Nonequilibrium spin fluctuations in single-electron transistors

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We show that nonequilibrium spin fluctuations significantly influence the electronic transport in a single-electron transistor, when the spin relaxation on the island is slow compared to other relaxation processes, and when size effects play a role. To describe spin fluctuations we generalize the ‘orthodox’ tunneling theory to take into account the electron spin, and show that the transition between consecutive charge states can occur via a high-spin state. This significantly modifies the shape of Coulomb steps and gives rise to additional resonances at low temperatures. Recently some of our predictions were confirmed by Fujisawa et al. [Phys. Rev. Lett. 88, 236802 (2002)], who demonstrated experimentally the importance of nonequilibrium spin fluctuations in transport through quantum dots.

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Electronic transport in single electron transistors (SETs) with metallic islands or semiconducting quantum dots (QDs) was usually described in terms of the ‘orthodox’ theory of sequential tunneling [1]. The relaxation processes on the island were characterized only by the energy relaxation time \(\tau_E\), with the tacit assumption that the spin-flip relaxation time \(\tau_{sf}\) is short or comparable to \(\tau_E\) [2–4]. In real systems, however, the ratio \(\tau_{sf}/\tau_E\) can be as high as \(10^3\) - \(10^4\) [5–7]. In this letter we show that, when the spin relaxation time is significantly longer than the energy relaxation time \(\tau_E\) and injection time \(\tau_I\), large nonequilibrium spin fluctuations (NSF) arise on the island. When the density of states for the island is low, e.g., due to size effects, the spin fluctuations lead to fluctuations of the spin-splitting of the chemical potential. This, in turn, can significantly modify the transport characteristics. In particular, the Coulomb steps are smeared because transitions to higher charge states may occur via a set of associated spin states, each of them corresponding to different temporary electrochemical potentials. Such NSF can limit the performance of spin electronic devices. The importance of spin fluctuations has been recently confirmed experimentally by Fujisawa et al. [8] for a few-electron QD.

The effects described here differ from other phenomena related to energy level quantization and electron spin, which have been investigated recently theoretically [2,3] and experimentally for paramagnetic [9–11] and ferromagnetic grains [12,13]. They also differ from further spin-related phenomena, like spin blockade due to exchange interaction [14], parity effects [15], Kondo effects [16,17], and quantum entanglement.

Before we analyze spin effects in normal metal SETs, we consider first the corresponding double tunnel junction without Coulomb blockade and with a linear current-voltage relation. Tunneling of a single electron increases or decreases the magnetization \(M\) of the island by 1. Here \(M = N_↑ - N_↓\), where \(N_↑\) (\(N_↓\)) are the numbers of excess electrons with spin \(\sigma = ↑\) (\(\downarrow\)). In the absence of intrinsic spin-flip processes, the time evolution of the magnetization \(M(t)\) can be mapped onto a one-dimensional diffusion process with \(\langle M \rangle = 0\) and \(\langle M^2 \rangle^{1/2} \sim \sqrt{t}\). There are, however, two main processes which restrict the increase of the fluctuations: (i) spin-flip relaxation processes, and (ii) spin splitting of the electrochemical potential \(\mu_\uparrow \neq \mu_\downarrow\) (two spin components of the island can be treated as two independent electron reservoirs with different electrochemical potentials \(\mu_\sigma\) due to spin accumulation \(M\) [18], which modifies the tunneling probability. In the non-equilibrium situation due to an applied bias \(V\), fluctuating numbers \(N_\sigma\) give rise to nonequilibrium fluctuations \(N_\sigma \Delta E\) of the electrochemical potentials \(\mu_\sigma\) (here \(\Delta E = 1/D(E_F)\) and \(D(E_F)\) denotes the density of states at the Fermi level in the island). These fluctuations influence the transport current. The currents flowing through the left \(L\) junction for both spin orientations are \(I_{L\uparrow} = 1/2 (VR_L/R + M\Delta E/2e)/R_L\) and \(I_{L\downarrow} = 1/2 (VR_L/R - M\Delta E/2e)/R_L\). \(R_R\) (\(R_L\)) denote the resistance of the right (left) junctions and \(R = R_R + R_L\). However, the total current \(I_L = I_{L\uparrow} + I_{L\downarrow} = V/R\) is independent of \(M\). Straightforward calculation shows that for \(eV/\Delta E \gg 1\) and for low temperatures the magnetization fluctuations are described by a Gaussian distribution with the standard deviation \(\langle M^2 \rangle^{1/2} \approx \sqrt{(eV/\Delta E) a/(a+1)^2}\), where \(a = R_R/R_L\).

