Cotunneling through two-level quantum dots weakly coupled to ferromagnetic leads

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Abstract. – The spin-polarized transport through two-level quantum dots weakly coupled to ferromagnetic leads is considered theoretically in the Coulomb blockade regime. It is assumed that the dot is doubly occupied, so that the current flows due to cotunneling through singlet and triplet states of the dot. It is shown that transport characteristics strongly depend on the ground state of quantum dot. If the ground state is a singlet, differential conductance \( G \) displays a broad minimum at low bias voltage, while tunnel magnetoresistance (TMR) is given by the Julliere value. If triplet is the ground state of the system, there is a maximum in differential conductance at zero bias when the leads are magnetized in antiparallel. The maximum is accompanied by a minimum in TMR. The different behavior of \( G \) and TMR may thus help to determine the ground state of the dot and the energy difference between the singlet and triplet states.

Introduction. – Transport properties of quantum dots coupled to ferromagnetic leads have been a subject of thorough studies since a few years [1, 2]. The considerations concerned both the strong coupling regime where the Kondo physics emerges [3], as well as the weak coupling regime. In the weak coupling regime most of theoretical works addressed the problem of spin-dependent sequential transport through quantum dots hosting single orbital level [4]. In the sequential tunneling regime, if the applied bias voltage exceeds a certain threshold voltage, electrons tunnel one by one through the system, otherwise the current is suppressed leading to the Coulomb blockade effect. Although in the Coulomb blockade regime the sequential transport is suppressed, the current can be still mediated by higher-order tunneling processes such as cotunneling which involves correlated tunneling of two electrons via virtual states of the dot [5]. Spin-polarized cotunneling transport has been addressed very recently. It has been shown that when the dot is singly occupied, a zero-bias anomaly appears in differential conductance when the magnetizations of the leads are aligned in antiparallel [6, 7]. These considerations were performed only for single-level quantum dots. In real systems, however, usually more than one energy level participate in transport, which can lead to further interesting effects [8].

The goal of this paper is to address the problem of spin-dependent transport through two-level quantum dots coupled to ferromagnetic leads in the cotunneling regime.
we analyze the case when the dot is doubly occupied at equilibrium and the system is in the Coulomb blockade regime. In this case, depending on the value of the exchange interaction, either the singlet or triplet state will be favored. We show that the differential conductance \( G \) and tunnel magnetoresistance (TMR) display a strong dependence on the ground state of the system. If the ground state is a singlet, \( G \) exhibits a broad minimum at low bias voltage in both magnetic configurations of the system, while TMR takes the Juliere value [9]. On the other hand, if the ground state is a triplet, there is a maximum in differential conductance at zero bias in the antiparallel configuration, accompanied by a minimum in TMR. The different behavior of both differential conductance and TMR provides thus a handle to determine the ground state of the system.

The systems discussed in this paper may be realized experimentally for example in metallic single-wall carbon nanotube (SWCNT) quantum dots contacted to ferromagnetic leads [10]. As shown recently, the shell filling of SWCNT quantum dots exhibits four-electron periodicity when sweeping the gate voltage [11]. By changing the gate, one can tune the system to the Coulomb blockade regime where the dot is doubly occupied at equilibrium.

Model and method. – We consider spin-polarized cotunneling transport through quantum dots with two orbital levels coupled to ferromagnetic leads. It is assumed that the magnetic moments of the leads can form either parallel or antiparallel magnetic configuration, see fig. 1. The Hamiltonian \( H \) of the system involves four terms, \( H = H_L + H_R + H_D + H_T \). The first two terms describe the noninteracting itinerant electrons in the leads, \( H_r = \sum_{k\sigma} \varepsilon_{rk\sigma} c_{rk\sigma}^\dagger c_{rk\sigma} \), for \( r = L, R \), corresponding to the left and right leads, with \( \varepsilon_{rk}\sigma \) being the energy of an electron with wave number \( k \) and spin \( \sigma \) in the lead \( r \), and \( c_{rk\sigma}^\dagger (c_{rk\sigma}) \) denoting the respective creation (annihilation) operator. The quantum dot is described by the Hamiltonian [12]

