Cotunneling current through a two-level quantum dot coupled to magnetic leads: the role of exchange interaction

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Abstract

The cotunneling current through a two-level quantum dot weakly coupled to ferromagnetic leads is studied in the Coulomb blockade regime. The cotunneling current is calculated analytically under simple but realistic assumptions as follows: (i) the quantum dot is described by the universal Hamiltonian, (ii) it is doubly occupied, and (iii) it displays a fast spin relaxation. We find that the dependence of the differential conductance on the bias voltage is significantly affected by the exchange interaction on the quantum dot. In particular, for antiparallel magnetic configurations in the leads, the exchange interaction results in the appearance of interference-type contributions from the inelastic processes to the cotunneling current. Such dependence of the cotunneling current on the tunneling amplitude phases should also occur in multi-level quantum dots weakly coupled to ferromagnetic leads near the mesoscopic Stoner instabilities.

1. Introduction

It is well known that the resistivity of a conductor depends on its magnetic state [1]. There are several quantum effects that increase this dependence, e.g., the Kondo effect [2], and colossal [3] and giant [4, 5] magnetoresistance effects. One can observe the latter in thin film structures composed of alternating ferromagnetic and non-magnetic layers. At room temperature the resistance for a parallel configuration of magnetizations in the leads is a few per cent less than the resistance for an antiparallel configuration. Thus, the spin degrees of freedom can considerably influence the magnetoresistance in the system.

Recently, the magnetic effect on the electron transport has attracted a lot of attention in different systems, and in particular in quantum dots (QDs) [6–16]. A QD is the simplest system in which one can study the interplay between spin and charge degrees of freedom. If the QD size is small enough, the characteristic electrostatic energy, necessary to change the number of electrons on the QD, can exceed the characteristic temperature and applied source–drain voltage (V). This regime of electron transport is referred to as Coulomb blockade. In this regime the processes of second order in tunneling amplitudes (sequential tunneling) are not suppressed only at specific values of the gate voltage (so-called Coulomb peaks) for which the electron QD charging levels become degenerate in the absence of tunneling [17]. Therefore, the current beyond the Coulomb peaks is determined by the cotunneling processes. These processes are of fourth order: tunneling of electrons from one lead to another through a virtual state on the QD [18, 19].

An additional factor, which strongly affects the current through a QD at low temperatures (T), is the discreteness of the energy spectrum [20]. For example, in the limit $T \gg \Delta$ the conductance due to inelastic cotunneling is proportional to $(e^2/h)(\Delta/E_c)^2$ [18, 19], where $\Delta$ denotes the single-particle spacing, $e$ is the electron charge, $h$ is the Planck constant, and $E_c = e^2/(2C)$ is the electrostatic energy of the QD with total capacitance $C$. In the opposite regime of low temperatures, $T \ll \Delta$, the (inelastic cotunneling) conductance is proportional to $(e^2/h)(\Delta^2/E_c^2 T) \exp(-\Delta/T)$, i.e., exponentially suppressed with temperature [21]. The same holds for the conductance due to elastic cotunneling. For high temperatures, $T \gg \Delta$, the
conductance is $\alpha (e^2/h)\Delta /E_c$ [18, 19] whereas for $T \ll \Delta$ it is proportional to $(e^2/h)(\Delta /E_c)^2$ [21]. We mention that the elastic cotunneling contribution dominates over the inelastic one at low temperatures.

The presence of the exchange interaction between the QD electrons results in the dependence of the conductance on the magnetic polarization of the leads. Recently, this dependence has been studied extensively in two-level quantum dots (2LQDs) [22]. In the paper [23] numerical solution of the rate equations describing transport through a 2LQD was employed.

As usual, in the rate equation approach [22, 23] it was assumed that the dominant relaxation process for electrons in a QD is their escape to the leads. This means that the QD electron distribution is fully determined by the distributions in the leads. In particular, at some finite source–drain voltage the QD electron distribution becomes non-equilibrium.

The aim of the present paper is to calculate analytically the cotunneling current through a 2LQD with the exchange interaction coupled to magnetic leads. In contrast to the previous works [22, 23], we assume that the dominant mechanism of relaxation of the electron distribution in the QD is governed by the electron–electron and electron–phonon interactions. Also we assume a fast spin relaxation in the QD, e.g., due to interaction with nuclear spins. In this case the QD electron distribution remains at equilibrium even in the presence of a finite source–drain voltage. We examine the interference and non-interference contributions to the cotunneling current and the contributions corresponding to elastic and inelastic processes. We obtain that at non-zero exchange energy ($J > 0$) the interference part of the cotunneling current for antiparallel magnetic alignment in the leads contains the inelastic terms which correspond to the transitions between QD states with different values of the total spin. Also we demonstrate that the low-temperature current asymptotics are similar to the results of [21] for a 2LQD without exchange interaction if we substitute $|\Delta - 2J|$ for the level spacing $\Delta$. At $J = \Delta/2$, when the transition from the singlet ($J < \Delta/2$) to the triplet ($J > \Delta/2$) ground state occurs, some of the inelastic processes become elastic. In addition, we find that the low-temperature current–voltage characteristics are non-linear. However, they become linear if the spin-triplet gap $|\Delta - 2J|$ vanishes. This fact can be used to observe the spin-triplet transition point experimentally. We have found that the inelastic cotunneling current has stairs corresponding to the transitions between the QD energy levels.

In general, contributions to the conductance of higher order (fourth and higher) in the tunneling amplitudes contain logarithmic divergences [24]. These divergences [25] are not relevant if $e|V|, T \gg T_K \sim \Delta \exp(-\Delta/\nu|t|_2^2)$, where $\nu$ and $t$ stand for the density of states in the leads and the tunneling amplitude, respectively. Thus, for $e|V|, T \gg T_K$, the cotunneling processes provide the main contribution to the current.

To observe the interplay of ferromagnetism and the discreteness of the energy spectrum in QDs, one should prepare a QD with a large level spacing. For ultra-small metallic islands (nanoparticles) [26] or in semiconducting QDs [27], the level spacing is comparable to the charging energy or even larger. This also happens in the case of a molecule attached to metallic leads [28] and in a two-dimensional (2D) electron gas with ferromagnetic leads [29].

