Spin accumulation in ferromagnetic single-electron transistors in the cotunneling regime

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We propose a new method of direct detection of spin accumulation, which overcomes problems of previous measurement schemes. A spin dependent current in a single-electron transistor with ferromagnetic electrodes leads to spin accumulation on the metallic island. The resulting spin-splitting of the electrochemical potentials of the island, because of an additional shift by the charging energy, can be detected from the spacing between two resonances in the current-voltage characteristics. The results were obtained in the framework of a real-time diagrammatic approach which allows to study higher order (co-)tunneling processes in the strong nonequilibrium situation.

73.23.Hk, 75.70.Pa, 73.40.Gk, 75.70.-i

The discovery of materials and devices with spin-dependent electronic transport properties opens new perspectives for research and applications. As examples we mention exchange-coupled metallic magnetic multilayers, heterostructures with ferromagnetic semiconductors, hybrid structures based on magnetic metals and nonmagnetic semiconductors, as well as perovskite oxides. The field has developed into a new branch of mesoscopic electronics – called magneto- or spin electronics. The balance of spin injection and spin diffusion in spin-polarized heterostructures in general leads to a nonequilibrium spin accumulation, which in turn produces a difference in the local electrochemical potentials for spin-up and down electrons. The predicted spin polarization was probed in experiments, either by measuring the resistance of metallic devices or in semiconducting systems by studying the circular polarized photoluminescence.

In spite of the reported successes in the indirect detection of spin accumulation, there remain controversies about the results. In these methods one usually measures the resistance and for a typical experimental configuration some other effects related to interface scattering, anisotropic magnetoresistance, etc., can occur. Even the question whether in principle it is possible to observe the spin splitting of the electrochemical potential by spectroscopic methods (analogous to the tunneling spectroscopy for the superconducting gap) remains open. The reason is the fact that the spin accumulation is a nonequilibrium effect, and the splitting of electrochemical potential is always smaller than the applied bias (in contrast to the superconducting energy gap which does not depend on the applied voltage).

In this article, we propose a method of direct detection of the nonequilibrium spin splitting of the electrochemical potentials in ferromagnetic single-electron transistors (FM SETs), which is free of the difficulties met in the methods used previously. We show that the splitting can be read from the spacing between two resonances in the current-voltage characteristics. The results are separated due to the spin dependence of the electrochemical potentials, but in addition they are shifted by the charging energy due to Coulomb blockade effects.

Spin-dependent transport in ferromagnetic double-barrier tunnel junctions has been studied before, both experimentally and theoretically in the sequential, cotunneling and strong tunneling regimes. The previous work covering the cotunneling regime is based on the approach developed by Averin and Nazarov for nonmagnetic SETs, which, however, is valid only away from the resonance. On the other hand, exactly the resonance is crucial for the effects considered in this paper. Therefore, we extend the previous descriptions by using the diagrammatic real-time technique, developed for normal-metal SETs, and extend it to FM SETs with spin-dependent tunneling. We take into account single-barrier cotunneling processes, vertex correction and propagator renormalization.

The system under study consists of a small nonmagnetic metallic grain (island) connected to two ferromagnetic leads (electrodes) via tunnel barriers. Its Hamiltonian takes the form

\[ H = \sum_{r=L,R} H_r + H_1 + H_{ch} + H_T = H_0 + H_T. \]  

Here \( H_r = \sum_{k\nu} \epsilon_{k\nu} a_{k\nu}^\dagger a_{k\nu} \) (for \( r = L, R \)) and \( H_1 = \sum_{q\sigma} \epsilon_{q\sigma} c_{q\sigma}^\dagger c_{q\sigma} \) describe noninteracting electrons in the two leads and island, respectively. The eigen-
states of the leads and island are described by the wave-vectors \( k \) and \( q \), transverse channel index \( \nu \), and spin \( \sigma \). The Coulomb interaction on the island is accounted for by \( H_{\text{ch}} = E_C(n - n_c)^2 \), where \( E_C = e^2 / 2C \) is the scale for the charging energy, and \( C = C_L + C_R + C_g \) is the total capacitance of the island, which is the sum of the left and right junction capacitances and the gate capacitance. The ‘external charge’ \( n_x \equiv C_L V_L + C_R V_R + C_g V_g \) accounts for the effect of the applied voltages, \( V_L \) and \( V_R \), in the left and right electrodes and the gate voltage \( V_g \). The last part of the Hamiltonian, \( H_T \), describes tunneling processes and may be written as

\[
H_T = \sum_{r=L,R} \sum_{kq} \sum_{\nu\sigma} T^\dagger_{kq\nu\sigma} a^\dagger_{kq\nu\sigma} e^{i\phi_{kq\nu\sigma}} + \text{h.c.}. \tag{2}
\]

The phase operator \( \hat{\phi} \) is the conjugate to the charge \( e\hat{n} \) on the island, and the operator \( e^{\pm i\phi_{kq\nu\sigma}} \) describes changes of the island charge by \( \pm e \).

