Negative differential conductance and magnetoresistance oscillations due to spin accumulation in ferromagnetic double-island devices

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Spin-dependent electronic transport in magnetic double-island devices is considered, theoretically, in the sequential tunneling regime. Electric current and tunnel magnetoresistance are analyzed as a function of the bias voltage and spin-relaxation time in the islands. It is shown that the interplay of spin accumulation on the islands and charging effects leads to periodic modification of the differential conductance and tunnel magnetoresistance. For a sufficiently long spin-relaxation time, the modulations are associated with periodic oscillations of the sign of both the tunnel magnetoresistance and differential conductance.

DOI: 10.1103/PhysRevB.73.033409

PACS number(s): 72.25.-b, 72.20.My, 73.23.Hk, 85.75.-d

I. INTRODUCTION

Spin-dependent transport in single-electron devices is currently attracting much attention from both fundamental and application points of view.^{1–8} It has been shown that the interplay of single-electron charging effects and spin dependence of tunneling processes (caused by ferromagnetism of external and/or central electrodes) in single-electron transistors gives rise to interesting phenomena, such as periodic modulation of the tunnel magnetoresistance (TMR) with increasing either bias or gate voltages, quasi-oscillatory behavior of the spin accumulation on the central electrode (referred to as an island in the following), enhancement of TMR in the Coulomb blockade regime, and others. Some of the theoretically predicted phenomena have already been confirmed experimentally.

Recently, transport characteristics of granular magnetic nanobridges connecting two external magnetic and/or nonmagnetic electrodes were investigated experimentally.9-11 The nanobridges consisted of magnetic grains distributed in a nonconducting matrix. The corresponding current-voltage (I-V) curves revealed characteristic steps due to discrete charging of the grains with single electrons (Coulomb steps). Apart from this, oscillations of the TMR effect (associated with the transition from one magnetic configuration to the other) with the bias voltage have also been observed.¹¹ Moreover, two additional features of the transport characteristics were found whose physical mechanism and origin needs further considerations. First, the differential conductance was found to change sign periodically with increasing bias voltage.9,10 Second, the TMR was shown to oscillate between positive and negative values with increasing bias voltage. Physical origin of these oscillations, however, remained unclear, although the role of spin accumulation was invoked following the results obtained for a single-island system.²

For small lateral and vertical dimensions of the nanobridges, electronic transport between the external leads occurs as a result of consecutive electron jumps via metallic grains, and the corresponding electronic paths can include either one, two, or more grains. Transport through a single grain and the associated TMR have already been analyzed,² but the results cannot describe the above-mentioned data obtained on magnetic multigrain nanobridges. However, the experimental setup described above can be modeled by assuming only two grains involved in the transport processes.⁹ Although the Coulomb steps have been accounted for by the existing theoretical models based on discrete charging with single electrons, the origin of negative differential conductance (NDC) as well as the oscillations in TMR with alternating sign remains unclear. Therefore, this problem is considered in this Brief Report in more details.

Following the above discussion, we consider transport through a double-island device consisting of two metallic grains attached to two external leads. The two islands are separated from each other and also from the external leads by tunnel barriers. In a general case, all the four metallic components of the device (two external electrodes and two islands) can be ferromagnetic,^{1,5} but in the following we will consider the situation with one external lead being nonmagnetic and the other three components being ferromagnetic. We also restrict our considerations to collinear [parallel (P) and antiparallel (AP)] alignments of the magnetic moments, as indicated in the inset of Fig. 1(d).

The key role in our analysis is played by the nonequilibrium spin accumulation, which occurs for a sufficiently long spin-relaxation time on the islands. Strictly speaking, the accumulation may take place when the spin-relaxation time is significantly longer than the time between successive tunneling processes. Assuming this is the case, we show that the interplay of charging effects and spin accumulation gives rise to both effects described above (i.e., to the NDC and periodic change of the sign of TMR with increasing bias voltage). This behavior of transport characteristics accounts for the experimental observations. We also show that the presence of NDC depends on the magnetic configuration of the device, and by switching from one configuration to the other, one may change sign of the differential conductance (e.g., by an external magnetic field). This behavior may be of some interest for possible future applications in spintronics and/or magnetoelectronics devices.