In contrast to the example discussed above, NSF in systems with nonlinear current-voltage characteristics can influence the average transport current. We will describe now how spin fluctuations influence transport through a SET, where the charge fluctuations are strongly sup-
pressed due to the Coulomb interaction, but spin fluctuations can be relatively large. For charging effects the relevant charging energy is $E_C = e^2/2C$, where $C = C_L + C_R + C_g$ is the total capacitance of the island, which is the sum of capacitances of the left ($C_L$) and right ($C_R$) junctions and of the gate ($C_g$). The electronic transport through the system in a stationary state is governed by the solution of the generalized master equation

$$0 = -\left\{ \Gamma(N^+,N^+\downarrow) + \Omega_{\downarrow} + \Omega_{\uparrow} \right\} P(N^+\uparrow,N^+\downarrow) + \Gamma_{\uparrow}(N^+\uparrow-1,N^+\downarrow) P(N^+\uparrow-1,N^+\downarrow) + \Gamma_{\downarrow}(N^+\downarrow+1,N^+\downarrow) P(N^+\downarrow+1,N^+\downarrow) + \Omega_{\downarrow}(N^+\downarrow-1,N^+\downarrow+1) P(N^+\downarrow-1,N^+\downarrow+1) + \Omega_{\uparrow} + \Omega_{\downarrow} \right\} P(N^+\downarrow,N^+\downarrow)$$

for the probability $P(N^+\uparrow,N^+\downarrow)$ to find $N^+\uparrow$ and $N^+\downarrow$ excess electrons on the island ($N = N^+\uparrow + N^+\downarrow$ is the total number of excess electrons). The first term in Eq. (1) describes the rate at which the probability of a given configuration decays due to electron tunnelling to or from the island, whereas other terms describe the rate at which this probability increases. The $\Omega$ terms account for spin-flip relaxation processes. The coefficients entering Eq. (1) are defined as $
abla_N^\pm(N^\uparrow\downarrow)$ and $\Gamma(N^\uparrow\downarrow)$ are the tunneling rates for electrons with spin $\sigma$, tunneling to (+) the grain from the lead $\ell = L, R$ or back (–). These coefficients are given by the following expression:

$$\frac{\Gamma_{\sigma}}{\Gamma_r} = \sum_{i,j}^{\gamma_{\sigma}} \sum_{N^\uparrow,N^\downarrow} \omega_{i\sigma,j\sigma} F_{i\sigma}(E_{i\sigma}) (N^\uparrow,N^\downarrow) f^\pm(E_{i\sigma} + E_{\sigma}^\pm(N^\uparrow,N^\downarrow) - E_F),$$

$$\Omega_{\sigma\sigma}(N^\uparrow,N^\downarrow) = \sum_{i,j}^{\gamma_{\sigma}} \sum_{N^\uparrow,N^\downarrow} \omega_{i\sigma,j\sigma} F_{i\sigma}(E_{i\sigma}) (N^\uparrow,N^\downarrow) \times F_{j\sigma}(E_{j\sigma}) (N^\uparrow,N^\downarrow).$$

Here, $f^\pm(E) = 1 - f^\pm$ is the Fermi distribution function, whereas $F_r^\pm(E_{i\sigma}(N^\uparrow,N^\downarrow))$ and $f^\pm E_{i\sigma}(N^\uparrow,N^\downarrow) - E_F)$ describes the probability that the energy level $E_{i\sigma}$ is occupied by an electron with spin $\sigma$ for the particular configuration $(N^\uparrow,N^\downarrow)$. The parameter $\gamma_{\sigma}$ is the tunneling rate of electrons between the lead $\ell$ and the energy level $E_{i\sigma}$ of the island, and $\omega_{i\sigma,j\sigma}$ is the transition probability from the state $i\sigma$ to $j\sigma$ due to the spin-flip processes. The energies $E_{i\sigma}^\pm(N) = C_{i\sigma}/eV + U^\pm(N)$ and $E_{i\sigma}^\pm(N) = -C_{i\sigma}/eV - U^\pm(N)$, where $U^\pm(N) = eC[2(N - N^\downarrow) \pm 1]$. We denote the gate voltage $V_g$ with $V_g$, from the solution $P(N^\uparrow,N^\downarrow)$ of the master equation (1) we obtain the current flowing through the island

$$I_r = e \sum_{\sigma} \sum_{N^\uparrow,N^\downarrow} P(N^\uparrow,N^\down\downarrow) \left\{ \Gamma_{\uparrow\sigma}(N^\uparrow,N^\down\downarrow) - \Gamma_{\down\sigma}(N^\up\down\downarrow) \right\}.$$