\[
H_D = \sum_{j\sigma} \varepsilon_j n_{j\sigma} + U \sum_j n_{j\uparrow} n_{j\downarrow} + U' \sum_{\sigma'\sigma} n_{1\sigma} n_{2\sigma'} - J \sum_{\alpha\beta\gamma\delta} d_{1\alpha\sigma}^\dagger d_{2\beta\gamma\sigma} d_{2\beta\gamma\sigma}^\dagger d_{1\alpha\sigma},
\]

where \( n_{j\sigma} = d_{j\sigma}^\dagger d_{j\sigma} \) and \( d_{j\sigma}^\dagger (d_{j\sigma}) \) is the creation (annihilation) operator of an electron with spin \( \sigma \) on the \( j \)th level \( (j = 1, 2) \), while \( \varepsilon_j \) is the corresponding energy. On-level and inter-level Coulomb interaction between electrons is described by \( U \) and \( U' \), respectively. The last term in \( H_D \) corresponds to the exchange energy due to the Hund’s rule, with \( J \) being the respective exchange coupling and \( \vec{\sigma} \) denoting a vector of Pauli spin matrices. To simplify further discussion of numerical results we assume \( U' = U \). For the energy of the first orbital level we write \( \varepsilon_1 = \varepsilon \) and \( \varepsilon_2 = \varepsilon + \delta\varepsilon \), where \( \delta\varepsilon \) is the splitting of the dot orbital levels. For example in metallic SWCNT quantum dots, \( \delta\varepsilon \) would correspond to the energy mismatch between the two subbands of the nanotube [11]. The mismatch can vary with the boundary conditions at the nanotube-electrode interface.

The last term of \( H \) describes the tunneling processes between the dot and electrodes

\[
H_T = \sum_{r=L,R} \sum_{jk\sigma} \left( T_{rj} c_{r\sigma}^\dagger d_{j\sigma} + T_{ij}^* d_{j\sigma}^\dagger c_{r\sigma} \right),
\]

Fig. 1 – Schematic of a quantum dot coupled to ferromagnetic leads. The magnetizations of the leads can form either parallel or antiparallel magnetic configuration. The system is symmetrically biased.
with $T_{r\sigma}$ denoting the tunnel matrix elements between the lead $r$ and $j$th level. Coupling of the $j$th dot level to external leads can be described by $\Gamma_{r\sigma}^{\pm} = 2\pi |T_{r\sigma}|^2 \rho_{\sigma}$, with $\rho_{\sigma}$ being the spin-dependent density of states in the lead $r$. By introducing the spin polarization of lead $r$, $p_r = (\rho_{\uparrow} - \rho_{\downarrow})/(\rho_{\uparrow} + \rho_{\downarrow})$, the couplings can be expressed as, $\Gamma_{r\sigma}^{\pm} = \Gamma_{r\sigma}(1 \pm p_r)$, with $\Gamma_{r\sigma} = (\Gamma_{r\sigma}^+ + \Gamma_{r\sigma}^-)/2$. Here, $\Gamma_{r\sigma}^+$ and $\Gamma_{r\sigma}^-$ describe the coupling of the $j$th level to spin-majority and spin-minority electron bands, respectively. We assume that the system is coupled symmetrically to the leads $\Gamma_{r\sigma} \equiv \Gamma/2$ and $p_r = p_L \equiv p$. As reported in ref. [13], typical values of $\Gamma$ in the weak coupling regime are of the order of tens of $\mu$eV.

In order to find the current flowing through the dot in the Coulomb blockade, one has to calculate the respective cotunneling rates. Within the second-order perturbation theory, the rate of a cotunneling process from lead $r$ to lead $r'$ which changes the dot state from $|\chi\rangle$ into $|\chi'\rangle$ is given by

$$\gamma_{r\sigma}^{\chi\chi'} = \frac{2\pi}{\hbar} \left| \sum_{v} \langle \Phi_{v \sigma}^{\chi'} | H_T | \Phi_v \rangle \langle \Phi_v | H_T | \Phi_{v \sigma}^{\chi} \rangle \right|^2 \frac{1}{\varepsilon_i - \varepsilon_f} \delta(\varepsilon_i - \varepsilon_f), \quad (3)$$

with $\varepsilon_i$ and $\varepsilon_f$ denoting the energies of initial and final states, $|\Phi_{v \sigma}^{\chi}\rangle$ being the state of the system with an electron in the lead $r$ and the dot in state $|\chi\rangle$, whereas $|\Phi_v\rangle$ is a virtual state with $\varepsilon_v$ denoting the corresponding energy. Among different cotunneling processes one can distinguish the single-barrier ($r = r'$) and double-barrier ($r \neq r'$) cotunneling as well as spin-flip ($\chi \neq \chi'$) and non-spin-flip ($\chi = \chi'$) cotunneling. The spin-flip processes change the state of the dot, whereas the non-spin-flip processes are fully coherent, i.e. they do not change the dot state. The current flows through the system due to double-barrier cotunneling processes. On the other hand, the single-barrier processes do not contribute directly to electric current, however, they can change the dot occupations, and this way also the current. The details of how to determine different cotunneling rates can be found in ref. [5].

