Fluctuation relations for spin currents

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(Dated: February 26, 2015)

The fluctuation theorem establishes general relations between transport coefficients and fluctuations in nonequilibrium systems. Recently there was much interest in quantum fluctuation relations for electric currents. Since charge carriers also carry spin, it is important to extend the fluctuation theorem to spin currents. We use the principle of microscopic reversibility to derive such theorem. As a consequence, we obtain a family of relations between transport coefficients and fluctuations of spin currents. These relations generalize the fluctuation-dissipation theorem to nonlinear transport. They do not depend on the microscopic model and can be used to test the validity of theoretical approximations in spin-transport problems.

PACS numbers: 72.25.-b, 05.40.-a, 72.10.Bg, 85.75.-d

I. INTRODUCTION

Both charge and spin of electron play major roles in solid state physics and its applications but the electric and spin currents behave in different ways. Coulomb interaction brings a high energy cost to any violation of charge neutrality. This fact and charge conservation make it easy to create and manipulate charge currents. At the same time, spin does not conserve. Its coupling to external probes is weaker than that of the electric charge. Thus, manipulating spin currents is a harder task than working with electric currents. Recently, much effort has focused on overcoming this challenge in the hope to get more control over the spin degree of freedom\textsuperscript{1}. That effort has been motivated by both potential applications and the interest in basic spin physics. An important open problem in the field is the understanding of nonlinear spin transport.

The problem is challenging because nonlinear transport occurs away from thermal equilibrium. Until recently, any progress in the theory of nonequilibrium transport was inevitably based on microscopic models\textsuperscript{2}. The situation changed after the discovery of fluctuation theorems\textsuperscript{3-5} which brought a powerful general principle to nonequilibrium statistical mechanics. The goal of this paper consists in the derivation of a family of fluctuation relations for spin currents and noises. Such relations are completely general and do not depend on microscopic details or the choice of a model. Thus, they give a useful tool for testing theoretical approximations.

Fluctuation theorems generalize the principle of detailed balance. Detailed balance establishes a fundamental relation between the rates of different processes. Fluctuation theorems go a step further and relate the probabilities of a process and its time-reversed version on a macroscopic time scale. At first sight, fluctuation relations apply to time-reversal invariant systems only. Indeed, a process and its time-reversed version can occur in the same system only if the time-reversal symmetry is present, i.e., in the absence of a spontaneous magnetization and external magnetic fields. Otherwise, the time-reversed process would require changing the sign of the magnetic field. However, interesting relations do not have to connect the properties of a single system. In particular, a number of fluctuation relations for electric currents in a magnetic field were derived in Ref. 6. Those relations contain currents and noises in two systems that differ by their directions of the magnetic fields. Moreover, in some cases it even proved possible to derive fluctuation relations that connect currents and noises in a single system in the absence of the time-reversal symmetry\textsuperscript{7,8}. A review of those developments can be found in Ref. 8.

Focusing on the spin degree of freedom means treating currents as multi-component with the components, corresponding to the up and down spin projections. A fluctuation theorem for multi-component currents was addressed in Ref. 9. That theorem is not directly relevant in our problem since Ref. 9 considers the currents of time-reversal-invariant quantities while spin changes its sign under time reversal. More specifically, the spin operator $S$ satisfies

$$
\Theta S \Theta^{-1} = -S,
$$

where $\Theta$ is the time reversal operator. In contrast, charge is invariant under time reversal: $\Theta Q \Theta^{-1} = Q$. Some fluctuation relations for spin currents were derived in Ref. 11. In this paper we establish a bigger family of relations which could not be obtained with the approach of Ref. 11. Indeed, Ref. 11 did not make use of microreversibility that allows us to go beyond the relations\textsuperscript{11}.

According to the principle of microscopic reversibility, the amplitudes of a process in a given system and the time-reversed process in the system with the reversed directions of all magnetic fields must be complex conjugate. On the basis of calculations within the Landauer-Büttiker formalism\textsuperscript{12,13}, it was proposed that microreversibility fails in nonzero magnetic fields. We believe that the conflict of the Landauer-Büttiker approximation and the principle of microscopic reversibility shows only the limitations of the Landauer-Büttiker approach.
Note that the conclusions, based on the microreversibility principle are supported by the experiment\textsuperscript{14,15} and agree with theoretical calculations beyond the Landauer-Büttiker approximation\textsuperscript{16,17}. A detailed discussion of microreversibility in the presence of a magnetic field can be found in the review\textsuperscript{14}. Clearly, it is important to address the consequences of microreversibility for spin currents and noises.

