Spin effects in single-electron tunneling in magnetic junctions

J. Martinek¹, J. Barnas²,³, G. Michałek¹, B.R. Bulka¹ and A. Fert³

¹ Institute of Molecular Physics, Polish Academy of Sciences, ul. Smoluchowskiego 17, 60-179 Poznań, Poland
² Department of Physics, A. Mickiewicz University, ul. Umultowska 85, 61-614 Poznań, Poland
³ Unite Mixte de Physique CNRS/Thomson, Domaine de Corbeville, 91-404 Orsay, France

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Spin dependent single electron tunneling in ferromagnetic double junctions is analysed theoretically in the limit of sequential tunneling. The influence of discrete energy spectrum of the central electrode (island) on the spin accumulation, spin fluctuations and tunnel magnetoresistance is analysed numerically in the case of a nonmagnetic island. It is shown that spin fluctuations are significant in magnetic as well as in nonmagnetic junctions.
Single electron tunneling in mesoscopic double-junctions has been recently extensively studied both experimentally and theoretically.\textsuperscript{1--3} For a small capacitance $C$ of the central electrode (island) the charging energy $E_c = e^2/2C$ can be larger than the thermal energy $k_B T$. Discrete charging of the island leads then to Coulomb blockade of the electric current at voltages below a certain threshold voltage, and to a characteristic 'Coulomb staircase' at higher voltages. When the number of electrons on the island is not large, the energy quantization may become important as well, and can lead to additional steps in the $I - V$ curves (or additional peaks in the $dI/dV$ characteristics).\textsuperscript{4,5}

The interplay of ferromagnetism and discrete charging was studied only very recently.\textsuperscript{6--11} Theoretical results show that discrete charging leads to oscillations in tunnel magnetoresistance (TMR) (that is in the change of the total junction resistance when relative orientation of the magnetization of both electrodes and of the island is varied).\textsuperscript{9,11} In Ref.\textsuperscript{[9]} the intrinsic spin relaxation time on the island was assumed to be sufficiently short (shorter than the time between two successive tunneling events) to neglect spin accumulation. This model has then been extended to describe the regime where intrinsic spin relaxation time on the island is larger than the time between successive tunneling events. In this regime spin accumulation has to be taken into account, which leads to enhanced TMR and several new phenomena, like inverse TMR or negative differential resistance. Spin accumulation can also generate TMR when the island is nonmagnetic.

In the model of Refs.\textsuperscript{[9,11]} the discrete nature of the density of states on
the island and also the fluctuations of the spin accumulation were neglected. These restrictions are relaxed in the present paper, where spin fluctuations as well as discrete structure of the density of single-electron states are taken into account. On the other hand, as in the models refered to above, we restrict our considerations to the limit of sequential tunneling (where the orthodox tunneling theory is applicable), that is to the limit when the resistance of each single junction is larger than the quantum resistance \( R_q, R_q = h/e^2 \). In that limit higher order processes (cotunneling) can be neglected. However, these processes may play an important role in the blockade regime.\(^{10}\)

Geometry of the junction and energy structure is shown schematically in Fig.1. In a general case both electrodes and the island can be ferromagnetic. We consider the case when the resistances of both left and right junctions are much larger than the quantum resistance \( R_q \). Moreover, we assume that \( k_B T >> \Gamma \), where \( \Gamma \) denotes width of the discrete energy levels on the island.

To calculate the electric current \( I \) which flows through the junction when a voltage \( V \) is applied between the source (right) and drain (left) electrodes, one can then use the orthodox tunneling theory.\(^{4,5,12−14}\)

Assume that there are \( N^* \) excess electrons on the island. Generally, one can write \( N^* = N^*_\uparrow + N^*_\downarrow \), where \( N^*_\sigma (\sigma = \uparrow \text{ or } \sigma = \downarrow) \) is the number of excess electrons of a given spin orientation. The excess spin polarization of the island is then equal to \( N^*_\uparrow - N^*_\downarrow \). If \( N_\sigma \) is the number of electrons with spin \( \sigma \) on the island when a voltage \( V \) is applied, and \( N^0_\sigma \) is this number at \( V = 0 \), then one can write \( N^*_\sigma = N_\sigma - N^0_\sigma \).
Denoting by $E_{i\sigma}$ the discrete energy levels of the island, one can express the electric current $I$ in the stationary state as

$$I = e \sum_{\sigma} \sum_i \sum_{\{n\}} |T_{i\sigma}|^2 P(\{n\})$$

$$\times \left\{ \delta_{n_{i\sigma},1}[1 - f(E_{i\sigma} + E_{lN}^- - E_F)] - \delta_{n_{i\sigma},0} f(E_{i\sigma} + E_{lN}^+ - E_F) \right\} . \quad (1)$$