This paper is organized as follows. We start with the formulation of the model describing the system under consideration (section 2). Then in section 3 we derive the general expression for the cotunneling current under several approximations which do not change the qualitative properties of the system. In section 4 we present the expressions for the non-interference and interference contributions to the cotunneling current. Finally, we present our discussion and conclusions in section 5.

2. The model

A QD coupled to the leads is described by the following Hamiltonian:

$$H = H_l + H_r + H_{QD} + H_T.$$  \hspace{1cm} (1)

Here, $H_i = \sum_{j=1, r, l} \epsilon_{\alpha\sigma} a_{\alpha\sigma}^\dagger a_{\alpha\sigma}$, $i = l, r$, are the Hamiltonians of the left and right leads, respectively, $a_{\beta\sigma}/a_{\beta\sigma}^\dagger$ denote the annihilation/creation operators in the corresponding leads, and $\sigma = \pm 1$ is the spin index. The term

$$H_T = \sum_{j=1, r, l} \sum_{k, l, \alpha, \sigma, \sigma} j_{k\alpha\sigma\sigma}^{(0)} a_{k\alpha\sigma}^\dagger d_{\alpha\sigma} + \text{h.c.}$$  \hspace{1cm} (2)

describes the tunneling between the QD and the leads, where $d_{\alpha\sigma}$ stands for the annihilation operator of the electrons on the QD. The QD is modeled by the so-called universal Hamiltonian [10]

$$H_{QD} = \sum_{\alpha, \sigma} \epsilon_{\alpha\sigma} d_{\alpha\sigma}^\dagger d_{\alpha\sigma} = E_c(\tilde{N} - N_0)^2 - JS^2.$$  \hspace{1cm} (3)

where $\tilde{N} = \sum_{\alpha, \sigma} d_{\alpha\sigma}^\dagger d_{\alpha\sigma}$ denotes the operator of the number of particles on the QD, $S = \sum_{\alpha\sigma} d_{\alpha\sigma}^\dagger d_{\alpha\sigma}$ is the operator of the total spin of the electrons, $N_0$ is the equilibrium number of electrons on the QD which minimizes the electrostatic energy and can be tuned by the gate voltage, and $J > 0$ is the exchange energy.

The main objective of this paper is an analytical investigation of the interplay between the spin and charge degrees of freedom in transport in the cotunneling regime. The simplest system revealing such effects is a QD with the two single-particle levels. Although Hamiltonian (3) is derived under the assumption that the number of QD levels involved in the transport is large [10], it is widely used to model a 2LQD [22, 23, 30, 31]. As we demonstrate in the appendix, Hamiltonian (3) can adequately describe the many-particle spectrum even for a QD with only two low-lying single-particle levels. In addition, Hamiltonian (3) appears in the effective description of a double QD involving spatially separated single-level QDs in which the exchange interaction is provided by small tunneling between the QDs [32].
In what follows, we consider a 2LQD, i.e., $\epsilon_\alpha$ will take only two values $\epsilon_1$ and $\epsilon_2 = \epsilon_1 + \Delta$. There are 16 (many-particle) eigen states. The energy of a state denoted by $|NSnS_\alpha\rangle$ is given by the following expression:

$$E_{NSnS_\alpha} = E_c(N - N_0)^2 + n\epsilon_1 + (N - n)\epsilon_2 - JS(S + 1).$$

(4)

Here, $N$ denotes the number of electrons on the QD, $n$ is the number of electrons on the first level, $S$ and $S_\alpha$ are the total spin and its projection on the z axis, respectively.

3. Contribution to the current of fourth order in $t_{ka,\sigma\sigma'}$

3.1. Perturbation theory

Let us assume that the Coulomb energy is much larger than the other energy scales, $E_c \gg T, \Delta, J$, and $N_0$ is close to the integer number (regime of a Coulomb valley). Then the main contribution to the current at low temperatures is of the fourth order in $t_{ka,\sigma\sigma'}$ [18, 19]. The second-order term corresponding to the real transitions changing the QD charge is exponentially suppressed ($\sim \exp(-E_c/T)$) for almost all the gate voltages with exception of those which correspond to the Coulomb peaks [17]. The contribution to the current of fourth order describes the electron transitions from lead to lead through the virtual QD states. These transitions are referred to as cotunneling of electrons.

The current operator is equal to the derivative of the charge operator in one of the leads:

$$\dot{\tilde{I}}_\sigma = e\tilde{N}_\sigma = i\tilde{X} - i\tilde{\dot{X}}^T, \quad \tilde{X} = \sum_{ka,\sigma\sigma'} t_{ka,\sigma\sigma'}^\dag a_{ka}^\dag a_{ka}. \quad (5)$$

Solving perturbatively the equation for the density matrix in the interaction representation to third order in tunneling (see e.g., [33]), we find the fourth-order correction in $t_{ka,\sigma\sigma'}^{(4)}$ to the current

$$\dot{I}_{\sigma\sigma'}^{(4)} = \frac{2e^2}{Z} \text{Re} \left\{ \int_{-\infty}^{+\infty} dt_1 \int_{-\infty}^{+\infty} dt_2 \int_{-\infty}^{+\infty} dt_3 \times \left[ (\tilde{X}^T(t)\tilde{H}_T(t_1)\tilde{H}_T(t_2)\tilde{H}_T(t_3)) \right. \right.$$}

$$- (\tilde{H}_T(t_3)\tilde{X}^T(t_1)\tilde{H}_T(t_2)\tilde{H}_T(t_3)) \right.$$}

$$- (\tilde{H}_T(t_1)\tilde{X}^T(t_2)\tilde{H}_T(t_1)\tilde{H}_T(t_3)) \right.$$}

$$+ (\tilde{H}_T(t_1)\tilde{H}_T(t_2)\tilde{X}^T(t_1)\tilde{H}_T(t_3)) \right.$$}

$$+ (\tilde{H}_T(t_1)\tilde{H}_T(t_2)\tilde{H}_T(t_1)\tilde{X}^T(t_3)) \right.$$}

$$+ (\tilde{H}_T(t_1)\tilde{H}_T(t_2)\tilde{H}_T(t_3)\tilde{X}^T(t_1)) \right.$$}

$$- (\tilde{H}_T(t_1)\tilde{H}_T(t_2)\tilde{H}_T(t_3)\tilde{X}^T(t_1)) \right\}, \quad (6)$$

where $Z \equiv \text{Tr}(\rho_0) = \text{Tr} \exp[-\beta(H_1 + H_T + H_{QD})]$ is the grand canonical partition function ($\beta = 1/T$) and $\langle \cdots \rangle \equiv \text{Tr}(\cdots | \rho_0)/Z$.