This is what we do below. We set the problem and derive a general fluctuation theorem for spin transport in Section II. In Section III we deduce from the theorem a family of fluctuation relations for spin currents and noises. We compare our results with Refs. 10,11 in Section IV.

II. FLUCTUATION THEOREM

We will study fluctuation theorems that are concerned with the transport through a finite system (conductor) attached to two or more large reservoirs. Each reservoir is assumed to be in thermal equilibrium but the reservoirs are not necessarily in equilibrium with each other. A difference between the reservoirs’ chemical potentials drives a current through the conductor. Our goal is to establish various relations between the correlation and response functions of the currents. The crucial assumption is that the system does not have long-term memory beyond a characteristic transport time through the conductor. Thus, we can assume any convenient choice of the system’s past. The standard choice is as follows. Initially, the reservoirs and the conductor are disconnected. Next, the particle and energy exchange between the system and reservoirs is turned on for some long time $\tau$. At the end, the reservoirs are again disconnected from the conductor. We are interested in the transport during the time interval $\tau$. Yet, it is most convenient to describe such transport in terms of the quantities, measured before the interaction between the conductor and the reservoirs was turned on at $t = 0$ and after the interaction was turned off at $t = \tau$. For example, the average current from reservoir number $k$ is defined as $- [Q_k(\tau) - Q_k(0)]/\tau$, where $Q_k(t)$ is the total charge of reservoir $k$ at time $t$. The energy and spin currents are defined in a similar way. Thus, the following measurement protocol is assumed: we measure the charge, the $z$-projection of the spin and the energy of each reservoir before and after the evolution period $\tau$.

Such simultaneous measurements are only possible, if the charge and the $z$-component of the spin commute with the energy operator and hence conserve. This poses an obvious problem: spin does not conserve because of the spin-orbit interaction. In addition, external magnetic fields may lead to the Larmor precession which breaks spin conservation. Moreover, if the spin-relaxation time is short it may be meaningless to even speak about spin currents, flowing from the reservoirs. Indeed, the spin current is defined as the total spin flowing per unit time through a cross-section near the boundary of the reservoir and conductor. In the absence of spin conservation such current depends on the choice of the cross-section.

The above discussion necessitates the following assumptions. First, to avoid the problem with the Larmor precession, we assume that the magnetic fields are oriented along the $z$-axis in the reservoirs. Such magnetic fields would break the conservation of the $x$- and $y$-components of the reservoir spin but we are only interested in the $z$-component. Second, we assume a weak spin-orbit interaction in the reservoirs. The interaction must be weak enough that it can be neglected on the time scale of the transport through the conductor and on the much longer time scale during which a quasiequilibrium establishes in a large portion of each reservoir near the interface with the conductor. Such quasiequilibrium can be described by two different chemical potentials in each reservoir for the up and down projections of the electron spin on the $z$-axis. In the simplest situation the two chemical potentials are equal but we will consider the most general case of arbitrary chemical potentials. We will assume that the quasiequilibrium survives during the evolution period $\tau$. Note that very long spin-relaxation times were reported for donor spins in pure silicon\textsuperscript{18}.

The reservoir spin is gradually lost due to the spin-orbit interaction and the spin current into the conductor. Hence, the two chemical potentials slowly depend on time. To keep them constant, one would need to compensate the spin loss by a spin current injection into the reservoir.

We do not make any assumptions about the spin-orbit interaction and the direction of the magnetic field in the conductor. If the $z$-projection of the spin does not conserve in the conductor then the spin currents, leaving the reservoirs, do not add up to zero.

We are now ready to derive the fluctuation theorem. Our method is connected with the approach of Refs. 19,20. We assume that initially all reservoirs are disconnected. It will be convenient to include the conductor as a part of one of the reservoirs. This requires us to make the Hamiltonian of the conductor time-dependent so that the spin conserves in the conductor at $t < 0$ and $t > \tau$. We will assume that the time-dependence is slow (i.e., $\tau$ is large) and hence the energy conserves. We will use the units such that $\hbar = k_B = 1$.

