Spin correlations at hopping in magnetic structures:
From tunneling magnetoresistance to single-spin transistor

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ABSTRACT

Spin correlations at hopping are responsible for large magnetoresistance at trap-assisted resonance tunneling between normal metallic and ferromagnetic electrodes. The reason for the spin correlations at hopping is the spin-selective escape rate, which results in non-zero average spin at a trap. This causes a dependence of the trap occupation and, therefore, the current on the average spin. Surprisingly, strong spin dephasing enhances the amplitude of the magnetoresistance at trap-assisted tunneling from a normal metal to a ferromagnet. Spin dephasing can also boost the tunneling magnetoresistance in magnetic tunnel junctions. Spin relaxation, however, reduces the spin correlations and associated effects, as expected.

Since the spin on the trap is a vector quantity, it produces unusual correlations in multi-terminal devices. Our analysis of a three-terminal device with normal metallic and ferromagnetic electrodes and trap-assisted hopping implies that the spin correlations result in current-voltage dependences characteristic to a single-electron transistor. Importantly, the transfer characteristics are determined by the spin correlations and the spin blockade alone as, because of the finite transition rate between the trap and the normal metallic electrodes, the current is not Coulomb blocked and it always flows through the trap-source, trap-drain, and trap-gate junctions. However, when both the gate and the source electrodes are ferromagnetic with high interface spin polarizations and anti-parallel, the current through all junctions is either suppressed or it flows only between source and drain depending on the voltages applied, in complete analogy to a single electron transistor.

Keywords: spin-dependent hopping, tunneling magnetoresistance, single-spin switch

1. INTRODUCTION

Single-electron hopping plays an important role in determining transport properties in undoped semiconductors and dielectrics. The Coulomb interaction plays an important role as well, as it leads to the repulsion of the charges on a trap and the Coulomb blockade, thus forbidding the double occupancy of the trap. The Coulomb repulsion results in strong charge correlations at transport. Indeed, when electrons tunnel through a trap [1], a second electron from a metallic contact cannot enter the trap, if it is already occupied by an electron. However, when the electron is released from the trap to a contact, the Coulomb repulsion does not prevent the second electron entering the trap. Therefore, the electron transport is performed in sequences consisting of an electron hopping from the source electrode to the trap, followed by the electron escaping from the trap to the drain electrode. Because the Coulomb repulsion is a purely classical interaction, the electron tunneling through the trap represents an example of classically correlated charge transport.

From the point of view of devices, the Coulomb blockade has led to the discovery of the single-electron transistor [1], which is a three-terminal device consisting of a small metallic island (dot) weakly coupled to the source and drain electrode by means of tunneling through the thick potential barriers. Because of the weak coupling, electrons are allowed to jump on the dot from the source and to escape to the drain one-by-one. However, due to the Coulomb blockade, the transport through the dot at small source-drain voltages is suppressed, unless the free energy to add an electron to the dot occupied with (N-1) electrons or to extract an electron from the dot occupied with N electrons is exactly zero [1]. As the potential of the dot and, therefore, the addition/extraction energies can be tuned by the gate electrode capacitively coupled to the dot, a current switch between the on/off states can be build. As the transport through the dot is carried out by a single electron jumping on/from the dot, the switch is termed the single-electron transistor.

In addition to its electron charge, the electron possesses spin. From the physical phenomena related to a single spin, the Pauli, or spin, blockade is known to the single-electron transport, in addition to the Coulomb blockade. The Pauli

exclusion principle forbidding two electrons with the same spin projections to occupy the trap quantum state results in yet another correlation affecting the transport through the double-quantum dot system in a magnetic field [2].

The spin blockade is very sensitive to the spin decoherence and relaxation effects and is therefore typically observed at low temperature. However, recently spin correlations resulting in large magnetoresistance and magnetoluminescence effects in organic semiconductors and organic light-emitting diodes [3] were reported at room temperature. The transport is due to the variable-range spin-dependent single-electron hopping through a network of traps. Due to the Pauli blockade, the current is very sensitive to the strength of the external magnetic field, which aligns the spins and inhibits the current.