Let us express all of the averages in equation (6) in terms of the exact two-particle correlators for the isolated dot ($H_{QD}$) and the following electron Green’s functions in the leads:

$$G_{\beta_1\beta_2}^{<}(t, t') \equiv -i \langle a^{\dagger}_{\beta_1}(t) a_{\beta_2}(t') \rangle,$$

$$G_{\beta_1\beta_2}^{>}(t, t') \equiv i \langle a_{\beta_1}(t') a^{\dagger}_{\beta_2}(t) \rangle.$$  \quad (7)

Here we introduce $\beta_j = (\xi, \sigma_j)$. Each of the eight terms in equation (6) can easily be evaluated. For example, $X_1(t)H_T(t_1)H_T(t_2)H_T(t_3)$

$$= \langle d_{\alpha_1}^\dag d_{\alpha_2} d_{\alpha_3}^\dag d_{\alpha_4} \rangle \tau_{1324} G_{\beta_1\beta_2}^{<}(t, t_1) G_{\beta_3\beta_4}^{<}(t_2, t_3) \right.$$}

$$+ \langle d_{\alpha_1}^\dag d_{\alpha_2} d_{\alpha_3}^\dag d_{\alpha_4} \rangle \tau_{1324} G_{\beta_1\beta_2}^{<}(t, t_1) G_{\beta_3\beta_4}^{<}(t_2, t_3) \right.$$}

$$+ \langle d_{\alpha_1}^\dag d_{\alpha_2} d_{\alpha_3}^\dag d_{\alpha_4} \rangle \tau_{1324} G_{\beta_1\beta_2}^{<}(t, t_1) G_{\beta_3\beta_4}^{<}(t_2, t_3) \right.$$}

$$+ \langle d_{\alpha_1}^\dag d_{\alpha_2} d_{\alpha_3}^\dag d_{\alpha_4} \rangle \tau_{1324} G_{\beta_1\beta_2}^{<}(t, t_1) G_{\beta_3\beta_4}^{<}(t_2, t_3) \right.$$}

$$- \langle d_{\alpha_1}^\dag d_{\alpha_2} d_{\alpha_3}^\dag d_{\alpha_4} \rangle \tau_{1324} G_{\beta_1\beta_2}^{<}(t, t_1) G_{\beta_3\beta_4}^{<}(t_2, t_3) \right.$$}

$$- \langle d_{\alpha_1}^\dag d_{\alpha_2} d_{\alpha_3}^\dag d_{\alpha_4} \rangle \tau_{1324} G_{\beta_1\beta_2}^{<}(t, t_1) G_{\beta_3\beta_4}^{<}(t_2, t_3), \quad (8)$$

where $\langle \cdots \rangle = \text{Tr} \cdots \exp(-\beta H_0) \text{Tr} e^{-\beta H_{QD}}$, $\alpha_k = (\xi, \sigma)$, $\tau_{ijkl} = \sum_{\sigma_1, \sigma_2} \langle \sigma_1 \sigma_2 | \tilde{X}_i \tilde{X}_j \tilde{X}_k \tilde{X}_l | \sigma_1 \sigma_2 \rangle$, and $\Delta_\alpha = \text{Tr} \tilde{d}_\alpha$. We mention that equation (8) involves the terms which are proportional to $\tilde{p}_{\sigma_1} \tilde{p}_{\sigma_2} \tilde{p}_{\sigma_3} \tilde{p}_{\sigma_4}$ and $\tilde{p}_{\sigma_1} \tilde{p}_{\sigma_2} \tilde{p}_{\sigma_3} \tilde{p}_{\sigma_4}$. If we set, e.g., $t^\prime = 0$, the current should vanish. However, the terms proportional to $\tilde{p}_{\sigma_1} \tilde{p}_{\sigma_2} \tilde{p}_{\sigma_3} \tilde{p}_{\sigma_4}$ remain unchanged. Thus such terms give no contribution to the current in fourth order. Therefore we shall omit them in what follows.

Due to the presence of interactions in $H_{QD}$ the correlators of the form $\langle \tilde{d}_\alpha^\dag \tilde{d}_\alpha^\dag \tilde{d}_\alpha \rangle$ in equation (8) cannot be simplified with the help of Wick’s theorem. In general, the absence of Wick’s theorem leads to a very cumbersome expression for the current. Therefore, we introduce some simplifications which do not affect the qualitative properties of the system but simplify the analytical calculations.

3.2. Approximations

We calculate the fourth-order correction to the current given by equation (6) under the following assumptions. The leads are made of a ferromagnetic metal with the magnetization along some axis $z$. Considering the exchange interaction in the leads to be isotropic, we assume the Green’s function $G_{\xi k,\xi' l, \sigma_1 \sigma_2}$ of the electrons in the lead to be diagonal in the spin indices $\sigma_1, \sigma_2$. Next, we assume that there are no magnetic impurities in the tunneling junctions. Therefore, we neglect the probability for the spin to flip during a tunneling event, i.e.,

$$\tilde{I}_{\xi k,\xi' l, \sigma_1 \sigma_2}^{\langle 1 \rangle} = \tilde{I}_{\xi k,\xi' l}^{\langle 1 \rangle} \delta_{\sigma_1 \sigma_2}. \quad (9)$$

Also, since only the energies near the Fermi level in the leads are essential for the calculation of the current, we will ignore the energy dependence of the tunneling amplitudes and introduce the dimensionless tunneling conductances

$$\bar{G}_{\xi k,\xi' l}^{R} = \frac{1}{\pi} \sum_k \text{Im} G_{\xi k,\xi' l}^{R}(E_F)|\tilde{I}_{\xi k,\xi' l}^{\langle 1 \rangle}|^2.$$ \quad (10)

Finally, we restrict our consideration to the QD states with two electrons only. If $\Delta > 2J$, the spin in the ground state is zero (both electrons occupy the lowest energy level). In the opposite case of $\Delta < 2J$ the ground state is ferromagnetic, the total spin equals unity, and each energy level is singly occupied. Therefore, it is possible to observe a drastic effect in the transport through the QD due to the change of the spin
in the ground state. In what follows, we assume such a value of the gate voltage that the only important states of the system are the states with two electrons on the QD, i.e., \( N_0 \simeq 2 \).