When writing Eq. \( (2) \) we assumed that the electron spin and transverse channel index are conserved during tunneling. We will further assume for simplicity that the transfer matrix elements \( T^\dagger_{kq\nu\sigma} \) are only dependent on the junction \( r \) and the spin orientation \( \sigma \), \( T^\dagger_{kq\nu\sigma} = T^\dagger_{\sigma} \). They can be related to the spin dependent tunneling resistance of the barriers via the relation \( 1/R_{\sigma r} = (2\pi e^2 / h) N T^2_{\sigma} D_{\sigma r} \), where \( D_{\sigma r} \) is the spin-dependent density of electron states at the Fermi level such that they satisfy \( \mu_{\uparrow} = -\mu_{\downarrow} \). Without loss of generality we can choose the reference energy such that they satisfy \( \mu_{\uparrow} = -\mu_{\downarrow} \). Spin current conservation, as described later, fixes these values.

The grand-canonical density matrix of the system depends on the electrochemical potentials of the two spin components. Following the standard procedure \[5, 10\] we expand it in \( H_T(t) \) and perform the trace over the reservoir degrees of freedom using Wick’s theorem. The electric current is given in the lowest order perturbation expansion by \( I^{(1)} = \sum_{\sigma} I^{(1)}_{\sigma} = -\sum_{\sigma} J^{(1)}_{\sigma \sigma} \), with

\[
J^{(1)}_{\sigma \sigma} = \frac{4\pi^2 e}{h} \sum_{n} \left[ p_n^{(0)} + p_{n+1}^{(0)} \right] \times \frac{\alpha^-(\Delta_n) \alpha^\dagger_{\sigma \sigma}(\Delta_n) - \alpha^+(\Delta_n) \alpha^-_{\sigma \sigma}(\Delta_n)}{\alpha(\Delta_n)} . \tag{3}
\]

Here \( \Delta_n = E_{\text{ch}}(n + 1) - E_{\text{ch}}(n) \), and \( \alpha_{\sigma \sigma}^\pm(\epsilon) \) are the forward and backward propagators on the Keldysh contour in Fourier space,

\[
\alpha_{\sigma \sigma}^\pm(\epsilon) = \pm i\frac{\epsilon - \Delta \mu_{\sigma \sigma}}{\pi} \exp\left(\pm \beta(\epsilon - \Delta \mu_{\sigma \sigma})\right) - 1 . \tag{4}
\]

The dimensionless conductance of the junction \( r \) for spin \( \sigma \) is \( G^r_{\sigma \sigma} = h / (4\pi^2 e^2 R_{\sigma r}) \). The energy of the tunneling electron, and \( \Delta \mu_{\sigma \sigma} = \mu_r - \mu_{\sigma \sigma} \). Apart from this, \( \alpha^\pm(\epsilon) = \sum_{\sigma} \alpha^\dagger_{\sigma \sigma}(\epsilon) \), \( \alpha(\epsilon) = \alpha^+(\epsilon) + \alpha^-(\epsilon) \), and the probabilities \( p_{n}^{(0)} \) obey the equation \( p_n^{(0)} \alpha^+(\Delta_n) - p_{n+1}^{(0)} \alpha^-(\Delta_n) = 0 \) with \( \sum_{n} p_{n}^{(0)} = 1 \).

The dominant second order (cotunneling) contribution to the electric current can be divided into three parts, \( I_{\sigma r}^{(2)} = \sum_{i=1}^{3} I_{i \sigma r}^{(2)} \), with \( I_{1 \sigma r}^{(2)} = \sum_{i} I_{i \sigma r}^{(2)} = -\sum_{i} I_{i r \sigma}^{(2)} \). These terms are given by the following equations:

\[
I_{1 \sigma r}^{(2)} = \frac{4\pi^2 e}{h} \sum_{n} \left[ p_{n}^{(0)} + p_{n+1}^{(0)} \right] \int d\omega \left[ \alpha^-(\omega) \alpha^\dagger_{\sigma \sigma}(\omega) \right] \Re R_{\sigma r}(\omega)^2 , \tag{5}
\]