II. MODEL

The system adapted to model the phenomena discussed above consists of the left and right leads and two central islands, separated from each other and from the leads by tunnel barriers, as shown schematically in the inset of Fig. 1(d). The bias voltage is applied to the external leads: $V_{\rm L}$ to the left one and $V_{\rm R}$ to the right one. In a general case, one can capacitively attach gate voltages to both islands. However, to model the experimental results described above, we neglect the gate voltages in the following. We assume that the size of each island is sufficiently small to have the charging energies of the islands significantly larger than the thermal energy $k_{\rm B}T$, but still large enough to neglect size quantization. Apart from this, we assume that the barrier resistances are much larger than the quantum resistance, R_r $\gg h/e^2$ (r=L,M,R). The system is then in a well-defined charge state described by n_1 and n_2 excess electrons on the first (left) and second (right) islands, respectively, and the electrostatic energy of the system is given by the formula^{12,13}

$$E(n_{1}, n_{2}) = E_{C_{1}} \left(n_{1} - \frac{C_{L}V_{L}}{e} \right)^{2} + E_{C_{2}} \left(n_{2} - \frac{C_{R}V_{R}}{e} \right)^{2} + 2E_{C_{M}} \left(n_{1} - \frac{C_{L}V_{L}}{e} \right) \left(n_{2} - \frac{C_{R}V_{R}}{e} \right), \quad (1)$$

where $C_{L(R)}$ is the capacitance of the left (right) junction, E_{C_1} and E_{C_2} denote the charging energies of the two islands, $E_{C_1(C_2)} = e^2 / (2C_{1(2)}) [1 - C_M^2 / (C_1 C_2)]^{-1}$, and E_{C_M} is the energy of electrostatic coupling between the islands, E_{C_M} $=e^{2}/(2C_{\rm M})(C_{1}C_{2}/C_{\rm M}^{2}-1)^{-1}$, with $C_{1(2)}$ being the total capacitance of the first (second) island, $C_{1(2)} = C_{L(R)} + C_M$, and $C_{\rm M}$ denoting the capacitance of the middle junction (the one between the islands). This allows us to employ the quasiclassical theory based on the master equation and the Fermi golden rule for the tunneling rates.¹⁴ Such an approach describes well transport in the sequential tunneling regime and corresponds to taking into account only the first-order tunneling processes, which are exponentially suppressed in the Coulomb blockade regime but give the dominant contribution to electric current when the applied voltage exceeds a certain threshold. Moreover, we take into account only nonspin-flip (spin-conserving) tunneling processes through the barriers.

When a bias voltage $V=V_L-V_R$ is applied to the system and spin-relaxation time on the islands is sufficiently long, a nonequilibrium magnetic moment can accumulate on each island. Let us denote the corresponding shift of the Fermi level for electrons with spin- σ on the *j*th island by ΔE_{Fj}^{σ} . When, in the initial state, there were n_1 and n_2 excess electrons on the islands, the spin-dependent tunneling rate from the left electrode to the first island is then given by

$$\Gamma_{L1}^{\sigma}(n_1, n_2) = \frac{1}{e^2 R_L^{\sigma}} \frac{\Delta E_{L1}^{\sigma}(n_1, n_2)}{\exp\left[\frac{\Delta E_{L1}^{\sigma}(n_1, n_2)}{k_B T}\right] - 1},$$
 (2)

where $R_{\rm L}^{\sigma}$ is the spin-dependent resistance of the left barrier and $\Delta E_{\rm L1}^{\sigma}(n_1, n_2)$ describes the change in the electrostatic energy of the system caused by the respective tunneling event, $\Delta E_{L1}^{\sigma}(n_1, n_2) = E(n_1 + 1, n_2) - E(n_1, n_2) + eV_L + \Delta E_{F1}^{\sigma}$. (According to our notation, the electron charge is -e with e > 0.)

