Spin accumulation in small ferromagnetic double-barrier junctions

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The nonequilibrium spin accumulation in ferromagnetic double barrier junctions is shown to govern the transport in small structures. Transport properties of such systems are described by a generalization of the theory of the Coulomb blockade. The spin accumulation enhances the magnetoresistance. The transient non-linear transport properties are predicted to provide a unique experimental evidence of the spin accumulation in the form of a reversed current on time scales of the order of the spin-flip relaxation time.
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In the 1970s it was understood that electron transport in tunneling and heterostructures involving metallic ferromagnets is associated with nonequilibrium spins.[1,2] Compared to other time scales in electron transport the spin relaxation time is generally very long at low temperatures, being limited only by scattering at paramagnetic impurities and by spin-orbit scattering. The spin-relaxation time and the spin-diffusion length which govern the spin accumulation has been measured by Johnson in polycrystalline gold films.[3] The concept of nonequilibrium spin accumulation plays an important role in the Boltzmann theory of transport of the giant magnetoresistance in the current perpendicular to the plane (CPP) configuration.[4,5] However, the experimental evidence for the spin accumulation is indirect at best. It can be shown that in the linear response regime the spin- and charge-distribution functions can be completely integrated out of the transport problem, which then depends exclusively on the scattering probabilities and the applied bias.[5] In this paper we show theoretically how unambiguous evidence for a nonequilibrium spin accumulation can be obtained by the dc and ac response of ferromagnetic double barrier junctions in the nonlinear regime. These junctions have to be small in order to observe large effects, which means that the complications of the Coulomb blockade have to be taken into account (for a review see Ref. 6). To this end we have to extend very recent theories of the Coulomb blockade in ferromagnetic double barrier junctions[7] to include time dependence and a nonzero spin relaxation time.[8] Ono *et al.* succeeded in fabricating a ferromagnetic single electron transistor,[9] which in principle can be used to test our predictions. Coulomb charging effects have also been seen in discontinuous multilayers[10] and in small cobalt clusters.[11]

We first show that the spin accumulation in ferromagnetic double barrier junctions becomes relevant when the number of electrons in the island between the tunneling barriers is relatively small. In ferromagnetic structures where the tunneling rates depend on the electron spin, a finite current through the system is accompanied by a spin current out of or into the island $(\partial s/\partial t)_{\mathrm{tr}}$. This creates a nonequilibrium excess spin s on the island, which decays with the spin-flip relaxation time τ_{sf} so that in steady state $(\partial s/\partial t)_{\mathrm{tr}}=s/\tau_{\mathrm{sf}}$. Energy relaxation is much faster than spin relaxation, so that the occupation of the states for each spin direction can be described by Fermi distributions.[6] The nonequilibrium spin accumulation on the island is equivalent to a chemical potential difference $\Delta\mu$ between the spin-up and the spin-down states. Since spin relaxation is slow and the structures of interest are small, $\Delta\mu$ is uniform over a sufficiently small island. In terms of the typical single-particle energy spacing (or inverse energy density of states at the Fermi energy) δ we have $\Delta\mu=s\delta$. Spin accumulation may be expected to interfere with the transport properties when $\Delta\mu$ is of the same order as the applied voltage V. The spin current is of the same order as the current, $e(\partial s/\partial t)_{\mathrm{tr}}\sim I\sim V/R$, where R is the typical junction resistance. The nonequilibrium spin accumulation is therefore important when the spin-relaxation time and/or the single-particle energy spacing are sufficiently large:

$$\tau_{\rm sf}\delta/h > R/R_K, \tag{1}$$

where the quantum resistance is $R_K = h/e^2$. The spin-flip relaxation time in polycrystalline aluminum is $\tau_{\rm sf} \sim 10^{-10}$ s (Ref. 1) [10^{-8} s in single-crystal aluminum at $T = 4.3$ K (Ref. 2)] and $\tau_{\rm sf} \sim 10^{-11}$ s for gold.[3] The single-particle energy spacing on the island is roughly $\delta \sim E_F/N$, where N is the number of atoms on the island and $E_F \sim 10$ eV is the Fermi energy. In an Al island with less than 10^6 atoms (10^8 atoms in single crystals) the spin accumulation may therefore be expected to play a significant role. ''Modern'' metals, such as arm-chair nanotubes[12] or (magnetic) semiconductor heterostructures,[13] can also be interesting as island materials. The former because of a possible huge spin-flip relaxation time and the latter since islands containing a small number of electrons can be created by depletion of the two-dimensional electron gas.[6]