In the case when the source and drain electrodes are ferromagnetic, the spin correlations may also be observed at the transport through a *single* quantum dot. Indeed, an electron impinging a ferromagnetic electrode from a trap has a larger probability to escape, if its spin is parallel to the magnetization of the electrode [4]. In this case the drain electrode plays a role similar to the second quantum dot in the Pauli spin blockade experiments [2], which results in the Pauli blockade-like trap-assisted tunneling transport between the ferromagnetic electrodes. This spin-dependent resonant tunneling is most likely responsible for the large magnetoresistance modulation [5] observed in three-terminal spin-injection experiments [6-12], as the spin accumulation has been proven to be not the source of the signal [13]. As the signal has been observed at temperatures as high as 500K [7], envisioning a single-spin switch seems conceivable.

Here we report on a three-terminal single-spin switching device employing for its operation both the spin and the Coulomb blockade. The switch consists of a trap weakly coupled to three ferromagnetic electrodes by tunneling transition rates. In this case the spin on the trap is a vector quantity determined by the magnetic polarizations of the elctrodes, transition rates as well as the weak non-quantizing magnetic field, which results in unusual correlations in the multi-terminal device. In order to describe the switch, we first ellaborate on spin-dependent trap-assisted hopping between a normal metallic and a ferromagnetic electrode. We then generalize the description to spin-dependent hopping in a magnetic tunnel junction with both electrodes ferromagnetic. After that we analyze a multi-terminal device governed by spin-dependent hopping by considering an example of a three-terminal switch, where the source, the gate, and the drain are ferromagnetic.

2. SPIN-DEPENDENT TRAP-ASSISTED HOPPING FROM A NORMAL METALLIC TO A FERROMAGNETIC ELECTRODE

We consider the transport due to spin-dependent trap-assisted tunneling. The trap is located between a normal metallic and a ferromagnetic electrode (Fig.1a). The trap can be empty or occupied by a single electron. The non-quantized magnetic field **B** is applied at an angle Θ with respect to the magnetization direction in the ferromagnetic drain contact.
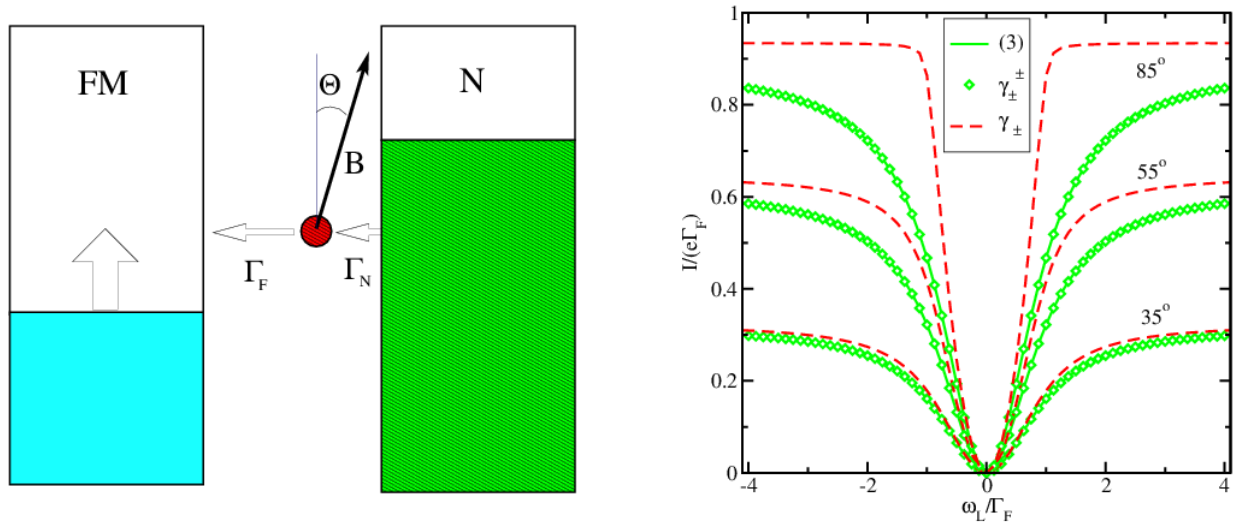

Figure 1. (a) Schematic illustration of trap-assisted tunneling, (b) Magnetoresistance computed with the stationary solution of (3) for spin-dependent sequential tunneling (diamonds). The results are compared with the findings in [5] (solid lines) and[16] (dashed lines).