The cotunneling current flowing through the system from the left to right lead is given by

$$I = e \sum_{\chi \chi'} \sum_{r\sigma} P_{\chi} \left[ \gamma_{LR}^{\chi\chi'} - \gamma_{RL}^{\chi\chi'} \right], \quad (4)$$

where $P_{\chi}$ denotes the corresponding occupation probability. The probabilities $P_{\chi}$ can be found from the master equation, $0 = \sum_{r\sigma} \sum_{\chi'} \left[ -\gamma_{r\sigma}^{\chi\chi'} P_{\chi} + \gamma_{r\sigma}^{\chi'\chi} P_{\chi'} \right]$, together with the normalization condition $\sum_{\chi} P_{\chi} = 1$.

We consider the case when the dot is doubly occupied at equilibrium and the system is in the Coulomb blockade regime. There are then six different two-particle states in the dot possible, these are three singlets $|S = 0, M = 0\rangle_1 = (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)/\sqrt{2}$, $|0, 0\rangle_2 = |\uparrow\uparrow\rangle|0\rangle$, $|0, 0\rangle_3 = |0\rangle|\uparrow\downarrow\rangle$, and three triplets $|1, 0\rangle_1 = (|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)/\sqrt{2}$, $|1, 0\rangle_2 = |\uparrow\downarrow\rangle|\downarrow\uparrow\rangle$, $|1, 0\rangle_3 = |\downarrow\uparrow\rangle|\downarrow\uparrow\rangle$, where the first (second) $ket$ corresponds to the first (second) orbital level of the dot. In the case of finite level spacing, $\delta \varepsilon > k_B T, \Gamma$, and $J < \delta \varepsilon$, the lowest singlet state is $|0, 0\rangle_2$. In the following, we show that transport characteristics strongly depend on the ground state of the system. It is therefore useful to introduce the difference between the energy of the lowest lying singlet ($\varepsilon_S$) and triplet ($\varepsilon_T$) states, $\Delta_{ST} = \varepsilon_S - \varepsilon_T = J - \delta \varepsilon$.

Results and discussion. – Figure 2 presents the bias voltage dependence of differential conductance $G$ in the parallel and antiparallel magnetic configuration for several values of $\Delta_{ST} = J - \delta \varepsilon$. First of all, one can see that the transport characteristics show a distinctly different behavior depending on $\Delta_{ST}$, i.e. the ground state of the quantum dot. For $\Delta_{ST} < 0$, the ground state of the dot is a singlet, $|0, 0\rangle_2$, whereas for $\Delta_{ST} > 0$, the ground state is a
triplet, which is three-fold degenerate, $|1, 0\rangle$, $|1, 1\rangle$, $|1, -1\rangle$. On the other hand, for $\Delta_{ST} = 0$, the dot is in a mixed state and the occupation of singlet and each triplet is equal at equilibrium and given by 1/4.

First, let us discuss the case of $\Delta_{ST} < 0$ shown in fig. 2(a) and (b). In this situation, at low bias voltage the dot is occupied by two electrons on the lowest energy level, $|0, 0\rangle_2 = |\uparrow\downarrow\rangle|0\rangle$, and the ground state is singlet, $S = 0$. Because for $\Delta_{ST} < 0$ and $|eV| < |\Delta_{ST}|$, $P_{|0, 0\rangle_2} \approx 1$, the current is mediated only by non-spin-flip cotunneling processes. However, once $|eV| \gtrsim |\Delta_{ST}|$, the triplet states start participating in transport (in the case of $\Delta_{ST} = -5\Gamma$ also the state $|0, 0\rangle_1$ contributes), leading to an increase in the differential conductance at $|eV| \approx |\Delta_{ST}|$.