The initial density matrix of the system is the product of the density matrices of the reservoirs:

$$
\rho_F = \prod_i \rho_i = \prod_i \frac{1}{Z_i} \exp \left[ \frac{V_i Q_i + V_{Si} S_{zi} - E_i}{T} \right],
$$

where

$$
Z_i = \text{Tr} \exp \left[ \frac{V_i Q_i + V_{Si} S_{zi} - E_i}{T} \right],
$$

$Q_i$ is the charge of reservoir $i$, $E_i$ is its energy, $S_{zi}$ is the $z$-component of the spin in reservoir $i$, $V_i$ is the elec-
trochemical potential and \( V_S \) is the chemical potential, conjugate to \( S_z \). The chemical potentials for the up and down spins are \((-e)V_i \pm V_S/2\) and equal \(-eV_i\) at \( V_S = 0\), where \(-e\) is the electron charge. Eq. (2) assumes that all reservoirs have the same temperature \( T \). A generalization of our calculations to the case of different temperatures is straightforward.

The evolution operator is

\[
U_F(t) = T \exp \left( -i \int_0^t H(t') dt' \right) \tag{4}
\]

where \( H(t) = \sum H_i(t) \) is the time-dependent Hamiltonian, and \( T \) is the time ordering operator.

We can now write the probability of the forward process. This is the probability that during the evolution period \( \tau \) the changes of the reservoirs change by \( \Delta Q_i \) and their spins change by \( \Delta S_{zi} \):

\[
P_F[\Delta Q, \Delta S_z] = \sum [(Q(0), S_z(0), n(0)] \rho_F[Q(0), S_z(0), n(0)]
\]

\[
\times \left| \langle (Q(\tau), S_z(\tau), n(\tau)|U_F(\tau)|Q(0), S_z(0), n(0)] \right|^2
\]

\[
\times \delta(\Delta Q - (Q(\tau) - Q(0))) \delta(\Delta S_z - (S_z(\tau) - S_z(0)))
\]

where \( \mathbf{A} \) stays for a vector whose component \( A_i \) describes reservoir \( i \). The summation in Eq. (5) extends over all initial and final states which we label with the charge vector \( Q \), the spin vector \( S_z \), and an additional quantum number \( n \) that distinguishes quantum states with identical charges and spins in the reservoirs.

We now introduce the time-reversed backward process. The initial density matrix

\[
\rho_B = \Theta \rho_F \Theta^{-1} = \prod \frac{1}{Z_i} \exp \left[ \frac{V_i Q_i - V_S S_{zi} - E_i}{T} \right]
\]

where \( \Theta \) is the antimunitary time-reversal operator, satisfying \( \Theta i = -i \Theta \), and we use the relation \( \Theta S_{zi} \Theta^{-1} = -S_{zi} \). The dynamics is controlled by the Hamiltonian

\[
H_B = \Theta H(\tau - t) \Theta^{-1}
\]

The evolution operator

\[
U_B(t) = T \exp \left( -i \int_0^t H_B(t') dt' \right)
\]

\[
= \Theta \left[ T^{-1} \exp (+i \int_{\tau-t}^\tau H(t') dt') \right] \Theta^{-1},
\]

where \( T^{-1} \) is the reversed time ordering operator. Hence,

\[
U_B(\tau) = \Theta U_F^d(\tau) \Theta^{-1}
\]

We compute the probability that during the evolution period \( \tau \) the changes of the reservoir charges are given by the vector \(-\Delta Q\) and the reservoir spins change by \(+\Delta S_z\):

\[
P_B[-\Delta Q, \Delta S_z] = \sum [(Q(\tau), S_z(\tau), n(\tau)] \rho_B[Q(\tau), S_z(\tau), n(\tau)]
\]

\[
\times \left| \langle (Q(0), S_z(0), n(0)|U_B(\tau)|Q(\tau), S_z(\tau), n(\tau)] \right|^2
\]

\[
\times \delta(-\Delta Q - (Q(0) - Q(\tau))) \delta(-\Delta S_z - (S_z(0) - S_z(\tau)))
\]