\[
I_{2 \sigma r}^{(2)} = -\frac{4\pi^2 e}{2h} \sum_{n} \left[ p_{n}^{(0)} + p_{n+1}^{(0)} \right] \times \frac{\alpha^-(\Delta_n) \alpha^\dagger_{\sigma \sigma}(\Delta_n) - \alpha^+(\Delta_n) \alpha^-_{\sigma \sigma}(\Delta_n)}{\alpha(\Delta_n)} \int d\omega \alpha(\omega) \Re R_{\sigma r}(\omega)^2 + R_{n+1}(\omega)^2 , \tag{6}
\]

\[
I_{3 \sigma r}^{(2)} = -\frac{4\pi^2 e}{2h} \sum_{n} \left[ p_{n}^{(0)} + p_{n+1}^{(0)} \right] \times \frac{\partial}{\partial \Delta_n} \left[ \frac{\alpha^-(\Delta_n) \alpha^\dagger_{\sigma \sigma}(\Delta_n) - \alpha^+(\Delta_n) \alpha^-_{\sigma \sigma}(\Delta_n)}{\alpha(\Delta_n)} \right] \int d\omega \alpha(\omega) \Re R_{\sigma r}(\omega) - R_{n+1}(\omega) , \tag{7}
\]

where \( R_{\sigma r}(\omega) = 1 / (\omega - \Delta_n + i\delta)^2 - 1 / (\omega - \Delta_n - i\delta)^2 \). The terms \( I_{2 \sigma r}^{(2)} \) and \( I_{3 \sigma r}^{(2)} \) describe renormalization of the tunneling conductance and energy gap, respectively, and become important at resonance.

We have numerically evaluated the current for parallel (P) and antiparallel (AP) alignment of the electron magnetizations. We assumed symmetric junctions such that for parallel alignment \( R_{\uparrow \uparrow} = R_{\downarrow \downarrow} = a R_Q \) and \( R_{\downarrow \uparrow} = R_{\uparrow \downarrow} = (1 - P)/(1 + P) R_{\uparrow \uparrow} \), where \( R_Q = h / e^2 \) and we assume \( a = 5 \). The parameter \( P \) denotes the spin polarization of the electrodes - \( P = 0.23 \) and 0.40 for Ni and Fe, respectively. For the antiparallel alignment one then finds \( R_{\uparrow \downarrow} = R_{\downarrow \uparrow} = a R_Q \), \( R_{\downarrow \downarrow} = R_{\uparrow \uparrow} = (1 - P)/(1 + P) R_{\uparrow \uparrow} \).

The spin accumulation on the island (or equivalently spin splitting of the electrochemical potential) is determined from the condition of spin current conservation

\[
\sum_{r} \left( I_{1 \sigma r}^{(1)} + I_{i \sigma r}^{(2)} \right) - e\mu_{\sigma \sigma} D_{i} / \tau_{sf} = 0 . \tag{8}
\]
Here the spin flip processes in the island are taken into account, and characterized by the relaxation time $\tau_{sf}$.

Equation (3) includes the components, which describe single-barrier cotunneling processes which transport spin but do not transport charge. They cancel after summation over the spin orientation and do not enter the current formula. On the other hand, they enter Eq. (2) and, by modifying the magnetic state of the island, have an indirect influence on the transport current.

In Fig. 3(a) we show the differential conductance for Ni and Fe electrodes in both parallel and antiparallel magnetic configurations, calculated for $T/E_C = 0.02$ in the absence of spin-flip processes. The corresponding value of tunnel magnetoresistance, $\text{TMR} \equiv (R_{AP} - R_P)/R_P$ where $R_P$ and $R_{AP}$ are total resistances for parallel and antiparallel configuration respectively, is shown in Fig. 3(a). In the AP configuration the splitting of the conductance peak, resulting from the spin splitting of the electrochemical potential of the island, is well resolved (see Fig. 3(b)). Different splitting for Fe and Ni is due to different magnetic polarizations $P$ of these metals.

The magnitude of the associated spin accumulation is shown in Fig. 3(b). It is interesting to note that no spin accumulation, and consequently no splitting of the resonance, occurs in the $P$ configuration (see Fig. 3(a)).

The cotunneling component of the current shows resonances when the bias voltage $V$ approaches the - effective - electrochemical potential of either spin component. The pure spin splitting of the electrochemical potentials is always smaller than the bias voltage. However, Coulomb blockade effects effectively shift the electrochemical potentials further by the charging energy (as illustrated in the diagram of Fig. 3). Due to this additional shift the resonances in the current should be resolvable. A direct observation of the resonances in the current may still be difficult because even at resonance cotunneling yields only a small fraction of the total current. The resonances are easier to detect in the differential conductance, which is shown in Fig. 3. For the interpretation of these results it should be noted that a resonance in the current corresponds to a maximum negative slope of the conductance.