In the above equation $P(\{n\})$ is the probability of a particular configuration of the occupation numbers of discrete energy levels of the island, $\{n\} \equiv \{n_{1\uparrow}, n_{2\uparrow}, \ldots, n_{1\downarrow}, n_{2\downarrow}, \ldots\}$, with $n_{i\sigma} = 1$ ($n_{i\sigma} = 0$) for occupied (empty) states and the sum over $\{n\}$ denotes the summation over all possible occupation configurations. This probability can be determined from an appropriate master equation.\(^{4,5}\) Apart from this, $T_{i\sigma}^l$ in Eq.(1) is the matrix element corresponding to the electron tunneling to (from) the left electrode from (to) the level $E_{i\sigma}$ of the island, while $E_{lN}^{\pm}$ is defined as $E_{lN}^{\mp} = e V_{lN}^l, \pm E_c$. Here, $V_{lN}^l = (C_r/C) V + N^* e/C$, where $C_r$ is the capacitance of the right junction, $C$ is the total capacitance of the island, and $-e$ is the electron charge ($e > 0$). Finally, we assumed in Eq.(1) the equilibrium Fermi-Dirac distribution of electrons in the electrodes, with the Fermi level $E_F$ at vanishing voltage. In the following, we will also introduce the quantity $E_{N^*}^{\mp} = e V_{N^*}^r, \mp E_c$, where $V_{N^*}^r = (C_l/C) V - N^* e/C$ and $C_l$ is the capacitance of the left junction ($C = C_l + C_r$), and the matrix element $T_{i\sigma}^r$ for tunneling through the right junction.

It is convenient to introduce the probability $P(N_{\uparrow}, N_{\downarrow})$ of finding on the
island $N_{\uparrow}$ electrons with spin $\sigma = \uparrow$ and $N_{\downarrow}$ electrons with spin $\sigma = \downarrow$,

$$P(N_{\uparrow}, N_{\downarrow}) = \sum_{\{n\}} P(\{n\})\delta_{N_{\uparrow}, \sum_n n_{\uparrow}} \delta_{N_{\downarrow}, \sum_n n_{\downarrow}}, \quad (2)$$

and the distribution function

$$F(E_{i\sigma}|N_{\uparrow}, N_{\downarrow}) = \frac{1}{P(N_{\uparrow}, N_{\downarrow})} \sum_{\{n\}} P(\{n\})\delta_{n_{\uparrow}, 1} \delta_{N_{\uparrow}, \sum_n n_{\uparrow}} \delta_{N_{\downarrow}, \sum_n n_{\downarrow}}. \quad (3)$$

Equation (3) describes the probability that the level $E_{i\sigma}$ is occupied under the condition that the island contains $N_{\uparrow}$ electrons with spin $\sigma = \uparrow$ and $N_{\downarrow}$ electrons with spin $\sigma = \downarrow$. The current $I$ is then given by

$$I = -e \sum_{N_{\uparrow}} \sum_{N_{\downarrow}} \sum_{\sigma} \sum_{i} P(N_{\uparrow}, N_{\downarrow}) \left\{ [1 - F(E_{i\sigma}|N_{\uparrow}, N_{\downarrow})]|T_{i\sigma}|^2 f(E_{i\sigma} + E_{N_{\uparrow}}^{F} - E_{F}) - F(E_{i\sigma}|N_{\uparrow}, N_{\downarrow})|T_{i\sigma}|^2 [1 - f(E_{i\sigma} + E_{N_{\downarrow}}^{F} - E_{F})] \right\}. \quad (4)$$

In the stationary state one finds the following master equation for the probability $P(N_{\uparrow}, N_{\downarrow})$:

$$\frac{\partial}{\partial t} P(N_{\uparrow}, N_{\downarrow}) = 0 = -P(N_{\uparrow}, N_{\downarrow})A(N_{\uparrow}, N_{\downarrow})$$

$$+ P(N_{\uparrow} + 1, N_{\downarrow})B_{\uparrow}(N_{\uparrow} + 1, N_{\downarrow}) + P(N_{\uparrow}, N_{\downarrow} + 1)B_{\downarrow}(N_{\uparrow}, N_{\downarrow} + 1)$$

$$+ P(N_{\uparrow} - 1, N_{\downarrow})C_{\uparrow}(N_{\uparrow} - 1, N_{\downarrow}) + P(N_{\uparrow}, N_{\downarrow} - 1)C_{\downarrow}(N_{\uparrow}, N_{\downarrow} - 1) + \mathcal{R}_s, \quad (5)$$