In small systems where Eq. (1) is satisfied the spin-flip relaxation time is longer than the charge relaxation time RC (C is the capacitance of the island). This can be seen from Eq. (1), $\tau_{\rm sf} > (2E_c/\delta)RC$, and noting that the charging energy is larger than the single-particle energy spacing except in few-electron systems, $E_C/\delta \sim (e^2/E_F a)N^{2/3}(e^2/E_F a \sim 1)$. Hence the long-time response of the system is dominated by the spin dynamics.

We consider a normal-metal island attached to two ferromagnetic leads by two tunnel junctions. We assume collinear magnetizations in the leads and disregard size quantization. The tunnel junctions are characterized by a capacitance C_i and magnetic configuration-dependent conductances $G_{i\sigma}$, where $i = 1,2$ denotes the first and the second junction and σ denotes up (+) or down (−) spin electrons on the island. There is a source-drain voltage V between the right and the left reservoir and a gate voltage source coupled capacitively to the island. Here we consider the situation with a maximum Coulomb gap where the offset charge controlled by the gate voltage is zero.[8]

We proceed from the assumptions of the orthodox theory, i.e., $G_{i\sigma} < G_K$ neglecting cotunneling,[14] with the difference that the transition rates becomes spin dependent. The transition rate from the left reservoir to the island is

$$\overrightarrow{\Gamma^{1\sigma}}_{n+1,n} = \frac{1}{e^2} G_{1\sigma} F(E_1(V,q) - \sigma\Delta\mu/2), \tag{2}$$

where the energy difference associated with the tunneling of one electron into the island through junction i is[6] $E_i(V,q) = \kappa_i eV + e(q - e/2)/(C_1 + C_2)$, the charge on the island is $q = -ne$, the total capacitance is $1/C = 1/C_1 + 1/C_2$, $\kappa_i = C/C_i$, $F(E) = E/[1 - \exp(-E/k_BT)]$, and k_BT is the thermal energy. The spin balance is

$$\frac{ds}{dt} = \left(\frac{ds}{dt}\right)_{\rm tr} + \left(\frac{ds}{dt}\right)_{\rm rel}, \tag{3}$$

where the spin-relaxation rate is $(ds/dt)_{\rm rel} = -s/\tau_{\rm sf} = -\Delta\mu/\delta\tau_{\rm sf}$, $\tau_{\rm sf}$ is the spin-flip relaxation time, and δ^{-1} is the density of states at the Fermi level in the island. The spin balance (3) can be written in the stationary case as $I_s = e(ds/dt)_{\rm tr} = G_s 2\Delta\mu/e$, where the ''spin relaxation conductance'' is introduced as $G_s \equiv e^2/2\delta\tau_{\rm sf}$. The master equation[6] determines the probability p_n to have n excess electrons on the island. The current through the first junction is $I_1 = (I_1^\uparrow + I_1^\downarrow)$, where the current of electrons with spin σ is $I_1^\sigma = e\Sigma_n p_n(\overrightarrow{\Gamma^{1\sigma}}_{n+1,n} - \overleftarrow{\Gamma^{1\sigma}}_{n-1,n})$ and there is a similar expression for the current through the second junction $I_2 = (I_2^\uparrow + I_2^\downarrow)$. The spin current is

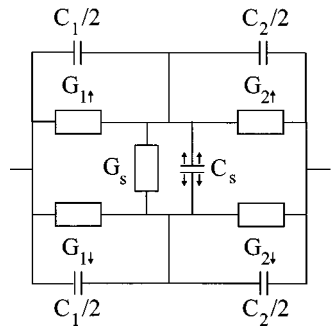

FIG. 1. The equivalent circuit for the current-voltage response of the system.