For the following discussion, it is convenient to distinguish between spin projection on the global quantization axis $(\sigma=\uparrow,\downarrow)$ and on the local quantization axis $(\sigma=+,-, \text{ with } \sigma=+ \text{ and } \sigma=- \text{ corresponding to spin-majority and spin-minority electrons, respectively). The spin asymmetry in tunneling processes follows from spin-dependent density of states and spin-dependent tunneling matrix elements. Let us define the parameters <math>\beta_r = D_r^+/D_r^-$ for the leads (r=L,R) and $\beta_j = D_{Ij}^+/D_{Ij}^-$ for the islands (j=1,2), where $D_r^{+(-)}$ and $D_{Ij}^{+(-)}$ are the appropriate densities of states for spin-majority (spin-minority) electrons in the leads (r=L,R) and islands (j=1,2), respectively. Thus, the Fermi-level shifts due to spin accumulation on the islands obey the condition $\Delta E_{Fj}^+/\Delta E_{Fj}^- = -\beta_j$ for the first (j=1) and second (j=2) islands.

The spin asymmetry of the barrier resistances can be described by the parameters $\alpha_r = R_r^{\uparrow}/R_r^{\downarrow}$ for r=L,M,R. Assuming a constant (independent of energy) density of states and constant (independent of energy and spin) matrix elements, one can write $\alpha_r = D_r^{\downarrow} D_{Ij}^{\downarrow} / D_r^{\uparrow} D_{Ij}^{\uparrow}$ for the left (r=L, j=1) and right (r=R, j=2) barriers, and $\alpha_r = D_{I1}^{\downarrow} D_{I2}^{\uparrow} / D_{I1}^{\uparrow} D_{I2}^{\uparrow}$ for the central (r=M) barrier. When the local and global spin quantization axes coincide (parallel configuration), then one can write $\alpha_r = 1/(\beta_r \beta_j)$ for the left (r=L, j=1) and right (r=R, j=2) barriers, and $\alpha_r = 1/(\beta_1 \beta_2)$ for the central (r=M) barrier. The above formulas are also applicable to the situation with magnetic moment of a lead or an island reversed (antiparallel configuration), but with the corresponding β replaced by $1/\beta$.

The probability $P(n_1, n_2)$ that the system is in a charge state (n_1, n_2) can be determined in a recursive way from the appropriate steady-state master equation.¹³ The electric current flowing through the left junction is then given by

$$I_{\rm L} = -e \sum_{\sigma} \sum_{n_1, n_2} \left[\Gamma_{\rm L1}^{\sigma}(n_1, n_2) - \Gamma_{\rm 1L}^{\sigma}(n_1, n_2) \right] P(n_1, n_2).$$
(3)

The associated shifts of the Fermi level can be calculated in a self-consistent way from the relations¹⁵

$$\frac{1}{e}(I^{\sigma}_{\rm M(R)} - I^{\sigma}_{\rm L(M)}) - \frac{D^{\sigma}_{\rm I1(2)}\Omega_{\rm I1(2)}}{\tau_{\rm sf,1(2)}}\Delta E^{\sigma}_{\rm F1(2)} = 0, \qquad (4)$$

where Ω_{Ij} is the volume of the island *j*, $\tau_{sf,j}$ denotes the spin-flip relaxation time in the *j*th island, and I_r^{σ} is the current flowing through the barrier *r* (*r*=L,M,R) in the spin channel σ .

