|_0,$$

which vanish identically due to Eq. (13). The last term in the sum represents the k th derivative of the generating function with respect to the counting fields, which is the k th cumulant at equilibrium. Solving the above equation for this last term leads to

$$\langle(\Delta I_\alpha)^k\rangle_\pm^{\text{eq}} = - \sum_{l=1}^{k-1} \binom{k}{l} (-k_B T)^{k-l} \left. \frac{\partial^{k-l} \langle(\Delta I_\alpha)^l\rangle}{\partial V_\alpha^{k-l}} \right|_\pm^{\text{eq}}. \quad (17)$$

This equation relates a correlation function at equilibrium to a linear combination of response coefficients of lower order correlations. Together with Eq. (16)—which determines the magnetic field symmetry—it defines new fluctuation relations for nonlinear transport. In Fig. 2, they are schematically represented and extended to the general case of a multiterminal conductor.

For the MZI, the fluctuation relation Eq. (12) can be explicitly verified within Hartree. We are concerned with temperatures and voltages low compared to the first plasma mode of an interferometer arm. Because of the separation of left and right movers, several response coefficients vanish, in particular, the nonlinear conductance $G_{3,31\pm}^{(2)}$ as well as the noise susceptibility $S_{33,1\pm}^{(1)}$. Also the third cumulant at equilibrium $C_{331\pm}^{(0)}$ is zero, because the scattering matrix is energy-independent for equal length of the interferometer arms. But the coefficients $G_{1,33}^{(2)}$ and $S_{31,3}^{(1)}$ are finite for $-B$ due to the internal potential, and vanish for B . Similar arguments hold for response coefficients with $1 \leftrightarrow 3$. Using $dg/dU \equiv (4e^3 \tau / h^2) \sqrt{R_B T_B R_A T_A} \sin \Phi$, Eq. (12) simplifies for the MZI to

$$2S_{31,3\pm}^{(1)} = k_B T G_{1,33\pm}^{(2)} = \pm k_B T u_3(-B) dg/dU, \quad (18)$$

$$2S_{31,1\pm}^{(1)} = k_B T G_{3,11\pm}^{(2)} = k_B T u_1(B) dg/dU. \quad (19)$$

The fluctuation relation (2), which does not account for magnetic field asymmetry in screening effects, would require that the antisymmetrized part $(-)$ of the above equations is identically zero [16]. Measuring a nonlinear conductance coefficient $G_{1,33}^{(2)}$ or noise susceptibility $S_{31,3}^{(1)}$, which is asymmetric in magnetic field, proves Eq. (2) wrong. The fluctuation relations Eqs. (18) and (19) are linear in temperature, periodic with the magnetic flux Φ , and depend on the reflection of the beam splitters; they can be experimentally verified.

Conclusion.—We have shown that electron-electron interactions lead to a breakdown of the usual fluctuation relations for the full counting statistics in the presence of a magnetic field. The reason is that interactions can induce effective deviations from reversibility of scattering processes out of equilibrium. Instead, fluctuation relations can be derived which relate correlation functions at equilibrium to response coefficients of correlations of lower order. These fluctuation relations are valid even in the presence of magnetic field asymmetry.

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