$$\left(\frac{ds}{dt}\right)_{\rm tr} = (I_1^\uparrow - I_1^\downarrow - I_2^\uparrow + I_2^\downarrow)/e. \tag{4}$$

In the Coulomb blockade regime the current is zero, $I = 0$, and it can be shown that $\Delta\mu$ vanishes, as expected.[8] The Coulomb gap in the low-temperature current-voltage characteristics is thus not modified by the nonequilibrium spin accumulation. We also want to point out that for symmetric tunneling junctions $G_{1\uparrow}/G_{1\downarrow} = G_{2\uparrow}/G_{2\downarrow}$ the nonequilibrium spin accumulation vanishes and our theory reduces to those in Refs. 7.

In this orthodox model the problem can be mapped on the equivalent circuit in Fig. 1 by introducing the ''spin capacitance'' $C_s \equiv e^2/2\delta$, so that

$$(es)/2 = C_s(\Delta\mu/e), \quad \Delta\mu/s = e^2/(2C_s) = \delta.$$

This ''charging energy'' of the spin capacitance is thus simply the single particle energy cost of a spin flip, δ, or more generally, the inverse of the magnetic susceptibility μ_B^2/χ_s.

We solve the general problem for the steady state as well as for the time-dependent properties by numerically integrating the master equation and the spin balance, Eq. (3). We choose symmetric capacitances $C_1 = C_2 = C$ in our calculations. Thus the important energy scale is the Coulomb energy $E_c = e^2/2C$ and the other relevant energies are renormalized by E_c. The thermal energy is $k_BT = 0.05E_c$. The spin-dependent junction conductances are described in units of the average junction conductance G and the currents are normalized by $Ge/2C$. In the parallel configuration, the conductances are $G_{1\sigma}^{\rm P} = G_1(1 + \sigma P)/2$ and $G_{2\sigma}^{\rm P} = G_2(1 + \sigma P)/2$, where P is the polarization of the ferromagnets. In the antiparallel configuration $G_{1\sigma}^{\rm AP} = G_1(1 + \sigma P)/2$ and $G_{2\sigma}^{\rm AP} = G_2(1 - \sigma P)/2$.

We consider first the steady-state transport properties where the spin capacitance C_s does not contribute. The junction magnetoresistance is the relative difference in the resistance when switching from the antiparallel to the parallel configuration. In the absence of the nonequilibrium spin ac-

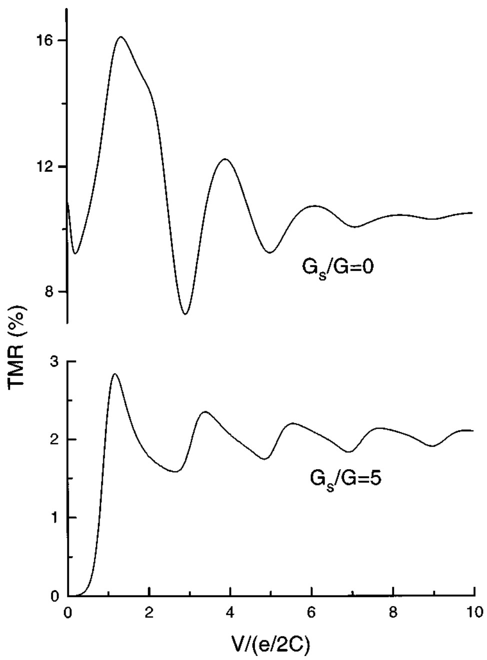

FIG. 2. The junction magnetoresistance in the limit of no spin relaxation in the island ($G_S/G=0$) and fast spin relaxation ($G_S/G=5$).