The tunneling rate Γ_N from the normal metallic contact to the trap does not depend on spin, while the tunneling rate from the trap to the ferromagnet -aligned to the magnetization is spin-dependent and is determined by the two rates $\Gamma_\pm$ for the spin projection on the trap aligned and anti **M**. The rates can be expressed as

$$\Gamma_\pm = \Gamma_F(1 \pm p), \tag{1}$$

where the spin current polarization at the interface of the ferromagnet $p \leq 1$ is defined as [5]

$$p = \frac{\Gamma_+ - \Gamma_-}{2\Gamma_F}. \tag{2}$$

The equations for the site occupation probability n and the spin on the trap **s** are determined from the equations [14]

$$\frac{d}{dt}n = \Gamma_N(1-n) - \Gamma_F n - \Gamma_F \mathbf{p}\mathbf{s}, \tag{3a}$$

$$\frac{d}{dt}\mathbf{s} = -\Gamma_F \mathbf{s} - \mathbf{p}\Gamma_F n + [\mathbf{s} \times \boldsymbol{\omega}_L], \tag{3b}$$

where $\mathbf{p} = p\frac{\mathbf{M}}{M}$ and $\boldsymbol{\omega}_L = \frac{e\mathbf{B}}{mc}$ is the Larmor precession frequency. In the case $p = 0$ one obtains the standard balance equation for the occupation decoupled from the spin. The spin polarization of the ferromagnetic drain electrode couples the spin degree of freedom into the equation for the site occupation, which results in the resistance dependence on the magnetic field [5] shown in Fig.1b by solid lines, for several values of Θ.

The transport process consists of two consecutive hops from the normal metallic source electrode to the trap and an escape from the trap to the ferromagnetic electrode [15]. The first step is characterized by a single rate Γ_N. However, the escape from the trap is described by a matrix because of the coupling between the spin and the occupation at the trap. The escape probability $P(t)$ from the trap in the case of spin-independent hopping is described as

$$P_2(t) = 1 - n(t), \tag{4}$$

where $n(t)$ is the solution of the differential equation [14]:

$$\frac{d}{dt}\begin{pmatrix} n \\ s_x \\ s_y \\ s_z \end{pmatrix} = -\mathbf{A}\begin{pmatrix} n \\ s_x \\ s_y \\ s_z \end{pmatrix} \tag{5}$$

The 4×4 relaxation matrix **A** is written in the coordinate system with the OZ axis parallel to the magnetic field **B**.

$$\mathbf{A} = \begin{pmatrix} \Gamma_F & p\Gamma_F \sin\Theta & 0 & p\Gamma_F \cos\Theta \\ p\Gamma_F \sin\Theta & \Gamma_F & \omega_L & 0 \\ 0 & -\omega_L & \Gamma_F & 0 \\ p\Gamma_F \cos\Theta & 0 & 0 & \Gamma_F \end{pmatrix} \tag{6}$$

Because the trap is populated by hopping from the normal metallic electrode, spin-up and spin-down states are occupied with equal probabilities. Thus, the initial spin at the trap is zero, and the initial condition for (6) is

$$\begin{pmatrix} n \\ s_x \\ s_y \\ s_z \end{pmatrix}(t=0) = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}. \tag{7}$$

We evaluate now the current as follows. The charge transport consists of a series of repeated cycles. An electron jumps from the normal metallic electrode to the trap with the rate Γ_N, and then escapes from the trap to the ferromagnetic electrode with the probability $P(t)$ (4). Double occupancy of the trap is prohibited by the Coulomb repulsion. One electron charge is transferred at each cycle during the time $\tau_1 + \tau_2$. The average time τ_1 is evaluated in accordance with the escape probability $P_1(t)=1- \exp(-t\,\Gamma_N)$. The time τ_2 is computed according to $P_2(t) = 1-n(t)$. In contrast to spin-independent tunneling, the probability is not determined by a single exponential distribution. As it follows from the solution of (5-7), the escape probability (4) is defined by the four eigenvalues of the 4×4 relaxation matrix **A** (6).