III. RESULTS AND DISCUSSION

In Fig. 1, we show transport characteristics of a device whose two islands as well as the right electrode are made of the same ferromagnetic metal, whereas the left electrode is nonmagnetic, as shown in the inset of Fig. 1(d), where also both parallel and antiparallel configurations are defined. This system geometry corresponds to the situation studied experimentally in Ref. 9. The transport characteristics have been



FIG. 1. Shifts of the Fermi levels for spin-majority electrons (a) and currents (b) in the P and AP configurations; differential conductance in the AP configuration (c); and TMR (d) as a function of the bias voltage for $\tau_{sf,1} = \tau_{sf,2} \rightarrow \infty$. The other parameters are: T = 140 K, $C_L = 0.45 \text{ aF}$, $C_M = 0.2 \text{ aF}$, $C_R = 0.35 \text{ aF}$, $\beta_1 = \beta_2 = \beta_R = 0.2$, and $\beta_L = 1$. The total barrier resistances in the P configuration, $R_{r,\uparrow}^{AP} = R_{r,\downarrow}^{AP} = (R_{r,\uparrow}^P R_{r,\downarrow}^P)^{1/2}$, for r = M, R. The parameters correspond to the experimental ones taken from Ref. 9.

obtained for the limit of long spin-relaxation time. The current-induced shifts of the Fermi level (due to spin accumulation) for the spin-majority electrons in both islands are shown in Fig. 1(a) for the P and AP configurations and for positive bias voltage (electrons flow from right to left). A nonzero spin accumulation occurs in both magnetic configurations. In the P configuration, the shift of the Fermi level for spin-majority electrons is negative for both islands, whereas in the antiparallel configuration it is positive for the first island and negative for the second one. Such a behavior can be accounted for by taking into account spin asymmetries of the tunneling processes through all the three barriers, similarly as it was done in the case of a double-barrier system.²

The Coulomb steps in the P and AP configurations are significantly different [see Fig. 1(b)]. There are two reasons for this difference. First, the overall resistance of a given barrier depends on the relative orientation of the magnetic moments of adjacent ferromagnetic components of the device. Second, the spin accumulations in the P and AP configurations are also different. The latter fact is of particular importance for the present analysis.

The differential conductance corresponding to the I-V curves from Fig. 1(b) is shown in Fig. 1(c). In both P and AP configurations, the differential conductance changes sign periodically with increasing bias. However, the bias voltage range of NDC for the P configuration is different from that for the AP one. The corresponding phase difference varies with the bias voltage. Furthermore, NDC is more pronounced in the AP configuration and its absolute magnitude increases with increasing bias voltage, as shown in Fig. 1(c). This behavior of NDC is consistent with experimental observation reported in Ref. 9.

The TMR effect associated with the transition from antiparallel to parallel configurations is visualized in Fig. 1(d). The TMR is periodically modulated with increasing bias voltage. The initial phase of the oscillations depends on the system parameters and can change from negative to positive. Moreover, the modulations are associated with a periodic change of TMR between positive and negative values.

Oscillations of the sign of TMR and differential conductance result from spin accumulations in both islands and are absent in the limit of fast spin-relaxation (no spin accumulation) on the islands. The results presented in Fig. 1 have been calculated for the long spin-relaxation limit. In such a limit, some spin accumulation may occur even for a very small current flowing through the system, giving rise to NDC and oscillations of the TMR sign for small bias voltages. However, both effects disappear when the spin-relaxation time is shorter than the time between successive tunneling events. Thus, for a finite relaxation time, one may expect no NDC and no TMR oscillations for small voltages and an onset of these effects at larger voltages. This is because at some voltage there is a crossover from the fast to slow spin-relaxation limits. In fact, such a behavior is consistent with experimental data presented in Ref. 9.

The disappearance of NDC with decreasing spinrelaxation time τ_{sf} is shown explicitly in Fig. 2, where the bias dependence of differential conductance is presented for different values of the spin-relaxation time. This figure clearly shows that NDC disappears when spin-relaxation time decreases, in agreement with the above discussion. In the limit of fast spin-relaxation time, the differential conductance is positive, although its periodic modulation still remains.