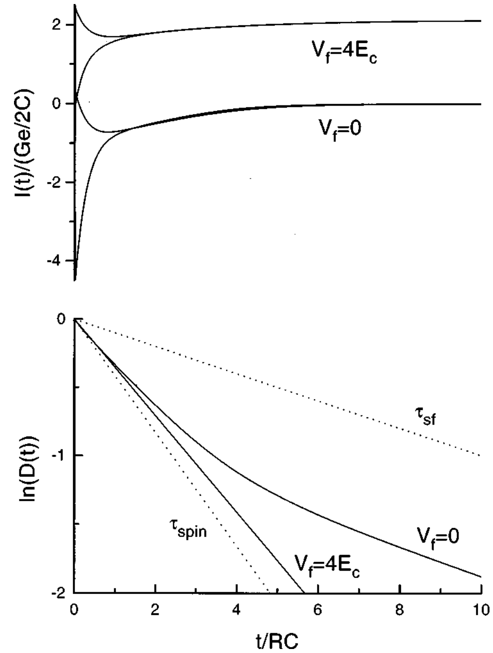

FIG. 3. The current as a function of time (upper panel). The relative change of the nonequilibrium spin as a function of time (lower panel). The source-drain voltage is switched from $V_i=10E_c$ to $V_f=4E_c$ or $V_f=0$ at $t=0$.

cumulation, the junction magnetoresistance vanishes for the $F/N/F$ junction. The spin accumulation causes a nonzero magnetoresistance. We show in Fig. 2 the calculated junction magnetoresistance for $G_1/G=1$, $G_2/G=2$ and a polarization $P=0.4$ in the limit of slow spin relaxation $G_s/G=0$ (upper curve) and fast spin relaxation $G_s/G=5$ (lower curve). We see the magnetoresistance oscillations as a function of the source-drain voltage.[7] The amplitude of the oscillations decreases with increasing source-drain voltage, where the Coulomb charging is less important.[7] The period of the oscillations is close to $2E_c$ for our system. There is only a small distortion of the shape of the magnetoresistance oscillations with increasing spin-relaxation rate in the island. The magnetoresistance and its oscillations are noticeable even when the spin-relaxation conductance is of the same order as the tunnel conductances, in agreement with Eq. (1). In the absence of the Coulomb charging energy, the tunnel magnetoresistance is

$$\text{TMR}=P^2\frac{1-\gamma^2}{1-P^2\gamma^2+\alpha^2}, \tag{5}$$

where $\gamma=(G_1-G_2)/(G_1+G_2)$ is a measure of the asymmetry of the junction conductances and $\alpha^2=4G_s/(G_1+G_2)$ determines the reduction of the magnetoresistance due to the spin relaxation. For a high source-drain bias when the Coulomb charging effects are negligible, the numerical results agree well with Eq. (5), TMR=11% for $G_s/G=0$ and TMR=2% for $G_s/G=5$.

For the transient response in the antiparallel configuration we use $P=0.5$, $G_1/G=1.3$, $G_2/G=2.6$, and $G_s/G=0.3$. Let us consider first a fixed source-drain voltage at a high bias until the system is stationary and then lower the source-drain voltage. We have used $\tau_{\text{sf}}=10RC$ (e.g., $E_c=0.2$ meV and $R/R_K=10$ gives $RC=2\times10^{-11}$ s). The initial high bias is $V_i=10E_c$ which gives a stationary current of $I_i=6.2Ge/2C$ and we investigate the behavior of the transient current when the final source-drain bias is below, $V_f=0$ $(I_f=0)$, and above the Coulomb charging energy, $V_f=4E_c$ $(I_f=2.1Ge/2C)$. We show in the upper panel in Fig. 3 the current through the first and the second junction for $V_f=4E_c$ (upper curves) and $V_f=0$ (lower curves) after the source-drain voltage is changed at $t=0$. It is clearly seen that the relaxation of the current is slow on the time scale RC. For time scales less than RC, we see that the current through the first and the second junction are not the same due to the charge depopulation in the island. The average of the upper curves ($V_f=4E_c$) where the final source-drain voltage is well above the Coulomb blockade energy, follows to within 10–20 % the description given by the equivalent circuit neglecting the Coulomb charging effects described below [(6), (7), and (8)] according to which the spin accumulation time is $\tau_{\text{spin}}=2.4RC$. When the source-drain voltage is switched off ($V_f=0$), we see that the transient current is negative. However, the spin accumulation time is much longer in this case, $\tau_{\text{spin}}\simeq\tau_{\text{sf}}=10RC$. This discrepancy becomes more evident when we consider the relative change of $\Delta\mu$ or s:

$$D(t)\equiv\left|\frac{s(t=\infty)-s(t)}{s(t=\infty)-s(t=0)}\right|.$$

In the lower panel of Fig. 3 we show the calculated time-dependent relative change $D(t)$ in the situations $V_f=4E_c$ (upper solid curve) and $V_f=0$ (lower solid curve), which are found to be remarkably different.