$$\gamma_{\pm}^{+} = \Gamma_F \pm \left(\frac{1}{2} \left(p^2 \Gamma_F^{\;2} - \omega_L^2 \right) + \sqrt{ \frac{ \left(p^2 \Gamma_F^{\;2} - \omega_L^2 \right)^2 }{4} + \omega_L^2 p^2 \Gamma_F^{\;2} \cos^2 \Theta } \right)^{1/2} \qquad (8a)$$

$$\gamma_{\pm}^{-} = \Gamma_F \pm \left(\frac{1}{2} \left(p^2 \Gamma_F^{\;2} - \omega_L^2 \right) - \sqrt{ \frac{ \left(p^2 \Gamma_F^{\;2} - \omega_L^2 \right)^2 }{4} + \omega_L^2 p^2 \Gamma_F^{\;2} \cos^2 \Theta } \right)^{1/2} \qquad (8b)$$

The solution of (5-7) is then written as

$$\begin{pmatrix} n \\ s_x \\ s_y \\ s_z \end{pmatrix} (t) = \sum_{\sigma,\sigma'} C_{\sigma'}^{\sigma} \, \boldsymbol{u}_{\sigma}^{\sigma'} \exp(-t \, \gamma_{\sigma}^{\sigma'}) \quad , \qquad (9)$$

where $\boldsymbol{u}_{\sigma}^{\sigma'}$ are the eigenvectors of **A** in (6). The coefficients $C_{\sigma'}^{\sigma}$ are uniquely determined by the initial condition (7).

The first component of (9) determines the escape probability $P_2(t) = 1-n(t)$. The average time τ_2 can be calculated analytically. The stationary current I computed as

$$I = \frac{e}{\tau_1 + \tau_2} \qquad (10)$$

is shown in Fig.1b with diamonds. The current from (10) coincides with the one obtained in [5] (Fig.1b, solid lines), but is significantly lower than the current computed with the expression from [16] shown with dashed lines in Fig.1b. In fact, the current values from [16] are too large for all directions of the magnetic field except the one, when **B** is parallel to **p**. This implies that the escape rates used in [16] are faster. The escape time distribution probability in [16] is determined by the two tunneling rates (8a) from each of the Zeeman levels renormalized by coupling to the ferromagnetic contact (Fig.1b, dashed lines). This approximation is appropriate only in the case, when the magnetic field is aligned with the magnetization. In the general case the consideration based on the matrix equation (5-7) is necessary to reproduce the correct transition probabilities and currents. This explains [14] the controversy between the results [5, 16].
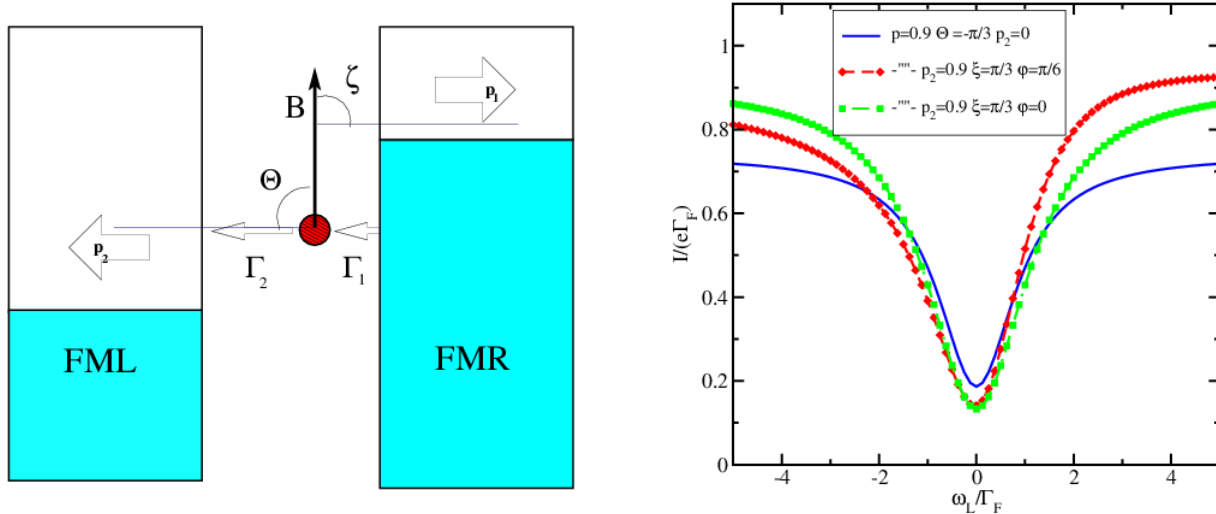

Figure 2. (a) When tunneling between two ferromagnetic electrodes, the direction of the source magnetization is specified by ζ and the polar angle φ; (b) Tunneling magnetoresistance for $p_1=0.9$ compared with that for $p_1=0$.