In the current (6) one can distinguish terms of two types. The terms of the first type depend only on the absolute values of the tunneling amplitudes (\( |t_i| \), \( |t_f| \)). The contributions of the second type involve also the relative phases of the tunneling amplitudes. One can refer to corrections of the first type as non-interference and to the second type as interference contributions. To single out the effects associated with the current dependence on phases of the tunneling amplitudes, it is convenient to analyze the interference and non-interference contributions to the current separately. As is well known [18, 19], there are processes of two types: inelastic processes during which the QD energy changes and elastic ones with the same energies of the initial and final QD states. Due to conservation of the energy of the total system we have \( E_i + E_f = E_i + E_f \) where \( \epsilon_1, \epsilon_2 \) denote the energies of an electron before and after the tunneling event, and \( E_i, E_f \) are the energies of the initial and final QD states. By definition, the inelastic cotunneling involves a change of the QD energy: \( E_i \neq E_f \) (an electron–hole pair arises). This means that during the inelastic cotunneling the state of the QD changes, \( |i\rangle \neq |f\rangle \).

So, one can call this process incoherent. For the corresponding discussion of a single-level QD see, e.g., [34]. An elastic cotunneling process can either change the state of the QD, \( |i\rangle \neq |f\rangle \), e.g. by electron spin-flip, or not, \( |i\rangle = |f\rangle \). One can refer to the latter type of cotunneling as coherent. In what follows, each term in the current will be discussed according to the definitions introduced above.

4. Cotunneling current

4.1. General expression

Analyzing equation (6), we obtain the following expression for the current:

\[
p_{\text{min}}^{(4)} = \frac{2}{Z_f} \int \frac{d\epsilon_1}{(2\pi)^2} \frac{d\epsilon_2}{(2\pi)^2} \left[ (1 - n_F^\uparrow(\epsilon_1))(1 - n_F^\uparrow(\epsilon_2)) + (1 - n_F^\downarrow(\epsilon_1))(1 - n_F^\downarrow(\epsilon_2)) \right] \chi_{\downarrow \downarrow}(\epsilon_1, \epsilon_2) + \left( 1 - n_F^\uparrow(\epsilon_1) \right) \chi_{\uparrow \downarrow}(\epsilon_1, \epsilon_2) + \left( 1 - n_F^\downarrow(\epsilon_2) \right) \chi_{\downarrow \uparrow}(\epsilon_1, \epsilon_2) + \frac{1}{2} \left( n_F^\uparrow(\epsilon_1) n_F^\downarrow(\epsilon_2) \right) \chi_{\uparrow \uparrow}(\epsilon_1, \epsilon_2).
\]

(11)

Here, \( n_F^\sigma(\epsilon) \) stands for the Fermi–Dirac distribution of electrons in the left/right lead. The terms in equation (11) proportional to \( (1 - n_F^\uparrow(\epsilon_1))(1 - n_F^\downarrow(\epsilon_2)) \) and \( n_F^\uparrow(\epsilon_1) n_F^\downarrow(\epsilon_2) \) have no physical meaning. As a consequence, direct calculation yields that \( \chi_{\uparrow \uparrow} \) and \( \chi_{\downarrow \downarrow} \) vanish. The detailed expressions for \( \chi_{\downarrow \uparrow} \) and \( \chi_{\uparrow \downarrow} \) are cumbersome. They are related as \( \chi_{\downarrow \uparrow}(\epsilon_1, \epsilon_2) = -\chi_{\uparrow \downarrow}(\epsilon_2, \epsilon_1) \). As an example, we present an expression for a term in \( \chi_{\downarrow \uparrow}(\epsilon_1, \epsilon_2) \) that contributes to the non-interference part of the cotunneling current:

\[
\chi_{\downarrow \uparrow}(\epsilon_1, \epsilon_2) = -\Delta^2 \text{Re} \int_{-\infty}^{t_1} dt_1 \int_{-\infty}^{t_1} dt_2 \left( \chi_{\downarrow \uparrow}(\epsilon_1, \epsilon_2) \right)
\]

(12)

Next,

\[
Z_2 = e^{-\beta(\epsilon_1 + \epsilon_2)}(\epsilon_1^\Delta + e^{-\beta\Delta} + 3e^{2i\beta} + 1)
\]

(13)

is the canonical partition function for the Hamiltonian \( H_{QD} \) with \( N = 2 \) electrons. The analytical expressions for the non-interference and interference contributions to \( \chi_{\downarrow \uparrow} \) are presented below for antiparallel and parallel alignments of magnetizations in the leads.

4.2. Antiparallel configuration

The antiparallel configuration of magnetizations in the leads corresponds to the following values of \( g^{(\downarrow)} \):

\[
g_{1,\uparrow} = g_{2,\downarrow} = -g, \quad g_{1,\downarrow} = g_{2,\uparrow} = 0.
\]

(14)

This choice of values \( g^{(\downarrow)} \) assumes that the spin-down electron band of the left lead and the spin-up electron band of the right lead are empty. Here, for the sake of simplicity, we assume equal tunneling amplitudes for levels 1 and 2. In addition, we introduce phases \( \phi_f \) and \( \phi_i \):

\[
\tilde{t}_{1\uparrow} = [\tilde{t}_{1\uparrow}^\downarrow || \tilde{t}_{2\downarrow}^\downarrow ] e^{i\phi}, \quad \tilde{t}_{1\downarrow} = [\tilde{t}_{1\downarrow}^\downarrow || \tilde{t}_{2\uparrow}^\downarrow ] e^{i\phi}.
\]