Such an effect may also arise in the sequential tunneling limit (high resistance junctions), but in that case it is not so evident and pronounced as in the cotunneling limit (see Fig. 3). The role of the cotunneling current is twofold: (i) the cotunneling current has a sharp maximum at resonance (described above), which can be easily detected on the conductance characteristics (see comparison of cotunneling and sequential limit in Fig. 3), and (ii) in the Coulomb blockade regime sequential tunneling is suppressed, so only the cotunneling processes contribute to spin accumulation (as can be concluded from Fig. 3(b) for $\epsilon V/2E_C < 1$).

In Fig. 4 we show the differential conductance versus gate $V_S$ and transport $V$ voltages in a gray-scale plot for both $P$ and AP magnetic configurations. The splitting of the resonance (shown in Fig. 3(b) for $V_S = 0$) is clearly visible in Fig. 4(b). From this figure one can read out the dependence of the electrochemical potential splitting on the transport voltage $V$. Figure 4(b) also shows that the splitting of the conductance peaks can be observed when the gate voltage $V_S$ is varied and the bias voltage $V$ is fixed.

Generally, spin-flip relaxation processes suppress spin accumulation. Figure 4 shows the values of the voltage corresponding to the upper and lower resonances from Fig. 3(b) (maximum negative slopes of the conductance curves) as a function of the intrinsic spin relaxation time $\tau_{sf}$ on the island. Generally, the spin-relaxation time $\tau_{sf}$ depends on sample properties. For instance, in Al one finds $\tau_{sf} \sim 10^{-8}$s. For small islands, $D_1 \approx 10^3/eV$, and for the other parameters as in Fig. 3, one finds $\tau_{sf} R_Q/\hbar (R_L + R_R) D_1 \approx 10^3$. This means that the spin relaxation process is relatively slow and the value of the spin accumulation is close to its maximum value for $\tau_{sf} \rightarrow \infty$. It is known that spin accumulation can be suppressed by a small perpendicular magnetic field (‘Hanle effect’) (4). In principle, this effect can be measured in the system under consideration and may be used to determine the spin relaxation time as well.

There are several experimental works (5) on ferromagnetic SETs both in the sequential and cotunneling regime with all electrodes (including island) ferromagnetic. The corresponding data, as well as the data obtained on ferromagnetic granular systems suggest that spin accumulation may occur in such systems, but there was no clear experimental evidence on this. But the range of the parameters indicates a possibility of a direct detection by the method proposed in this paper.

In conclusion, we have analyzed electron tunneling in ferromagnetic SETs in the cotunneling regime. We showed that the spin accumulation on the island can be observed directly in the current-voltage characteristics as a splitting of the resonance. The distance between two resonances arises due to the spin-splitting of the electrochemical potential due to spin accumulation. This effect should be observable experimentally, and could give clear evidence of the spin accumulation, overcoming the objections to the previous measurements (6).

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FIG. 1. Differential conductance vs. bias voltage $V$ in the (a) parallel and (b) antiparallel configurations, calculated for $T/E_C = 0.02$ and symmetric junctions with $R_{\uparrow\downarrow} = 5R_Q$. The spin polarization is $P = 0.23$ and 0.40 for Ni and Fe electrodes, respectively. Dotted line is for the sequential tunneling limit for Fe electrodes.

FIG. 2. (a) Tunnel magnetoresistance TMR and (b) spin accumulation for Ni and Fe electrodes as functions of bias voltage $V$. The other parameters are the same as in Fig. 1. Dotted line is for the sequential tunneling limit for Fe electrodes.
FIG. 3. Energy diagrams for a symmetric magnetic double tunnel junction with a normal metallic island for the antiparallel configuration (a) without and (b) with the Coulomb blockade. Here $\mu_L$, $\mu_R$ are the electrochemical potentials for the left and right electrodes and $\mu_{\uparrow}$, $\mu_{\downarrow}$ are the electrochemical potentials for spin-up and spin-down electrons on the island. $N = 1.0$ denotes the island charge.

FIG. 4. The differential conductance vs. gate $V_g$ and transport $V$ voltages in a gray-scale representation in the (a) parallel and (b) antiparallel configurations calculated for Fe electrodes and the other parameters as in Fig. 3.

FIG. 5. Dependence of the upper and lower resonance voltages on the intrinsic spin relaxation time $\tau_{sf}$, calculated for Fe electrodes and the other parameters as in Fig. 3. The dotted line denotes a regime of insufficiently resolved resonances.