3. SPIN-DEPENDENT HOPPING IN MAGNETIC TUNNEL JUNCTION

We consider the spin-dependent hopping transport between two non-collinear ferromagnetic contacts (Fig.2a) with the interface current polarizations $\mathbf{p}_{1,2} = \frac{\Gamma_{1,2}^+ - \Gamma_{1,2}^-}{2\Gamma_{1,2}} \frac{\mathbf{M}_{1,2}}{M_{1,2}}$ in the source(drain) electrode with the saturation magnetization $M_{1,2}$, and $\Gamma_{1,2} = \frac{\Gamma_{1,2}^+ + \Gamma_{1,2}^-}{2}$. In order to determine the escape probability $P_2(t) = 1\text{-}n(t)$ from the trap one has to solve equation (4). The tunneling rate from the ferromagnetic source electrode to the trap defining the occupation is the sum of the tunneling rates with the spin-up and the spin-down projections on the axis oriented along the source polarization $\mathbf{p}_1$:

$$\Gamma_{S \to Trap} = \Gamma_1^+ + \Gamma_1^- = 2\Gamma_1 \qquad (11)$$

The rate (11) plays a similar role as Γ_N in sequential tunneling considered in the previous section. Although $\Gamma_{S \to Trap}$ does not depend on the spin polarization of the source, the electron on the trap originating from the ferromagnetic source electrode exhibits non-zero spin defined by the spin polarization $\mathbf{p}_1$ as the ferromagnetic source electrode injects electrons with the spin defined by the source polarization vector. The non-zero initial spin generalizes the initial condition (7) to the case of trap-assisted tunneling between the two ferromagnetic electrodes [14]:

$$\begin{pmatrix} n \\ s_x \\ s_y \\ s_z \end{pmatrix}(t = 0) = \begin{pmatrix} 1 \\ p_1 \sin\zeta \cos\varphi \\ p_1 \sin\zeta \sin\varphi \\ p_1 \cos\zeta \end{pmatrix} \qquad (12)$$

Now in order to evaluate the stationary current it remains to solve (4-6) with the initial condition (12), and the matrix **A** modified so that **p** is replaced by $\mathbf{p}_2$ and Γ_F by Γ_2.

Results of the calculations are shown in Fig.2b. The magnetoresistance shows a more pronounced minimum at small magnetic fields as compared to tunneling from a normal metallic electrode. This is due to the fact that the probability to inject spins with the orientation opposite to $\mathbf{p}_2$ causing the spin blockade is mostly determined by Γ_1^- for the parameters chosen in Fig.2b and is thus reduced as compared to the probability of tunneling of 50% from a normal metallic source electrode. The magnetoresistance also shows a dependence on the direction of the magnetic field in the case when **B**, $\mathbf{p}_1$, and $\mathbf{p}_2$ are not lying within the same plane, although at high magnetic fields the symmetry with respect to the direction of the field is restored.

4. SINGLE SPIN TRANSISTOR

We analyze a three-terminal device in the configuration shown in Fig.3a, where the source "1", the gate "2", and the drain "3" are ferromagnetic, each described by the corresponding spin polarization $\mathbf{p}_i$ ($i = 1,2,3$). The potential at the trap is determined by the gate voltage $V_{GS,}$ the drain-source voltage $V_{DS,}$ and the capacitances C_i (i=1,2,3), which are assumed equal to C. The current I_α is defined positive, if it flows from the electrode α=G(gate), S(source), or D(drain) to the trap. The current continuity $I_G + I_S + I_D = 0$ is thus automatically ensured.
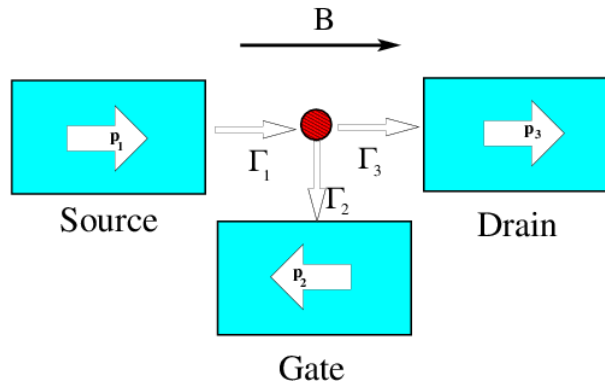