In the antiparallel case we obtain (\( \phi = \phi_f - \phi_i \))

\[
\chi_{\Delta \uparrow}(\epsilon_1, \epsilon_2) = \frac{-4\pi \Delta^2 g^{(\downarrow)} g^{(\uparrow)}}{E_2^2} \times \\
\times (e^{i\phi}(\epsilon_1 + \epsilon_2 - 2J)[2\delta(\epsilon_1 - \epsilon_2)(1 + \cos \phi) + \delta(\epsilon_1 - \epsilon_2 - 2J)(1 - \cos \phi) + \delta(\epsilon_1 + \epsilon_2 - \Delta - 2J)] + \\
\times (\epsilon_1 - \epsilon_2 - \Delta + 2J) + e^{-2i\beta} \delta(\epsilon_1 - \epsilon_2 - 2J) + \\
\times (1 - \cos \phi) + e^{i\phi} \delta(\Delta - 2\phi) \delta(\epsilon_1 - \epsilon_2 - \Delta - 2J) + \\
e^{-2i\phi} \delta(\epsilon_1 - \epsilon_2 - \Delta - 2J).
\]

(16)

Each term in \( \chi_{\Delta \uparrow} \) has transparent physical interpretation since it can be written as \( \sim \exp(-E_i/T) \delta(\epsilon_i - \epsilon - E_i) \).

For example, the term proportional to \( \delta(\epsilon_1 - \epsilon_2) \) corresponds to elastic cotunneling which results in the QD transition
between the states \([2110]\) and \([211\bar{1}]\). Similarly, the term proportional to \(\delta(\epsilon_1 - \epsilon_2 - \Delta + 2J)\) in equation (16) describes inelastic cotunneling and corresponds to the transition of the QD from the state \([2110]\) to \([210]\). This transition can be realized through two virtual states: with three electrons and one electron on the QD. Under our assumption of large \(E_c\), both of them provide the same contributions to the current since the energies of the virtual states are equal to \(E_c\) with our accuracy. At non-zero value of the exchange energy \(J\), the terms proportional to \(\delta(\epsilon_1 - \epsilon_2 \pm 2J)\) correspond to the inelastic cotunneling but depend on the phase difference \(\phi\).

They describe the transitions between the states \([2110]\) and \([211 \pm 1]\) (see figure 1).

Employing equation (11), we find

\[
I_{\text{AP, in}}^{(\text{inel})} = -\frac{2\Delta^2 g^4 g^2}{\pi Z^2 E_c^2} e^{-\beta(\epsilon_1 + \epsilon_2 - 2J)} \left[2F(V)(1 + \cos \phi) + F(V + 2J)(1 - \cos \phi) + F(V + \Delta + 2J) + e^{-2\beta J} F(V - 2J)(1 - \cos \phi) + e^{\beta(-2\Delta - 2J)} F(V + \Delta - 2J) + e^{\beta(\Delta + 2J)} F(V - \Delta - 2J) - (V \rightarrow -V)\right]
\]

and

\[
I_{\text{AP, in}}^{(\text{el})} = \frac{4g^4 g^2 \Delta^2}{\pi E_c^2 Z^2} e^{-\beta(\epsilon_1 + \epsilon_2 - 2J)} V,
\]

respectively. Here we use the following relation: \(F(V) - F(-V) = -V\). The interference term of the cotunneling current is split into the inelastic and elastic parts as follows

\[
I_{\text{AP, in}}^{(\text{inel})} = \frac{2\Delta^2 g^4 g^2}{\pi Z^2 E_c^2} e^{-\beta(\epsilon_1 + \epsilon_2 - 2J)} [F(V + 2J) \cos \phi + e^{-2\beta J} F(V - 2J) \cos \phi - (V \rightarrow -V)],
\]

and

\[
I_{\text{AP, in}}^{(\text{el})} = \frac{4g^4 g^2 \Delta^2}{\pi E_c^2 Z^2} e^{-\beta(\epsilon_1 + \epsilon_2 - 2J)} V \cos \phi.
\]

As we have mentioned above, some inelastic transitions (e.g. from \([210]\) to \([211]\)) can be implemented through two different virtual states. The interference of these two processes depends on the phases of the tunneling amplitudes. Therefore, there is an interference term in the current which involves the inelastic contributions in the form \(F(\pm 2J + V)\) (see equation (20)). In the regime of low temperatures and voltages, \(|V|, T, |\Delta - 2J| \ll \Delta, J\), the inelastic terms of the interference contribution (20) are suppressed due to the small exponential factor \(\exp(-2J/T)\). In the case of \(J = 0\) the contribution \(I_{\text{AP, in}}^{(\text{inel})}\) becomes elastic and exactly compensates \(I_{\text{AP, in}}^{(\text{el})}\). Therefore, at \(J = 0\) the cotunneling current becomes independent of the phase \(\phi\).

In figure 2 we present the dependence of the differential conductance \(dI_{\text{AP}}^{(4)}/dV\) on \(T\) and \(V\) for \(\phi = \pi/2\) and different values of the exchange interaction \(J\). For low temperatures \(T \ll J\), a staircase structure in the differential conductance appears. It corresponds to step-by-step switching-on of different inelastic processes with increasing voltage. As follows from equation (17), one can expect some features at \(|V| = |\Delta - 2J|, 2J, \Delta + 2J\). However, some of the stairs have exponentially small heights at low temperatures and, therefore, are invisible, as shown in figure 2. At \(J = 0\) there is only one stair which corresponds to a change of the QD energy by \(\Delta E_{\text{QD}} = \Delta\). For small values of the exchange energy \(J \ll \Delta/2\), the feature at \(|V| = \Delta E_{\text{QD}} = \Delta - 2J\) is visible. In the regime \(\Delta - 2J \ll \Delta, J\), stairs corresponding to the processes with \(\Delta E_{\text{QD}} = \Delta + 2J, 2J\) and \(\Delta - 2J\) appear. As is expected, the latter disappears at \(J = \Delta/2\). All the three stairs survive at \(J > \Delta/2\). The evolution of the differential conductance \(dI_{\text{AP}}^{(4)}/dV\) with increasing \(J\) at a fixed temperature is shown in figure 3 for \(\phi = \pi/2\). At \(J > \Delta/2\) the feature corresponding to the inelastic process with \(\Delta E_{\text{QD}} = 2J\) disappears at \(\phi = 0\).