where

$$A(N_{\uparrow}, N_{\downarrow}) = \sum_{\sigma} \sum_i \left[ 1 - F(E_{i\sigma}|N_{\uparrow}, N_{\downarrow}) \right] |T_{i\sigma}|^2 f(E_{i\sigma} + E_{N_{\uparrow}}^{F} - E_{F})$$

$$+ |T_{i\sigma}|^2 f(E_{i\sigma} - E_{N_{\downarrow}}^{F} - E_{F}) + \sum_{\sigma} \sum_i F(E_{i\sigma}|N_{\uparrow}, N_{\downarrow}) |T_{i\sigma}|^2 [1 - f(E_{i\sigma} + E_{N_{\downarrow}}^{F} - E_{F})]$$
In Eq. (5) \( R_s \) stands for terms responsible for magnetic relaxation on the island. Note that spin-conserving relaxation processes are included in Eq. (5) only through the distribution function \( F(E_{i\sigma}|N_\uparrow N_\downarrow) \).

We assume in the following that the energy relaxation time due to spin-conserving relaxation processes is significantly shorter than the time between successive tunneling events, and also shorter than the spin relaxation time. This allows us to use an equilibrium form \( F_{eq}(E_{i\sigma}|N_\uparrow N_\downarrow) \) of the distribution function \( F(E_{i\sigma}|N_\uparrow N_\downarrow) \), which depends on the spin orientation. In a general case the equilibrium distribution \( F_{eq}(E_{i\sigma}|N_\uparrow N_\downarrow) \) can be found from the Gibbs distribution. In the limit \( k_B T \gg \Delta E \) the distribution function \( F_{eq}(E_{i\sigma}|N_\uparrow N_\downarrow) \) is equal to by the Fermi-Dirac distribution

\[
F(E_{i\sigma}|N_\uparrow N_\downarrow) = f(E_{i\sigma} - \mu_{\sigma}(N_{\sigma})) ,
\]

where the chemical potential \( \mu_{\sigma}(N_{\sigma}) \) is determined by the equation

\[
\sum_i f(E_{i\sigma} - \mu_{\sigma}(N_{\sigma})) = N_{\sigma} .
\]

As noticed by Beenakker [5], the equilibrium distribution for \( k_B T \approx \Delta E \) differs significantly from the Fermi distribution.
Consider now some numerical results. For simplicity, we will restrict the following considerations to the situation when the island is nonmagnetic and the magnetizations of the electrodes can be either parallel or antiparallel. To emphasize the role of spin accumulation we assume that the intrinsic spin relaxation time on the island is long enough to neglect all intrinsic spin-flip processes. The magnetic relaxation on the island takes then place only through tunneling processes. For simplicity, we assume that the energy levels on the island are equidistinct, with the inter-level spacing $\Delta E$. Apart from this, we assume that $k_B T < \Delta E$ (but still $k_B T >> \Gamma$) and $\Delta E < E_c$. In Fig.2a we show the I-V characteristics for parallel and antiparallel configurations. In both cases the electric current is blocked below a threshold voltage equal approximately to 13 mV. Above the threshold voltage typical ‘Coulomb staircase’ appears with additional small steps due to discrete levels. The existence of those steps leads to additional peaks in the derivative $dI/dV$, as shown in Fig.3b for the antiparallel configuration (the corresponding curve for the parallel configuration was not shown there for clarity).

The difference between I-V curves for the parallel and antiparallel configurations is due to a different spin accumulation in both geometries (note, that both I-V curves become identical when no spin accumulation occurs in the nonmagnetic island\(^9\)). To show correlations between the I-V curves and spin accumulation, we present in Fig.2c the average value of the difference between the numbers of spin-up and spin-down excess electrons on the island, $\langle N^*_+ - N^*_\downarrow \rangle$, for both antiparallel and parallel configurations (up to a
constant factor, Fig.2c shows an average magnetic moment induced on the island due to spin accumulation). For the symmetrical structure assumed here ($R_{l\uparrow}/R_{l\downarrow} = R_{r\uparrow}/R_{r\downarrow}$ for the parallel configuration, where $R_{l(r)\sigma}$ are the junction resistances for a given spin channel) there is no significant spin accumulation in the parallel configuration. Only small nonzero values occur around the Coulomb steps. For asymmetrical junctions spin accumulation also occurs in the parallel configuration. Discrete structure of the density of states on the island appears in the voltage dependence of $\langle N_\uparrow^* - N_\downarrow^* \rangle$ as the fine steps clearly visible in Fig.2c.