Figure 3. Schematic illustration of the single-electron switch. Electron transport is caused by spin-dependent hopping between the ferromagnetic contacts.
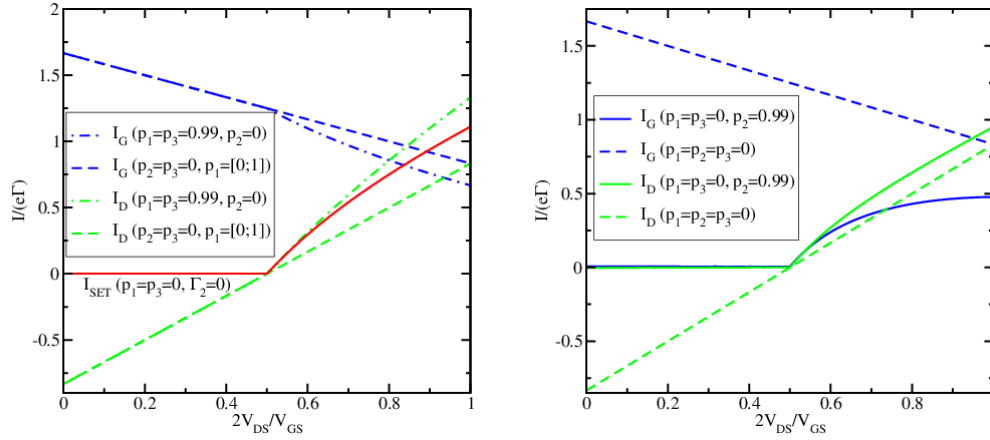
Figure 4. (a) Gate and drain currents, when the gate is non-ferromagnetic (p_2=0). The single-electron transistor's drain current is also shown; (b). The drain and gate currents are suppressed, if the trap-drain junction is backward biased and the gate is ferromagnetic (p_2=0.99). The gate current is nonzero for V_{DS}>V_{GS}/4.

First we consider the single-electron transistor mode, where there is no tunneling between the trap and the gate $\Gamma_2 = 0$ and all electrodes are non-ferromagnetic ($p_i = 0$). We apply a constant gate voltage V_{GS}=e/C_2. For zero drain-source voltage V_{DS} we are in the middle of the Coulomb blockade region with a single electron on the trap, and no current can flow between the source and drain.

For $V_{DS} < V_{GS} C_2/(C_1+ C_2)$ the junction's "trap-drain" is biased in opposite direction, and there is no source-drain current (Fig.4a, solid line). As soon as $V_{DS} > V_{GS} C_2/(C_1+ C_2)$, the Coulomb blockade is overcome, and the current starts flowing (Fig.4a, solid line). We evaluate the current by using the rates standard to single-electron tunneling as [17]

$$\Gamma_i(V_{eff}) = \Gamma_i \left(\frac{V_{eff}}{V_{GS}}\right) , \qquad (13)$$

where $V_{eff} = (V_b + V_a)/2 > 0$ is the effective voltage of the junction i calculated as an average of the voltages before V_b and after V_a the electron jump.