The above formula differs from Eq. (4) in that \( \tau \) stays in place of 0 and vice versa inside the bra and ket vectors. This change of notation emphasizes the time-reversed dynamics. For the same reason, all matrix elements are written in the basis of the time-reversed states \( \Theta Q, S_z, n \). Next, we combine Eqs. (5), (6), (8), and (9) and use the conservation of the energy \( \sum E_i \) on the evolution period \( \tau \). We find

\[
P_F(\Delta Q, \Delta S_z) = P_B(-\Delta Q, \Delta S_z) \exp \left[ -\frac{\sum(V_i \Delta Q_i + V_S \Delta S_{zi})}{T} \right].
\]

Finally, we note that \( P_B \) can be interpreted as a probability of a forward process in the system with the same chemical potentials \( V \) and the opposite chemical potentials \( V_S \) in the opposite magnetic field. Here, the fact that \( V_S \) changes its sign follows from the comparison of Eqs. (2) and (9). Strictly speaking, in addition to the sign changes of the magnetic field and \( V_S \), one also needs to demand the opposite sign of the reservoir magnetizations in a system with spontaneously broken time-reversal symmetry. In what follows we instead assume that the time-reversal symmetry is always broken explicitly by some (possibly infinitesimal) magnetic field. Interpreting Eq. (11) as a relation between the probabilities of the two forward processes and getting rid of the indices \( B \) and \( F \), one obtains the fluctuation theorem

\[
P(\Delta Q, \Delta S_z, V, V_S, B) = P(-\Delta Q, \Delta S_z, -V, -V_S, -B) \exp \left[ -\frac{\sum(V_i \Delta Q_i + V_S \Delta S_{zi})}{T} \right]
\]

where \( \mathbf{B} \) is the coordinate-dependent magnetic field.

It what follows it will be convenient to slightly modify the notations. We will redefine \( V_i \to V_i - \mu \), where \( \mu \) is the common electrochemical potential of all reservoirs in equilibrium. This will not change the form of Eq. (12) but will allow us to assume that \( V_i \) are small in the limit of the low voltage bias. On the other hand, \( V_S \) are already small in the limit of the low voltage bias since spin is not conserved far from the conductor and hence its chemical potential is zero in a true thermal equilibrium.

## III. FLUCTUATION RELATIONS FOR TRANSPORT COEFFICIENTS

We now consider transport quantities. We first intro-
Duce a piece of notation:

\[
|{\Delta Q^m_{\alpha S}^{\nu}}|_{V,V_S} = \left( \prod_i \Delta Q_{i \alpha}^{n_{i}} \right)_{V,V_S} \sum_{\Delta Q,\Delta S_S} P(\Delta Q, \Delta S_S, V,V_S, B) \prod_i \Delta Q_{i \alpha}^{n_{i}} \Delta S_{S i}^{n_{i}}. \tag{13}
\]

With this notation, the average charge and spin currents, injected into the conductor from reservoir \( i \), can now be compactly written as

\[
I_i = -\langle \Delta Q_i \rangle / \tau; \tag{14}
\]

\[
I_{Si} = -\langle \Delta S_{Si} \rangle / \tau. \tag{15}
\]

We will also be interested in various cumulants of the currents, for example, the noise of electric currents

\[
S_{ij} = 2 \int dt dt' (I_i(t) - I_i)(I_j(t') - I_j) / \tau = 2 \int dt dt' \left[ I_i(t) I_j(t') - \bar{I}_i \bar{I}_j \right] / \tau, \tag{16}
\]

the noise of spin currents and the cross noises

\[
S_{ij}^S = 2 \int dt dt' (I_{Si}(t) I_{Sj}(t') - \bar{I}_{Si} \bar{I}_{Sj}) / \tau, \tag{17}
\]

\[
S_{ij}^{\text{cross}} = 2 \int dt dt' (I_i(t) I_{Sj}(t') - \bar{I}_i \bar{I}_{Sj}) / \tau \tag{18}
\]

and the third cumulant of electric currents

\[
C_{ijk} = \frac{2}{\tau} \int dt dt' dt''(I_i(t) - I_i)(I_j(t') - I_j)(I_k(t'') - I_k) = \frac{2}{\tau} \int dt dt' dt'' \left( I_i I_j I_k - [I_i I_j I_k I_i + I_k I_i I_j] + 2 \bar{I}_i \bar{I}_j \bar{I}_k \right), \tag{19}
\]

where the bar means the average with respect to quantum and thermal fluctuations.