In other words, the suppression of cotunneling through $S = 1$ states gives rise to a broad minimum in differential conductance which is present in both magnetic configurations of the system, see fig. 2(a) and (b). The width of this minimum is determined by the splitting between the singlet and triplet states, $2|\Delta_{ST}|$. By measuring the width, one can thus obtain information about the energy difference between the $S = 0$ and $S = 1$ states.

In the case when the current flows only due to non-spin-flip cotunneling through the state $|0, 0\rangle_2$, one can find approximative formulas for the differential conductance. At low temperature and for $\delta \varepsilon \gg k_B T$, $G$ in the parallel configuration can be expressed as

$$G_P^{S=0} = \frac{e^2\Gamma^2}{2\hbar}(1 + p^2) \left[ \frac{1}{(\varepsilon + U)^2} + \frac{1}{(\varepsilon + \delta \varepsilon + 2U)^2} \right],$$

(5)
on the other hand, for the antiparallel configuration one finds, $G_{AP}^{S=0} = (1 - p^2)G_P^{S=0}/(1 + p^2)$. These expressions approximate the minimum in differential conductance at low bias voltage shown in fig. 2(a). The analytical formulas for the general case when more states participate in transport are too cumbersome to be presented here.
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Figure 2(c) displays the differential conductance in the case when the singlet and triplet states are degenerate, $\Delta_{ST} = 0$. First of all, it can be seen that $G$ exhibits a distinctively different behavior in both magnetic configurations. In the parallel configuration the differential conductance has a smooth parabolic dependence on the bias voltage, whereas in the antiparallel configuration there is a maximum at the zero bias voltage. This effect bears a resemblance to that found in the case of singly occupied one-level quantum dots [6, 7]. Here, however, the mechanism leading to the maximum is different – the zero-bias peak appears due to cotunneling through singlet and triplet states of the dot. In the case of $\Delta_{ST} = 0$ and at low bias voltage, all the four dot states, i.e. $|0,0\rangle_2, |1,0\rangle, |1,-1\rangle, |1,1\rangle$, participate in transport on an equal footing. Consequently, the current flows due to both spin-flip and non-spin-flip cotunneling processes. To understand the mechanism leading to the zero-bias peak, one should bear in mind that in the antiparallel configuration the spin-majority electrons of one lead tunnel to the spin-minority electron band of the other lead. For example, for positive bias voltage (electrons tunnel then from the right to left lead), the spin-$\uparrow$ electrons can easily tunnel to the left lead (the spin-$\uparrow$ electrons are the majority ones), while this is more difficult for the spin-$\downarrow$ electrons (they tunnel to the minority electron band). Thus, with increasing the bias voltage, the occupation of state $|1,-1\rangle$ ($|\downarrow\rangle|\downarrow\rangle$) is increased, while the occupation of state $|1,1\rangle$ ($|\uparrow\rangle|\uparrow\rangle$) decreases. The unequal occupations of these triplet states lead thus to a nonequilibrium spin accumulation in the dot, $P_{|1,1\rangle} - P_{|1,-1\rangle} < 0$, which is shown in fig. 3. It is further interesting to note that in the antiparallel configuration the possible non-spin-flip cotunneling processes are proportional to $\Gamma^+_L \Gamma^-_R$ and $\Gamma^-_L \Gamma^+_R$, whereas the spin-flip cotunneling is proportional to $\Gamma^+_L \Gamma^+_R$ and $\Gamma^-_L \Gamma^-_R$. It is clear that the fastest cotunneling processes are the ones involving only the majority spins, i.e. $\Gamma^+_L \Gamma^+_R$. However, because of nonequilibrium spin accumulation, with increasing the bias ($V > 0$), the dot becomes dominantly occupied by majority electrons of the right lead, $P_{|1,-1\rangle} > P_{|1,1\rangle}$, and the processes proportional to $\Gamma^+_L \Gamma^+_R$ are suppressed. As a consequence, the differential conductance drops with the bias voltage, leading to the zero-bias peak, see the dashed line in fig. 2(c). This is thus the nonequilibrium accumulation of spin $S = 1$ which is responsible for the maximum in $G$ at low bias voltage. Because the dot is coupled symmetrically to the leads, there is no spin accumulation in the parallel configuration and the differential conductance exhibits a smooth parabolic dependence on the bias voltage, see the solid line in fig. 2(c).