If we now allow tunneling between the trap and the gate, $\Gamma_1 = \Gamma_2 = \Gamma_3 \equiv \Gamma$, the Coulomb blockade is lifted, as an electron from the trap can escape to the gate at V_{GS}=e/C_2. The electrodes are considered metallic with $p_i = 0$. At V_{DS}=0 the source is not biased with respect to the drain, and the equal currents flow through the source and drain junctions from the electrodes, which then combine into the gate current (Fig.4a, dashed lines). At V_{DS}= V_{GS} the drain and gate junctions are equivalent, and the corresponding currents are equal. Making the source electrode ferromagnetic with
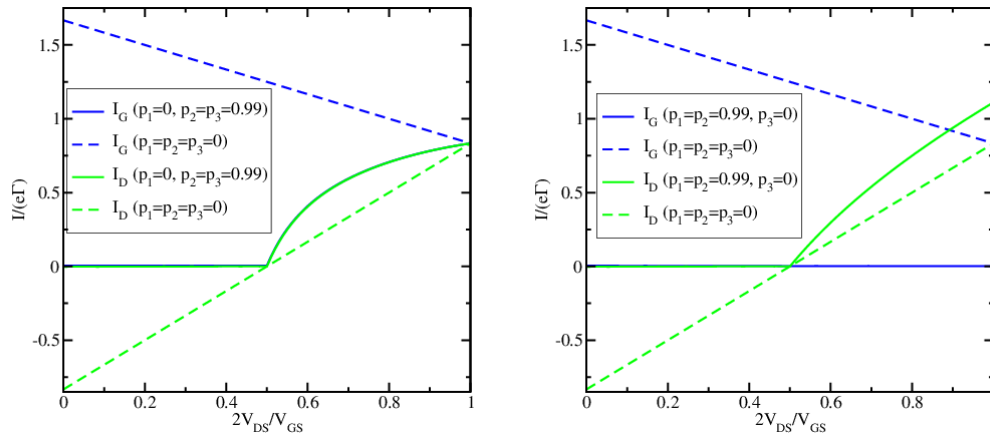

Figure 5. (a) The currents are similar to those in Fig.4b, when, in addition to the gate, the drain is ferromagnetic (p_2=p_3=0.99); (b) The gate current is suppressed, when the source and the gate are ferromagnetic (p_1=p_2=0.99).
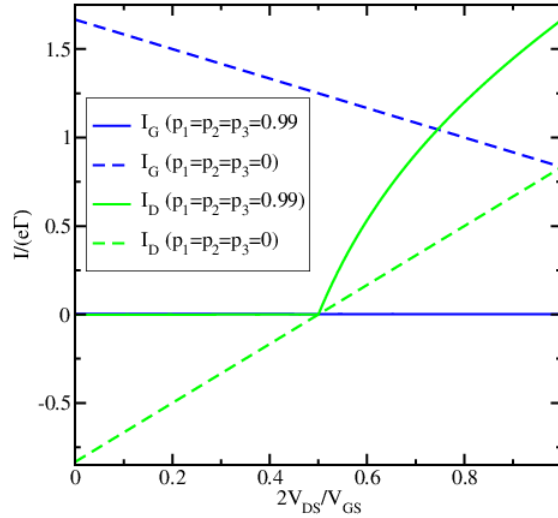
Figure 6. The drain current is the largest, while the gate current is suppressed, when all electrodes are ferromagnetic ($p_1=p_2=p_3=0.99$) as in the configuration shown in Fig.3.

high spin current polarization $p_1=0.99$ does not modify the current-voltage characteristics as the escape from the trap is spin-independent in the case of a non-ferromagnetic gate and drain $p_2=p_3=0$ (Fig.4a, dashed lines).

However, when the drain is also ferromagnetic, $p_1=p_3=0.99$, $p_2=0$ (Fig.3), one observes an increase of the drain current at $V_{DS} > V_{GS}/2$ shown by the dashed-dotted lines in Fig.4a. At these conditions the drain junction is positively biased, and the current flows easily between the source and drain since the magnetizations of the electrodes are parallel $\mathbf{p}_1=\mathbf{p}_3$. Therefore, the spin at the trap is also parallel to the magnetizations. Because the electron at the trap is spin-polarized, its probability to escape through the gate junction is reduced as only a single spin-polarized channel is open, in contrast to a spin-un-polarized electron, when both channels for spin-up and spin-down are open.

The situation changes dramatically when the gate is ferromagnetic, $p_2=0.99$, $p_1=p_3=0$. At $V_{DS} < V_{GS}/2$ both the source and drain junctions are negatively biased, so that the electrons flow from these junctions to the trap and then escape to the highly polarized gate. The system is similar to the one with the trap-assisted tunneling between the normal metallic and the ferromagnetic electrode. In complete analogy to this case, the current suppression characteristic to spin-dependent hopping is observed in Fig.4b, solid lines.