The number $N_\uparrow^* - N_\downarrow^*$ of spins accumulated on the island fluctuates around its average value $\langle N_\uparrow^* - N_\downarrow^* \rangle$, as shown in Fig.2d, where the standard deviation $[(N_\uparrow^* - N_\downarrow^*)^2 - \langle N_\uparrow^* - N_\downarrow^* \rangle^2]^{1/2}$ is plotted against the voltage $V$. It is worth to note that although there is almost no spin accumulation in the parallel configuration, the corresponding fluctuations are relatively large and comparable with the fluctuations in the antiparallel configuration. The fine structure in the voltage dependence of the standard deviation originates from the discrete electronic structure of the island.

As we have already pointed above, spin accumulation on the island gives rise to a difference between the I-V curves in the parallel and antiparallel configurations. This difference, in turn, leads to the tunnel magnetoresistance (TMR) defined quantitatively as $(R_{ap} - R_p)/R_p$, where $R_{ap}$ and $R_p$ are the total junction resistances respectively in the antiparallel and parallel configurations. The ratio $(R_{ap} - R_p)/R_p$ is shown in Fig.2e, where the broad
peaks correspond to the Coulomb steps while the fine structure originates from the discrete structure of the density of states.

In the limit of a nonmagnetic junction (nonmagnetic island and nonmagnetic electrodes) there is no spin accumulation on the island and no TMR. However, spin fluctuations still occur as shown in Fig.3 for the limit of no intrinsic spin relaxation on the island. As in Fig.2, the discrete energy levels on the island lead to fine steps in the voltage dependence of the standard deviation.

In summary, we developed in this letter a formalism for calculating electric current and tunnel magnetoresistance in ferromagnetic double junctions, which takes into account spin accumulation on the island and discrete structure of the density of states. The discrete energy levels lead to fine steps in the $I - V$ curves and to additional peaks in the first derivative $dI/dV$. Moreover, we showed that the discrete levels give rise to fine structure in the spin accumulation and spin fluctuations on the island, as well as in TMR. To single out the charging effects we neglected in this letter tunneling processes with simultaneous spin flip, which lower TMR and also lead to a smooth decrease of TMR with increasing voltage.$^{15,16}$

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Fig. 1 Schematic of the double-junction considered in this paper. Magnetization of the island and of the right electrode can be either parallel or antiparallel to the magnetization of the left electrode, as indicated by the solid and open arrows. Discrete levels of the island are marked schematically by the solid (dashed) lines for spin-up (spin-down) electron states.
Fig. 2 Voltage dependence of the tunnel current $I$ (a), first derivative $dI/dV$ (b), spin accumulation $\langle N_\uparrow^* - N_\downarrow^* \rangle$ (c), standard deviation $[(\langle N_\uparrow^* - N_\downarrow^* \rangle^2 - \langle N_\uparrow^* - N_\downarrow^* \rangle^2)^{1/2}$ (d), and tunnel magnetoresistance TMR (e), calculated at $T = 2.3K$. The solid and dotted curves in (a), (c) and (d) correspond to the antiparallel and parallel configurations, respectively. The other parameters assumed in the numerical calculations are: $\Delta E = 3\text{meV}, C_1 = 6.6a\text{F}, C_2 = 1.32a\text{F}$ (which gives $E_c = 10.1\text{meV}$), $R_{\uparrow\uparrow} = 200\text{M}\Omega, R_{\downarrow\downarrow} = 100\text{M}\Omega, R_{\uparrow\downarrow} = 4\text{M}\Omega$ and $R_{\uparrow\uparrow} = 2\text{M}\Omega$ for the parallel configuration ($R_{\downarrow\uparrow} = 2\text{M}\Omega, R_{\uparrow\uparrow} = 4\text{M}\Omega$ for the antiparallel configuration).
Fig.3 Fluctuations of the spin accumulated on the island in the limit of a non-magnetic double junction. Standard deviation \[\langle (N_{i}^{\uparrow} - N_{i}^{\downarrow})^2 \rangle^{1/2} - \langle N_{i}^{\uparrow} - N_{i}^{\downarrow} \rangle^2 \] is shown as a function of the voltage \(V\) for the parameters: \(T = 2.3K, \Delta E = 3\text{meV}, C_1 = 6.6\text{aF}, C_2 = 1.32\text{aF} (E_c = 10.1\text{meV}), R_{\uparrow\uparrow} = R_{\downarrow\downarrow} = 100\text{M}\Omega,\) and \(R_{\uparrow\downarrow} = R_{\downarrow\uparrow} = 4\text{M}\Omega.\)