In the case of $\Delta_{ST} > 0$, at equilibrium the dot is occupied by a triplet. Therefore, at low bias voltage, $|eV| < |\Delta_{ST}|$, transport is mediated by $S = 1$ states. When the voltage is
increased, at $|eV| \approx |\Delta_{ST}|$, the occupation of the singlet state $|0,0\rangle_2$ increases and all the four states start to participate in transport. This leads a step in differential conductance which is clearly visible in both magnetic configurations, see fig. 2(d). The width of the transport region where cotunneling through the singlet state is suppressed is given by $2|\Delta_{ST}|$. As one can see in the figure, in the parallel configuration there is one minimum, while in the antiparallel configuration there are two minima separated by a zero-bias peak. The mechanism leading to the peak at zero bias in the antiparallel configuration is the same as in the aforementioned case of $\Delta_{ST} = 0$. Spin accumulation induced in the dot, see fig. 3, leads to a decrease in differential conductance when increasing the bias voltage from $V = 0$.

The nonequilibrium spin accumulation in the antiparallel configuration as a function of the bias voltage is displayed in fig. 3. In the case of $\Delta_{ST} \geq 0$, spin accumulation is always present for $V \neq 0$, while for $\Delta_{ST} < 0$, spin accumulation appears when $|eV| \approx |\Delta_{ST}|$, i.e. at the onset of cotunneling through triplet states. As pointed previously, spin accumulation is responsible for the maximum in differential conductance. Thus, one could expect that some maxima in $G$ should also appear in the case of $\Delta_{ST} < 0$, where spin accumulation is induced for $|eV| \geq |\Delta_{ST}|$, see fig. 3. This is indeed visible in fig. 2(a) and (b) where two small maxima develop at $|eV| \approx |\Delta_{ST}|$, symmetrically with respect to the zero bias.

![Fig. 4 – (color online) Tunnel magnetoresistance as a function of the bias voltage for $\Delta_{ST} = -5, -2.5, 0, 2.5\Gamma$, as indicated in the figure. The other parameters are the same as in fig. 2](image)

Another feature visible in fig. 2 is the difference between conductance in the parallel and antiparallel magnetic configuration. Conductance in the parallel configuration is generally larger than that in the antiparallel configuration. This is due to the asymmetry between the couplings of the dot levels to the spin majority and spin minority electron bands in the antiparallel configuration [7]. This difference gives rise to tunnel magnetoresistance, $TMR = (I_P - I_{AP})/I_{AP}$, where $I_P$ ($I_{AP}$) is the current flowing through the system in the (anti)parallel configuration [7, 9]. The bias voltage dependence of TMR is shown in fig. 4. As one can see, TMR displays a nontrivial dependence on the ground state of the dot. When the dot is occupied by a singlet, $\Delta_{ST} < 0$, TMR at low bias displays a maximum plateau. With the same assumptions as made when deriving eq. (5), one finds that the maximum in TMR for $\Delta_{ST} < 0$ and $|eV| < |\Delta_{ST}|$ can be approximated by, $TMR_{S=0} = 2p^2/(1 - p^2)$. This value corresponds to the Julliere TMR, which is characteristics of single tunnel junction [9]. In our case, it results from the fact that in this transport regime current flows due to non-spin-flip cotunneling. On the other hand, for $\Delta_{ST} \geq 0$, the TMR exhibits a minimum at zero bias, which can be seen in fig. 4 for $\Delta_{ST} = 0$ and $\Delta_{ST} = 2.5\Gamma$.
Summary. – We have considered spin-polarized transport through a doubly occupied two-level quantum dot coupled to ferromagnetic leads in the cotunneling regime. In this case, due to finite exchange interaction, cotunneling current is mediated by singlet and triplet states of the dot. We have shown that transport strongly depends on the ground state of the system. If the dot is occupied by a singlet at equilibrium, $G$ exhibits a minimum at low bias, whose width is given by the strength of splitting between the singlet and triplet states. In this transport regime the TMR is given by the Julliere value, indicating that transport is coherent. If triplet is the ground state, a zero-bias peak evolves in differential conductance for antiparallel configuration. The zero-bias maximum results from the nonequilibrium accumulation of spin $S = 1$ in the dot. The maximum is accompanied by a minimum in TMR. The different behavior of differential conductance and TMR may help in determining the ground state of the dot as well as the splitting between the singlet and triplet states in the dot.

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