At $V_{DS} > V_{GS}/2$ the drain junction is positively biased, and the spin blockade is lifted. An electron from the trap can always escape to the drain regardless of its spin. This leads to a non-zero drain current (Fig.4b, solid line). Because the trapped electron with its spin opposite to $\mathbf{p}_2$, which could not tunnel to the gate for $V_{DS} < V_{GS}/2$ and was responsible for blocking the gate current, can easily escape to the drain, the gate current also starts flowing (Fig.4b). Adding ferromagnetism to the drain in the configuration shown in Fig.3 ($\mathbf{p}_2 = -\mathbf{p}_3$, $p_2=p_3=0.99$, $p_1=0$) does not alter the current-voltage behavior significantly (Fig.5a). Since the source remains non-ferromagnetic, it provides electrons with spin parallel or anti-parallel to $\mathbf{p}_2$ (or vice versa for $\mathbf{p}_3$) with equal probability 50%, the drain and gate currents are equal at $V_{DS} = V_{GS}$, although it is surprising that the currents are actually equal in the whole range $V_{DS} > V_{GS}/2$ (Fig.5a).

Adding ferromagnetism to the source instead of the drain in the configuration in Fig.3 ($\mathbf{p}_2 = -\mathbf{p}_1$, $p_1=p_2=0.99$, $p_3=0$) does not alter the currents blocking at $V_{DS} < V_{GS}/2$ (Fig.5b). Indeed, the drain junction is negatively biased which prevents an electron to tunnel from the trap to the drain. The tunnel current into the gate is suppressed by the spin blockade as the drain electrode delivers electrons with the spin anti-parallel to $\mathbf{p}_2$ with 50% probability, while the source with the probability 99%. These electrons cannot tunnel to the gate thus blocking the gate current.

Because the source provides electrons with their spins mostly opposite to $\mathbf{p}_2$, they cannot tunnel from the trap to the gate. The drain junction is positively biased at $V_{DS} > V_{GS}/2$, so the drain does not deliver electrons to the trap. Therefore, in contrast to Fig.5a, the gate current remains negligible in the whole range of V_{DS}. The drain current is substantial at $V_{DS} > V_{GS}/2$ as electrons are allowed to tunnel to the normal metallic drain electrode.

Finally, if one considers all electrodes to be ferromagnetic in the configuration shown in Fig.3 ($\mathbf{p}_1 = \mathbf{p}_3 = -\mathbf{p}_2$, $p_1 = p_2 = p_3 = 0.99$), mostly the electrons with their spins opposite to $\mathbf{p}_2$ are delivered to the trap. The gate current remains negligible in the whole range of V_{DS} (Fig.6). The drain current is blocked at $V_{DS} < V_{GS}/2$ (Fig.6) as the gate junction is negatively biased, which prevents electrons tunneling to the drain. At $V_{DS} > V_{GS}/2$ the drain current flows. Since the source and the drain are strongly polarized with their magnetizations aligned, the current value shown in Fig.6 is larger than in the case of a non-ferromagnetic drain (Fig.5b) as only one-half of the states is now available in the drain to accommodate polarized electrons from the source. We stress that, although the current-voltage characteristic in Fig.6 is qualitatively similar to that of a single-electron transistor (Fig.4a, solid line), the suppression of the gate current and the switching is due to the *spin correlations* and the *spin blockade* at spin-dependent trap-assisted hopping in a multi-terminal device with ferromagnetic electrodes.

5. CONCLUSION

The Coulomb interaction leads to a strong repulsion on the trap - a Coulomb blockade, while the Pauli exclusion results in spin-dependent correlations at transport. The spin correlations manifest themselves in unusual transport properties at trap-assisted hopping between normal metallic and ferromagnetic electrodes as well as in magnetic tunnel junctions. The spin correlations are due to spin-dependent escape rates, which result in a non-zero spin on a trap. Because the average spin is a vector property, it results in an unusual behavior in multi-terminal devices with ferromagnetic electrodes. In particular, a single-spin switch with transfer characteristics similar to those of a single-electron transistor is demonstrated.

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