In our calculations we assume that discrete energy levels $E_{i\sigma}$ are equally separated, with the level spacing $\Delta E$. The tunneling rates $\gamma_{\sigma}$ are then given by the formula

$$\gamma_{\sigma} = \Delta E/e^2 R_{\sigma}$$

for spin-flip rates $\gamma_{\sigma}$ and slow $\left( \gamma_{\sigma} \ll \gamma_{\sigma} \right)$ spin-flip relaxation rates, where $\gamma_{\sigma} \sim 1/\tau_{\sigma} \sim e/I$ (for QDs, it is shown in Ref. [6] that $\tau_{\sigma} > 1\mu s$, $\tau_{\sigma} < 1 ns$ and $\tau_{\sigma} \sim 1 \div 10 ns$ for $I \sim 0.1 \div 1 nA$). In Fig. 1(a,f) the conductance-voltage characteristics are presented for both limits. The effect on the current is relatively weak (a few per-cent at maximum), as shown in Fig. 1(b,g). However, the effect on the differential conductance can be significantly larger, about a few tens of per-cent at maximum, as shown in Fig. 1(c,h). Generally, the most pronounced effect of the spin fluctuations is on the nonlinear parts of the transport characteristics. This effect is similar to the one produced by an increase in effective temperature $T_{\text{eff}}$ [22,23] of the system (different from the bath temperature $T$). However, both effects can be easily distinguished because of the parabolic dependence on the bias voltage $V$ of the former effect (see the discussion below). For intermediate spin-flip times, $0.1 < \tau_{\sigma} \ll 10$, the conductance smoothly crosses over from one limit to the other.

In Fig. 1(d,i) we show the bias dependence of the spin fluctuations ($M^2$) in the limits of both short and long spin relaxation times. In the former case the fluctuations are almost constant and close to 1. In the latter case and for symmetrical junctions, the fluctuations nearly follow the law for junctions without Coulomb effects. However, for asymmetric junctions there are pronounced oscillations in the fluctuations amplitude with increasing bias $V$, which are related to the charge accumulation and correlated with the charge fluctuations [21] (see Fig. 1(j)). For comparison, we show in Fig. 1(e,j) the amplitude of charge fluctuations. For asymmetric junctions the fluctuations are due to charge accumulation.

In the inset of Fig. 1(e), we show the probability $P(N = 1)$ in the transition range (from $N = 0$ to $N = 1$) for the symmetrical junction. One can see that the effect of the Coulomb interaction is similar to the one produced by an increase in the nonlinear parts of the transport characteristics. This effect is similar to the one produced by an increase in effective temperature $T_{\text{eff}}$ [22,23] of the system (different from the bath temperature $T$). However, both effects can be easily distinguished because of the parabolic dependence on the bias voltage $V$ of the former effect (see the discussion below). For intermediate spin-flip times, $0.1 < \tau_{\sigma} \ll 10$, the conductance smoothly crosses over from one limit to the other.

In Fig. 2(d,i) we illustrate the mechanism how the spin fluctuations assist the system to enter the higher charge states. For strong NSF, an electron needs a lower energy to enter the island, because of the double degenerate states below the Fermi energy are empty (compare Fig. 2(b) and Fig. 2(a)). Figure 2(c) illustrates that the onset of the transition to the next charge state is linked to the existence of a high spin state, and the
new charge state at the onset can appear only via the high spin state. The effect is relevant as long as the energy of thermal fluctuations is lower than the energy of NSF, $2\langle M^2 \rangle^{1/2} \Delta E > k_B T$. Thus, even if the discrete states are not resolved ($k_B T \gtrsim \Delta E$), the effects due to spin fluctuations can be important. In Fig. 3 we show the amplitude of NSF as a function of $\Delta E$. From this figure follows that the NSF effects can be observed in electronic transport when $\Delta E/E_C \gtrsim 0.01$. This condition can be easily achieved for typical QDs [1] of radius 200nm, $2E_C \sim 1$meV, $\Delta E \sim 0.03$meV and for $T < 1K$. For smaller QDs the effect is important also at higher temperatures. For larger islands with high density of states $D(E_F)$ the fluctuations of $M$ have very large amplitude ($\sim \sqrt{eV/\Delta E}$), but the associated effective spin splitting of the electrochemical potential $M \Delta E$ is small ($\sim \sqrt{eV \Delta E}$). For metallic systems the condition $2\langle M^2 \rangle^{1/2} \Delta E > k_B T$ could be fulfilled for small metallic particles with diameter in the range of several $nm$. For strong exchange interactions QDs can have a ground state with spin $S > 1/2$ [14,24]. But even in this case spin fluctuations are important, as pointed out by Kleff and von Delft [13] for ferromagnetic grains.

The time $\tau_{sf}$ depends mainly on the strength of the spin-orbit interaction and on the impurity contents [7]. Thus counter-intuitively, for SETs with dirty island and strong spin scattering one should expect sharper transport characteristics. The NSF effects can be detected by measurements of the conductance peak or peak widths as a function of temperature. The widths should saturate at a value that scales with the bias voltage as $V^{1/2}$. In the absence of NSF there will be no saturation observed, or the saturation will be at a value (related to some $T_{eff}$) which does not depend on the bias as $V^{1/2}$.