Similarly, periodic oscillations of the sign of TMR also disappear with decreasing spin-relaxation time τ_{sf} . This behavior is shown in Fig. 3, where the bias dependence of TMR is shown for several values of τ_{sf} . First, the transitions to negative TMR disappear with decreasing τ_{sf} . The TMR



FIG. 2. (Color online) Differential conductance in the antiparallel configuration as a function of the bias voltage calculated for different values of the spin relaxation time $\tau_{sf,1} = \tau_{sf,2} = \tau_{sf}$ and for $D_{11}^+\Omega_{11} = D_{12}^+\Omega_{12} = 1000/\text{eV}$. The other parameters are the same as in Fig. 1.

becomes then positive, although some periodic modulations survive. Second, the phase of the modulations shifts by about π when the spin-relaxation varies from fast to slow limits.

IV. CONCLUSION

We have analyzed transport through a double-island device and shown that the negative differential conductance measured experimentally is due to nonequilibrium spin accumulation in the islands. Furthermore, spin accumulation may also lead to oscillations in TMR between negative and posi-



FIG. 3. (Color online) The TMR effect as a function of the bias voltage calculated for different values of spin relaxation time $\tau_{\rm sf,1} = \tau_{\rm sf,2} = \tau_{\rm sf}$ and for $D_{\rm I1}^+ \Omega_{\rm I1} = D_{\rm I2}^+ \Omega_{\rm I2} = 1000/\rm eV$. The other parameters are the same as in Fig. 1.

tive values. The effect of NDC occurs in both configurations, and transition from one configuration to the other may result in transition from positive to negative differential conductance, which may be of some importance for applications in spintronics devices.

ACKNOWLEDGMENTS

The work was supported by the Polish State Committee for Scientific Research through the Projects No. PBZ/KBN/ 044/P03/2001 and No. 2 P03B 116 25, and by EU through RTNNANO Contract No. MRTN-CT-2003-504574.

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- ¹K. Ono, H. Shimada, S. Kobayashi, and Y. Ootuka, J. Photogr. Sci. 65, 3449 (1996).
- ²J. Barnaś and A. Fert, Phys. Rev. Lett. **80**, 1058 (1998); Europhys. Lett. **24**, 85 (1998).
- ³S. Takahashi and S. Maekawa, Phys. Rev. Lett. 80, 1758 (1998).
- ⁴H. Imamura, S. Takahashi, and S. Maekawa, Phys. Rev. B **59**, 6017 (1999).
- ⁵H. Shimada and Y. Ootuka, Phys. Rev. B **64**, 235418 (2002).
- ⁶I. Weymann and J. Barnaś, Phys. Status Solidi B 236, 651 (2003).
- ⁷F. Ernult, K. Yamane, S. Mitani, K. Yakushiji, K. Takanashi, Y. K. Takahashi, and K. Hono, Appl. Phys. Lett. 84, 3106 (2004).
- ⁸K. Yakushiji, F. Ernult, H. Imamura, K. Yamane, S. Mitani, K. Takanashi, S. Takahashi, S. Maekawa, and H. Fujimori, Nat. Mater. 4, 57 (2005).

- ⁹H. Imamura, J. Chiba, S. Mitani, K. Takanashi, S. Takahashi, S. Maekawa, and H. Fujimori, Phys. Rev. B **61**, 46 (2000).
- ¹⁰K. Takanashi, S. Mitani, J. Chiba, and H. Fujimori, J. Appl. Phys. 87, 6331 (2000).
- ¹¹K. Yakushiji, S. Mitani, K. Takanashi, and H. Fujimori, J. Appl. Phys. **91**, 7038 (2002).
- ¹²W. G. van der Wiel, S. De Franceschi, J. M. Elzerman, T. Fujisawa, S. Tarucha, and L. P. Kouwenhoven, Rev. Mod. Phys. **75**, 1 (2003).
- ¹³I. Weymann and J. Barnaś, J. Magn. Magn. Mater. **272**, 1477 (2004).
- ¹⁴Single Charge Tunneling, edited by H. Grabert and M. Devoret (Plenum, New York 1992).
- ¹⁵J. Barnaś and A. Fert, J. Magn. Magn. Mater. **192**, 391 (1999); A.
 N. Korotkov and V. I. Safarov, Phys. Rev. B **59**, 89 (1999).