In the most interesting regime in the vicinity of the singlet–triplet transition, where \(\Delta = 2J - \kappa\) with \(|V|, T, |\kappa| \ll \Delta, J\), the expression for \(I_{\text{AP, in}}^{(\text{inel})}\) can be drastically simplified:

\[
I_{\text{AP, in}}^{(\text{inel})} = \frac{2g^4 g^2 \Delta^2}{\pi (3 + e^{E_c/E_c T}) E_c^2} \left[(V + \kappa) \frac{1 - e^{V/T}}{1 - e^{(V + \kappa)/T}} + (V - \kappa) \frac{1 - e^{-V/T}}{1 - e^{-(V - \kappa)/T}}\right].
\]

At \(\kappa = 0\) the current (22) acquires a simple form, \(I_{\text{AP, in}}^{(\text{inel})} = g^4 g^2 \Delta^2 V/\pi E_c^2\), since the spin-flip process becomes elastic.
Figure 2. The dependence of the differential conductance $dI/V$ on $V$ and $T$ for antiparallel ((a), (c), (e), (g), and (i)) and parallel ((b), (d), (f), (h), and (j)) configurations for different values of $J$. We use $g_l = 0.1$, $g_r = 0.2$, $E_c/\Delta = 10$, $\phi = \pi/2$ and $J/\Delta = 0.2$ for (a) and (b), $J/\Delta = 0.45$ for (c) and (d), $J/\Delta = 0.5$ for (e) and (f), $J/\Delta = 0.55$ for (g) and (h), and $J/\Delta = 0.95$ for (i) and (j).
This is the reason why the conductance does not vanish when \( V = 0 \). For \( |\kappa| \gg T \), the current \( I_{\text{AN,in}}^{(\text{inel})} \) becomes

\[
I_{\text{AN,in}}^{(\text{inel})} = \frac{2e\Delta^2 g^1 g^2}{\pi E_c^2} e^{-|\kappa|/T} \left( \frac{2}{|\kappa|} \sinh \frac{V}{T} + \left[ 1 - \cosh \frac{V}{T} \right] \right),
\]

for \( |V| < |\kappa| \),

\[
e^{\kappa/T} (V - |\kappa| \text{ sgn}(V)),
\]

for \( |V| > |\kappa| \),

where

\[
c_{\kappa} = \begin{cases} 
1/3, & \text{for } \kappa > 0; \\
1, & \text{for } \kappa < 0.
\end{cases}
\]

Expression (23) demonstrates an exponential suppression of conductance at low temperatures due to the spacing between the energy levels. In our case this is the spacing between the triplet and singlet energy levels, which is equal to \(|\Delta - 2J|\).

In the case of different tunneling amplitudes for levels 1 and 2, the contributions from different tunneling processes to the cotunneling current in equation (17) are multiplied by factors depending on the ratios \( \alpha = |t_{11}^{(11)}|^2 / |t_{11}^{(12)}|^2 \) and \( \beta = |t_{11}^{(12)}|^2 / |t_{11}^{(11)}|^2 \). In addition, in the interference part of the cotunneling current, one should substitute \( 2ab \cos \phi / (1 + a^2 b^2) \) for \( \cos \phi \). Therefore, in the case of different tunneling amplitudes for levels 1 and 2 the results for the cotunneling current remain qualitatively the same as for \( a = b = 1 \).

4.3. Parallel configuration

The parallel configuration of magnetizations in the leads corresponds to the following values of \( g^{1T} \):

\[
g^1_{1,1} = g^1_{2,1} = g^1_{1,1} = g^1_{2,1} = 0.
\]

This choice of the values of \( g^{1T} \) assumes that the spin-down electron bands of both leads are empty. In addition, we introduce phases \( \phi_1 \) and \( \phi_2 \):

\[
\begin{align*}
\hat{t}_{1,1}^{(1T)} & = |\vec{t}_{1,1}^{(11)}| |\vec{t}_{1,1}^{(12)}| e^{i\phi_1}, \\
\hat{t}_{1,1}^{(2T)} & = |\vec{t}_{1,1}^{(11)}| |\vec{t}_{1,1}^{(12)}| e^{i\phi_2}.
\end{align*}
\]

In this case we obtain

\[
\chi_{P,^<;^>}^{(\text{inel})}(\epsilon_1, \epsilon_2) = \frac{-2\pi \Delta^2 g^1 g^2}{E_c^2} e^{-\beta(\epsilon_1 + \epsilon_2 - 2J)} |\delta(\epsilon_1 - \epsilon_2)| \\
\times (1 + \cos \phi) + \delta(\epsilon_1 - \epsilon_2 - 2J) (1 - \cos \phi) \\
+ \delta(\epsilon_1 - \epsilon_2 - \Delta - 2J) + \delta(\epsilon_1 - \epsilon_2 + \Delta - 2J) \\
+ e^{-\beta J} [\delta(\epsilon_1 - \epsilon_2 + 2J) (1 - \cos \phi) \\
+ \delta(\epsilon_1 - \epsilon_2 - \Delta) + \delta(\epsilon_1 - \epsilon_2 + \Delta)] \\
+ e^{\beta J} [\delta(\epsilon_1 - \epsilon_2 - 2J) (1 - \cos \phi) \\
+ \delta(\epsilon_1 - \epsilon_2 - 2J)] (\epsilon_1 - \epsilon_2)] (1 - \cos \phi) \\
+ \delta(\epsilon_1 - \epsilon_2 + \Delta + 2J) + \delta(\epsilon_1 - \epsilon_2 + \Delta)] (1 - \cos \phi).
\]

Like in the case of the antiparallel alignment of magnetizations, each term in \( \chi_{P,^<;^>}^{(\text{inel})} \) has a transparent physical interpretation. As compared with \( \chi_{\text{AP,}^<;^>}^{(\text{inel})} \), equation (27) demonstrates that the parallel magnetizations permit more elastic processes. In addition, transitions in which the QD energy is changed by \( \Delta E_Q \), \( \pm \Delta \) are possible.