We are particularly interested in the limit of the low biases \( V \) and \( V_S \) and thus focus on the Taylor expansion of the above correlators. We assume that the currents and correlation functions are analytic in the biases \( V \) and \( V_S \). Then, up to the linear terms in \( V \) and \( V_S \), the expressions for the noises considerably simplify:

\[
S_{ij} = 2 \bar{I}_i \bar{I}_j = 2 \langle \Delta Q_i \Delta Q_j \rangle / \tau; \tag{20}
\]

\[
S_{ij}^S = 2 \bar{I}_{Si} \bar{I}_{Sj} = 2 \langle \Delta S_{Si} \Delta S_{Sj} \rangle / \tau; \tag{21}
\]

\[
S_{ij}^{\text{cross}} = 2 \bar{I}_i \bar{I}_{Sj} = 2 \langle \Delta Q_i \Delta S_{Sj} \rangle / \tau. \tag{22}
\]

In the zeroth order in the bias, the third cumulant reduces to

\[
C_{ijk} = 2 \bar{I}_i \bar{I}_j \bar{I}_k = -2 \langle \Delta Q_i \Delta Q_j \Delta Q_k \rangle / \tau. \tag{23}
\]

Thus, in the simplest cases, the cumulants reduce to the correlation functions \( \langle \rangle \). Higher-order cumulants are combinations of the correlation functions \( \langle \rangle \).

We now use Eq. \( 12 \) to establish fluctuation relations for the Taylor coefficients of the correlation functions \( \langle \rangle \). We define the coefficients \( L_{\nu,\mu}^{n,m}(B) \) according to

\[
\langle \Delta Q^m_{\alpha S}^{\nu} \rangle_{V,V_S} = \sum_{\nu,\mu} L_{\nu,\mu}^{n,m}(B) \prod_i \left( \frac{V_i}{T} \right)^{\nu_i} \left( \frac{V_{Si}}{T} \right)^{\mu_i}. \tag{24}
\]

Next, we substitute Eq. \( 12 \) into the right hand side of Eq. \( 13 \) and expand the exponent in powers of \( V \) and \( V_S \). A comparison with Eq. \( 24 \) yields a family of general relations among the correlation functions at the opposite orientations of the magnetic field:

\[
L_{\nu,\mu}^{n,m}(B) = (-1)^{\Sigma_{i}[n_i,\mu_i]} \sum_{\nu,\mu} \prod_{i=0}^{n} \frac{1}{u_i! u_i!} L_{\nu-u,\mu-w}^{n+m-w}(B), \tag{25}
\]

where \( \nu - u \) denotes the vector with the components \( \nu_i - u_i \), \( \sum_{i=0}^{n} \nu_i = 0 \), \( \sum_{i=0}^{n} \mu_i = 0 \) and the coefficients \( L_{\nu,\mu}^{n,m} \) can only be nonzero for nonnegative \( \rho_i \) and \( \sigma_i \). The fact that spin is odd under time reversal manifests itself in the sign factor \( (-1)^{\Sigma_{i}[n_i,\mu_i]} \) in the above equation. If spin were even then the sign factor would be \( (-1)^{\Sigma_{i}[n_i,\mu_i]} \). This illustrates the difference of the fluctuation relations for spin currents from fluctuation relations for multi-components currents of time-reversal-invariant quantities.

Eq. \( 25 \) is our main result.

**IV. EXAMPLES**

In this section we derive several consequences of Eq. \( 25 \) and compare them with the results of the related work\footnote{Note 11}. We start with a simple helpful identity \( \langle \Delta Q^0 \Delta S^0 \rangle = 1 \). As a consequence,

\[
L_{\nu,\mu}^{0,0} = \delta_{\nu,0} \delta_{\mu,0}. \tag{26}
\]

We will use this equation to derive nontrivial relations between transport coefficients.