Very recently, Fujisawa et al. [8] obtained results for very small QDs at low temperatures, which confirm the importance of NSF and which can be accounted for by our model. In Fig. 2(d) we show schematically the positions of the conductance peaks (current steps), i.e., dc excitations in the low-temperature limit, $k_B T \ll \Delta E$, where discrete energy levels are resolved. In addition to the standard resonance peaks (corresponding to the dotted lines), there are new ones indicated by the dashed lines which start inside the diamond corresponding to single electron tunneling transport (SETT) (not at its border) when a particular spin excitation appears. Double electron tunneling transport (DETT) can occur already within the SETT diamond region due to NSF (the regions marked in gray in Fig. 2(d), where also the magnetic state of the excitations is indicated). We find that not only in results of Ref. [8] but also in earlier data (see e.g. Fig.1.(A) in Ref. [25]) the borders between SETT and DETT diamonds could be affected by spin fluctuation. The new resonance peaks are related to the fact that the ratio $E_C/\Delta E$ is generally not integer, and the electron levels for adjacent charge states are effectively shifted by $mod(E_C,\Delta E)$.

One may expect a similar effect of NSF on the cotunneling current, too. The fluctuations can influence in particular the first step in the current-voltage characteristic (the first peak in the conductance). One may also expect that NSF modify the current shot noise.

In summary we have generalized earlier spinless descriptions of SETs by taking into account the fact that spin-flip relaxation time $\tau_{sf}$ is usually much longer than the energy relaxation time $\tau$. The spinless models describe properly only the situation when $\tau_{sf}$ is short or comparable to $\tau$. When, however, $\tau_{sf} \gg \tau$, large nonequilibrium spin fluctuations may occur in the island. These fluctuations have a significant influence on the transport characteristics. Our model accounts for recent experimental observations [8].

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FIG. 1. (a,f) Differential conductance vs. bias voltage $V$ for two limits: short (dashed line) and long (solid line) spin-flip relaxation time; (b,g) relative difference $(I_1 - I_s)/I_s$, where $I_1$ and $I_s$ are the currents for the two limits; (c,h) relative difference $(G_1 - G_s)/G_s$; (d,i) spin fluctuations $(M^2)^{1/2}$; (e,j) charge fluctuations $(\langle N^2 \rangle - \langle N \rangle^2)^{1/2}$. The curves are calculated for symmetric (left column) and asymmetric junctions (right column). Inset: The probability $P$ of the $N = 1$ charge state for a symmetrical junction as a function of the bias voltage $V$. For symmetric junctions $C_R = C_L$, $R_0 = R_L$, and the gate voltage $N_s = 0.5 + 1/4 \Delta E/E_C$ (at resonance), whereas for asymmetrical junctions $C_R/C_L = 3$, $R_0/R_L = 100$ and the gate voltage $N_s = 0$. The other parameters are $k_B T = 0.4 \Delta E$ and $\Delta E/E_C = 0.1$ for both cases.
FIG. 2. Examples of specific spin configurations of the island for the same charge state of the system with (a) short and (b) long spin-flip relaxation times. Part (c) shows how the system enters the next charge state. The probability $P(N_\uparrow, N_\downarrow)$ for $N = (N_\uparrow + N_\downarrow) = 1$ is shown there as a function of $M = (N_\uparrow - N_\downarrow)$ for several values of the bias voltage $V$. All parameters are the same as in Fig. 1(a). The lines are a guide to eye only. Data for different $V$ are offset vertically. (d) The scheme of conductance peaks (current $I$ plateaus) for dc excitation transport in the $V - V_g$ plane for symmetric junction, $E_C/\Delta E = 4.5$ and $k_B T \ll \Delta E$. Solid lines determine the Coulomb blockade (CB) diamond structure indicating also the onset of single electron tunneling transport (SETT) and double electron tunneling transport (DETT) (two excess electrons on the island are possible) in the absence of NSF. Dashed lines are usual effects related to offset in transport of the next discrete energy levels. Dashed and dotted-dashed lines indicate the onset of the consecutive charge states due to particular spin excitations (as indicated) which without spin fluctuations are not accessible. Dashed lines indicate also formation of new resonances due to NSF. Symmetry is broken due to the parity effect.

FIG. 3. Spin fluctuations $\langle M^2 \rangle^{1/2}$ (dashed line) and the corresponding splitting of electrochemical potential $\langle M^2 \rangle^{1/2} \Delta E/E_C$ (solid line) as a function of energy level spacing $\Delta E/E_C$, calculated for $k_B T = 0.04E_C$ and $eV/2E_C = 3$. The other parameters are as in Fig. 1(a).