Using equation (11), we find

\[
I_P^{(\text{inel})} = -\frac{\Delta^2 g^1 g^2}{\pi Z_c E_c^2} e^{-\beta(\epsilon_1 + \epsilon_2 - 2J)} [F(V + 2J) + F(V - \Delta + 2J)] \\
+ F(V + \Delta + 2J) + e^{-\beta J} [F(V - \Delta - 2J) \cos \phi + F(V + \Delta)] \\
+ e^{\beta J} [F(V + \Delta - 2J) + F(V - \Delta)] \\
\times (V \rightarrow -V) \right) (V \rightarrow -V).
\]

The inelastic and elastic parts of the non-interference contribution to the cotunneling current are as follows:

\[
I_{\text{P,in}}^{(\text{inel})} = \frac{-\Delta^2 g^1 g^2}{\pi Z_c E_c^2} \left( F(V + 2J) + e^{-\beta(\epsilon_1 + \epsilon_2 - 2J)} \right) \\
+ (2 + e^{-\beta(\epsilon_1 + \epsilon_2 - 2J)}) V.
\]

The inelastic and elastic terms of the interference part of the cotunneling current are given by

\[
I_{\text{P,in}}^{(\text{inel})} = \frac{\Delta^2 g^1 g^2}{\pi Z_c E_c^2} e^{-\beta(\epsilon_1 + \epsilon_2 - 2J)} [F(V + 2J) + e^{-\beta J} F(V - 2J)] \cos \phi,
\]

and

\[
I_{\text{P,in}}^{(\text{el})} = \kappa \frac{\Delta^2 g^1 g^2 \Delta^2}{\pi Z_c E_c^2} e^{-\beta(\epsilon_1 + \epsilon_2 - 2J)} [F(V + 2J) + e^{\beta J} F(V - 2J)] \cos \phi - (V \rightarrow -V).
\]

In the expression (28) there are two types of additional term in comparison to the case of the antiparallel configuration. The first ones (\( \alpha F(V \pm \Delta) \)) correspond to the singlet–singlet transitions of the QD, e.g. [2020] \( \rightarrow [2010] \), during the inelastic cotunneling. They include a transfer of one electron to the other level. The terms of the second type (\( \alpha F(V) (1 - \cos \phi) \)) describe the elastic cotunneling due to transitions of the QD between the states in which one level is doubly occupied, e.g. [2020]. These elastic terms lead to the dependence of the cotunneling current for the parallel alignment of magnetizations on the phase difference \( \phi \) even.
at $J = 0$. The differential conductance $dI_p^{(4)}/dV$ as a function of $T$ and $V$ for $\phi = \pi/2$ is plotted in figure 2. As compared to the antiparallel case, there is an additional stair at $|V| = \Delta$ visible at $0 < J < \Delta/2$ (see figure 3) and corresponding to $\Delta E_{\text{QP}} = \Delta$.

In the most interesting regime near the singlet–triplet transition $\Delta = 2J - \kappa$ with $|V|, T, |\kappa| \ll \Delta, J$, the expression for $I_{\text{P, min}}^{(\text{inel})}$ can be written as

$$I_{\text{P, min}}^{(\text{inel})} = \frac{\Delta^2 g_1^4 g_2^4}{\pi E_0^2 (3 + e^{-\kappa/T})} \left( (V - \kappa) \frac{1 - e^{-V/T}}{1 - e^{-(-\kappa+V)/T}} + (V + \kappa) \frac{1 - e^{V/T}}{1 - e^{V/(\kappa+V)/T}} \right).$$

(33)

Note that in this regime $I_{\text{P, min}}^{(\text{inel})} = (1/2)I_{\text{AP, min}}^{(\text{inel})}$. In the case $|\kappa|, |V|, T \ll \Delta, J$ additional terms $\propto F(V \pm \Delta)$ are suppressed by the small factor $\exp(-\Delta/T)$ and, therefore, do not contribute to the current.

For different tunneling amplitudes for levels 1 and 2, the result (28) varies similarly to the antiparallel case and remains qualitatively the same.

5. Discussion and conclusions

In this paper the cotunneling current through a 2LQD coupled to ferromagnetic leads has been calculated analytically. The results have been presented for the most interesting case of a two-electron QD and for parallel and antiparallel magnetic configurations in the leads.

The inelastic cotunneling current has a singular behavior due to electron transitions between the QD’s energy levels. The singularities in the current characteristics can be used to determine the structure of the QD’s energy levels. In the cases of both parallel and antiparallel configurations the non-interference part of the conductance has a minimum at low temperatures and voltages $|V|, T \ll \Delta$. The width of this minimum near the transition ($\Delta = 2J$) between singlet and triplet ground states is determined by the gap between the singlet and triplet two-electron states $|\Delta - 2J|$. Near the transition our results for the differential conductance resemble the expression derived in the paper [21] for a 2LQD at $J = 0$ if we substitute the level spacing by the singlet–triplet gap $|\Delta - 2J|$. For low temperatures $T \ll |\Delta - 2J|$, the inelastic part of the current is suppressed by a factor of $\exp(-|\Delta - 2J|/T)$ in comparison with the elastic one. However, at temperatures $T \sim |\Delta - 2J| \ll \Delta$, the elastic and inelastic parts of the cotunneling current are of the same order of magnitude. This is because the spin-flip processes become almost elastic near the singlet–triplet transition in contrast to the case of $J = 0$.

As we mentioned in section 1, numerical calculations of the differential conductance based on a rate equation approach were performed in [23]. It was found that there is a zero-bias peak with a width of the order of $T$ in the antiparallel case (in contrast to the parallel one). This zero-bias peak has been explained by the non-equilibrium difference in the occupation probabilities of the states [211] and [211]. Such a peak is absent in our results since we assume that the electron distribution in the QD remains at equilibrium in the presence of a finite source–drain voltage.

However, we emphasize here that both the perturbation theory and the rate equation method produce the same results for the linear conductance and for the positions of the stairs in the differential conductance–voltage dependence.

In the regime $V, T \sim |\Delta - 2J| \ll \Delta$ only the singlet–triplet transitions are important. It is worthwhile to mention that in this regime the inelastic interference contributions to the cotunneling current are exponentially suppressed. Thus, in this regime expressions (23) and (33) for the inelastic part of the cotunneling current $I_{\text{inel}}^{(\text{cir})}$ are valid for a QD with a large number of levels. The elastic part of the cotunneling current $I_{\text{el}}^{(\text{cir})}$ in such QDs is determined by the transitions via the energy levels in the range $\sim E_0$. Therefore, the expression for $I_{\text{el}}^{(\text{cir})}$ in QDs with a large number of levels is $\sim E_0/\Delta$ times greater than $I_{\text{el}}^{(\text{cir})}$ for a 2LQD (see equation (19), (21), (30) and (32)). This allows us to go over to the result of elastic cotunneling in a multi-level QD.