As a warming-up exercise, we use Eqs. \( 25 \) \( 26 \) to develop the linear response theory for a two-terminal conductor, i.e., for a system with two reservoirs. To simplify our notations, we note the asymmetry between the indices \( n \) and \( m \) and between the indices \( \nu \) and \( \mu \). Charge conservation dictates that \( \sum_i \Delta Q_i = 0 \). Thus, the components of \( \Delta Q \) are not independent. For a two-terminal conductor, \( \Delta Q_1 = -\Delta Q_2 \). Thus, without the loss of
generality we can set \( n_2 = 0 \). Besides, gauge invariance tells us that the currents and noises depend only on the voltage difference between the reservoirs and not the absolute values of their chemical potentials. Thus, without the loss of generality, we are allowed to set \( V_2 = 0 \). In such situation all nonzero contributions to correlation functions come from the coefficients \( L^{m,n}_{\nu^2} \) with \( \nu_2 = 0 \). Below we will omit the zero indices \( n_2 \) and \( \nu_2 \) and write \( L_{\nu^1,\mu_1,\nu_2}^{m,n} \) for \( L^{m,n}_{\nu^2} \). We now extract the following consequence from Eqs. (25,26):

\[
0 = L^{000}_{110}(\bar{B}) = -\frac{1}{2} \left[ \partial I_{110} - \partial I_{010} \right](-\bar{B}) = \frac{1}{2} \left[ \partial I_{110} - \partial I_{010} \right](-\bar{B}),
\]

and hence

\[
L^{110}_{000}(\bar{B}) = -L^{100}_{010}(\bar{B}) - L^{010}_{100}(\bar{B}). \tag{27}
\]

A similar relation can be found for \( L^{010}_{000}, L^{100}_{000} \) and \( L^{000}_{000} \). In terms of currents and noises, the relation (28) is written as

\[
S_{11}^{\alpha} = 2T \left( \frac{\partial I_1}{\partial S_1} + \frac{\partial I_{S1}}{\partial V_1} \right). \tag{29}
\]

Noticing that \( \frac{\partial I_1}{\partial V_1} \) and \( \frac{\partial I_{S1}}{\partial V_1} \) are conductances, we see that this relation is a generalization of the standard Nyquist formula.

Other fluctuation-dissipation relations that follow from Eq. (25) include

\[
L^{110}_{000}(\bar{B}) = -L^{100}_{010}(\bar{B}); \tag{30}
\]

\[
L^{100}_{010}(\bar{B}) = L^{110}_{000}(\bar{B}) + L^{100}_{000}(\bar{B}); \tag{31}
\]

\[
L^{010}_{100}(\bar{B}) = L^{110}_{000}(\bar{B}) + L^{100}_{000}(\bar{B}). \tag{32}
\]

Combining Eqs. (27,32) one finds

\[
L^{100}_{000}(\bar{B}) = -L^{010}_{100}(\bar{B}), \tag{33}
\]

which can be more explicitly written as

\[
\frac{\partial I_1(\bar{B})}{\partial V_1} = \frac{\partial I_{S1}(\bar{B})}{\partial V_1}. \tag{34}
\]

The latter resembles the famous Onsager reciprocal relation \( G_{i\gamma}(\bar{B}) = G_{\gamma i}(-\bar{B}) \), where \( G \) are electric conductances. However, (34) has an additional minus sign, which essentially comes from the fact that spin is odd under time reversal. A similar relation to (33) for multicomponent currents of time-reversal-invariant quantities, obtained in Ref. 10, has no minus sign in front of \( L^{100}_{000} \) either.

What about nonlinear response? To facilitate the comparison with Ref. 11 we modify our notations. Instead of the charge and spin currents we will speak about the charge currents of spin-up and spin-down electrons, \( I_{\alpha} \), where \( \alpha = + \) corresponds to the spin-up current and \( \alpha = - \) corresponds to the spin-down current. The charges of the spin-up and -down electrons in reservoir \( i \) are \( Q_{\alpha i} = Q_i/2 - \alpha e S_i \). The conjugate chemical potentials \( V_i = V_i - \frac{1}{2} V_N \). Under time-reversal, \( Q_{\alpha i} \rightarrow Q_{\alpha i}^* \), \( V_i \rightarrow V_i^* \), where \( \alpha = -, \bar{\alpha} = + \). The fluctuation theorem (12) becomes

\[
P(\Delta Q,V, B) = \exp \left[ -\frac{\mathbf{V} \cdot \Delta \mathbf{Q}}{\overline{T}} \right] P(-\Delta \mathbf{Q}, \nabla, -\mathbf{B}), \tag{35}
\]

where \( Q \) and \( V \) stay for the vectors with the components \( Q_{\alpha i} \) and \( V_i \), \( Q \) and \( V \) are the vectors with the components \( Q_{\bar{\alpha}i} \) and \( V_i \), and \( \mathbf{V} \cdot \Delta \mathbf{Q} = \sum_{\alpha i} V_{\alpha i} \Delta Q_{\alpha i} \).