In order to understand the dynamics it is useful to inspect the device without the Coulomb charging effects, i.e., the capacitances C_1 and C_2 in the equivalent electric circuit in Fig. 1. We set the voltage on the left lead to zero and apply a time-dependent potential $V(t)$ to the right lead. The complex impedance $Z_{\rm spin}(\omega)=V(\omega)/I(\omega)$ is

$$\frac{1}{Z_{\rm spin}(\omega)}=\frac{G_1G_2}{G_1+G_2}-\frac{G_{1\uparrow}G_{2\downarrow}-G_{1\downarrow}G_{2\uparrow}}{(G_1+G_2)}\frac{\Delta\mu(\omega)}{eV(\omega)}, \quad (6)$$

where

$$\frac{\Delta\mu(\omega)}{eV(\omega)}=\frac{1}{1+i\omega\tau_{\rm spin}}\frac{G_{1\uparrow}G_{2\downarrow}-G_{1\downarrow}G_{2\uparrow}}{(G_s+G')(G_1+G_2)}. \quad (7)$$

Here the spin accumulation time is

$$\tau_{\rm spin}=\frac{C_s}{G_s+G'}, \quad (8)$$

where $1/G'=1/(G_{1\uparrow}+G_{2\uparrow})+1/(G_{1\downarrow}+G_{2\downarrow})$. From the relations (6) and (7) we see why switching off the source-drain voltage ($V_f=0$) reverses the transient current as found in the upper panel in Fig. 3. Without the Coulomb blockade this transient decays on the time scale $\tau_{\rm spin}$. In the limit that the junction conductances are much smaller than the spin conductance, the spin accumulation time (8) reduces to the spin-flip relaxation time, $\tau_{\rm spin}\approx\tau_{\rm sf}$. In the opposite limit where the junction conductances are much larger than the spin conductance, the spin accumulation time is $\tau_{\rm spin}\sim C_sR$. The spin capacitance is much larger than the charge capacitance C in the regime where the orthodox theory is valid ($\delta\ll E_C$) and thus the spin accumulation time is much larger than the charge-relaxation time.

The dashed lines in the lower panel in Fig. 3 correspond to the spin accumulation time in the absence of charging, $\tau_{\rm spin}=2.4RC$ as well as to the spin-flip relaxation time $\tau_{\rm sf}=10RC$. We see that the calculated spin accumulation time agrees well with the equivalent circuit described above [Eq. (8)] for $V_f=4E_c$, but disagrees with this expression for $V_f=0$ where the spin accumulation time is close to $\tau_{\rm sf}$. The latter is a result of the Coulomb charging which is seen to affect the spin accumulation time. In this case the nonequilibrium spin accumulation decays slower since the spins must relax through the spin conductance G_s on the island and the transport through the junctions is suppressed. In this situation, the relaxation time of the nonequilibrium spins and the current for long times is equal to the spin-flip relaxation time $\tau_{\rm sf}$, as observed in Fig. 3.

It should be noted that the magnon assisted inelastic tunneling, which reduces the TMR, gives negligibly small contribution in our case because of a magnon excitation gap, presumably due to magnetic anisotropy and/or size effects.[15] This magnon gap is larger than the bias voltage applied in our study. For very small islands like the metallic cluster studied in Ref. 11 when the Coulomb charging energy is larger than the magnon gap, magnon inelastic tunneling can interfere with the Coulomb charging effects.

In conclusion, we have investigated the influence of a nonequilibrium spin accumulation on the transport properties of a ferromagnetic single-electron transistor. For a $F/N/F$ junction we find a finite magnetoresistance due to the nonequilibrium spin accumulation. The spin accumulation can have a drastic effect on the ac transport properties. A transient response can be found on time scales much larger than the charge relaxation time RC. The same slow response is also expected if other external parameters such as the gate voltage or the magnetization are changed.

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