As the first example of a fluctuation relation for nonlinear transport, we derive a relation that connects the nonlinear conductance with the noise (20) and the third cumulant (23). We sum the right and left hand sides of Eq. (35) over all possible \( \Delta Q \). We get \( \left\langle 1 \right\rangle_{V,B} = 1 \) on the left and expand the sum on the right to the third order in \( V \). We find

\[
C_{i\alpha,j\beta,k\gamma}(V = 0, B) = T \left[ \frac{\partial S_{i\alpha,j\beta,k\gamma}(V = 0, B)}{\partial V_i} + (\text{cyclic permutations}) \right] - 2T^2 \left[ \frac{\partial^2 I_{k\gamma}(V = 0, B)}{\partial V_i \partial V_j} + (\text{cyclic permutations}) \right], \tag{36}
\]

where (cyclic permutations) stays for the cyclic permutations of the pairs of the indices \( \alpha, j \beta \) and \( k \gamma \). This result is similar to the fluctuation relations from Refs. 11,12 and could be obtained without the use of microreversibility.

How does one go beyond what can be obtained with the method in Ref. 11? We use Eq. (35) to compute \( \langle \Delta Q_{i\alpha} \rangle_{V,B} - \langle \Delta Q_{i\alpha} \rangle_{\nabla,-B} \). We next subtract the right hand side of the equation from its left hand side and expand to the second order in \( V \). The result is

\[
C_{i\alpha,j\beta,k\gamma}(V = 0, B) - C_{i\alpha,j\beta,k\gamma}(V = 0, -B) =
T \left[ \frac{\partial S_{i\alpha,j\beta,k\gamma}(V = 0, B)}{\partial V_i} + \frac{\partial S_{i\alpha,k\gamma}(V = 0, B)}{\partial V_j} \right] - \frac{\partial S_{i\alpha,j\beta,k\gamma}(V = 0, -B)}{\partial V_j}, \tag{37}
\]

Summing Eq. (37) over all possible choices of the spin indices and taking the limit \( V = \nabla \), one reproduces a fluctuation relation for electric currents. The latter relation cannot be obtained without microreversibility and the same is true for our result (37) which thus goes beyond the set of the fluctuation relations that could be derived with the method of Ref. 11.
V. DISCUSSION

Equations (27, 34–37) are the simplest consequences of the fluctuation theorem (25). Several other relations can be obtained by combining these equations, or using the symmetry of the correlation functions. For example, the third cumulants and noises are symmetric under permutations of the indices. One may also find fluctuation relations for higher-order correlation functions and for higher-order derivatives of the currents, noises and third cumulants. We note that equations (27–34, 36, 37) only involve up to the second derivatives of currents and first derivative of noises.

Our derivation relies on the combination of microreversibility with the assumption that the \( z \)-component of spin conserves approximately in large parts of the reservoirs near the conductor. If the latter assumption does not hold then it may not be even meaningful to speak about the spin currents, injected from the reservoirs into the conductor. Note that very long spin-coherence times have been reported for donor spins in silicon\textsuperscript{18}.

Our results do not depend on a particular model and thus apply to many systems. One example is the spin current rectifier\textsuperscript{21}. Since our fluctuation relations are exact, they can be employed for testing approximations in theoretical calculations.

The discussion in the preceding sections is most directly connected with mesoscopic conductors that can carry both spin and electric currents. At the same time, the presence of an electric current is not essential for the validity of our results. They also apply to pure spin currents. For example, our fluctuation relations hold for spin diffusion in insulators\textsuperscript{22}. The above calculations assume that the spin current is carried by particles with spin \( 1/2 \). It is straightforward to extend our results to higher spins. Another interesting setting is topological matter with chiral transport\textsuperscript{8}. We expect our fluctuation relations to simplify considerably in such systems since chirality implies the vanishing of many transport coefficients\textsuperscript{8}.

In conclusion, we prove a general fluctuation theorem for spin currents. It imposes a great number of restrictions on transport coefficients and fluctuations of the spin and charge currents in linear and nonlinear transport.

Acknowledgments

CW acknowledges the support by the NSF under Grant No. DMR-1254721. DEF was supported in part by the NSF under Grant No. DMR-1205715.

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