In QDs with a large number of levels transitions between the ground states with $S$ and $S + 1$ are possible at $J = J_5 = \delta(2S + 1)/(2S + 2)$, where $\delta$ denotes the mean single-particle level spacing [10]. Our results indicate that in the vicinity of such transitions, at $|V|, T \ll |J - J_5| \ll J, \delta$, the inelastic part of the cotunneling current will be suppressed. However, exactly at the transition ($J = J_5$) the current $I_{\text{inel}}^{(\text{cir})}$ becomes linear in $V$ and temperature-independent. Therefore, the increase of conductance should occur at the transition point. One can use this fact in order to observe experimentally the transition between the ground states with $S$ and $S + 1$ in multi-level QDs.

It is worthwhile to note that the interference part of the cotunneling current (20) and (31) involves the terms corresponding to inelastic processes ($\propto F(eV \pm 2J)$). This results from the presence of the two-particle eigenstates [211] and [210] which admit inelastic transitions with different paths. As usual, it leads to a dependence of the probabilities of such transitions on the phases of the tunneling amplitudes. We recall that in the lack of the exchange
interaction only the elastic cotunneling terms depend on the phases of the tunneling amplitudes [18, 19]. In order to measure the interference part of the cotunneling current, one needs to change the phase difference $\phi_1 - \phi_4$. For reasons to be explained shortly, this seems impossible in a setup with a 2LQD. However, as we have mentioned above, the two-electron states of Hamiltonian (3) can be realized in a double QD involving two spatially separated single-level QDs. As is shown in [32], the exchange interaction between electron spins in different QDs will be mediated by small tunneling between the QDs. Recently, such a system was successfully operated experimentally [35, 36]. In this setup one can use the Aharonov–Bohm effect to control the phase difference $\phi_1 - \phi_4$, which will be proportional to the magnetic flux (see, e.g., [34]).

Our analytical results for the cotunneling current have been derived under the assumption that the dominant mechanism of relaxation of the electron distribution in the QD is governed by the electron–electron and electron–phonon interactions. For a multi-level QD one can estimate the relaxation rate due to the electron–electron interaction as [37, 38] $1/\tau_{ee} \sim \max[N_e^{-1}, g_{Th}(T/\delta)f_{\beta_l}(T/\delta)]$. Here, $N_e$ stands for the total number of electrons in the QD, and the Thouless conductance is determined by the ratio of the Thouless energy $E_{Th}$ to the mean level spacing $\delta$: $g_{Th} = E_{Th}/\delta$. The function $f_{\beta_l}(x)$ has the following asymptotic behavior: $f_{\beta_l}(x) \sim 1$ for $x \gg 1$ and $f_{\beta_l}(x) \sim 6\beta_l x$ for $x \ll 1$, where $\beta_l = 1$ for the Gaussian orthogonal ensemble, $\beta_l = 2$ for the Gaussian unitary ensemble and $\beta_l = 4$ for the Gaussian symplectic ensemble. The relaxation rate due to escape of electrons to the leads is of the order of $1/\tau_E \sim g^6$, where $g = g_1 + g_2$. Therefore, in the limit of small enough $g$ one can achieve the regime in which $1/\tau_{ee} \gg 1/\tau_E$. The relaxation rate in QDs due to electron–phonon interaction is not universal and depends on the type of phonon considered. For example, for a 2D ballistic QD in GaAs/Al$_x$Ga$_{1-x}$As heterostructure in which the dominant electron–phonon coupling is due to the piezoelectric interaction, the relaxation rate at $T \ll \delta$ can be estimated as [39] $1/\tau_{ee} \sim (\delta^3/\omega_{D}^4)(T/\delta)^{b/3}$, where $\omega_D$ stands for the Debye frequency. This indicates that at low temperatures the relaxation due to electron–phonon interaction is larger than that due to electron–electron interaction. The theoretical estimates [40, 41], which take into account the particular structure of the single-particle levels in a QD as well as experimental observations (see e.g., [42]), suggest that the electron–phonon interaction can provide the main relaxation mechanism in QDs. In few-electron QDs the numerical estimate of energy relaxation due to electron–phonon coupling [43] yields a relaxation time about 1 ns in agreement with experiments [44].

To justify our analytical results for the cotunneling current, we should also assume the fast relaxation of a QD spin to equilibrium. In general, there are three mechanisms for the spin relaxation: spin–orbit interaction, magnetic impurities and nuclear spins (see, e.g. [45]). Experimentally, for few-electron 2D QDs in heterostructures the dominant spin-relaxation mechanism is the interaction with the surrounding nuclear spins [46, 47]. The spin-relaxation time is very sensitive to the presence of a magnetic field and increases drastically at large magnetic fields. In the absence of a magnetic field, the typical time of spin relaxation is about 10–100 ns and is shorter than the time for electrons to escape to reservoirs which can reach 0.01 ms [47]. Therefore, our analytical results for the cotunneling current can be useful for experiments on few-electron QDs in 2D electron systems.

In this work we have treated the collinear magnetization in fully polarized ferromagnetic leads. In the general case of noncollinear magnetization in partially polarized leads we also expect the existence of the interference part of the inelastic contribution to the cotunneling current. The details will be published elsewhere.

To realize a 2LQD effectively, one can use any system with doubly degenerate levels and study it at voltages and temperatures much less than the level spacing. For example, it can be a carbon nanotube with orbital degeneracy of levels [31] or a 2D electron gas in a Si(001)-MOSFET [48] or Si/SiGe heterostructure [49]. In addition, one should observe the 2LQD behavior of the inelastic cotunneling current for an arbitrary QD in the regime $|V|, T \sim |J - J_3| \ll J, \delta$.

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Appendix. The general form of the Hamiltonian for a 2LQD

Although there is a vast body of studies on the spectra in few-electron QDs [50], in this appendix we present a general expression for the 2LQD Hamiltonian and discuss when it can be simplified to equation (3). We start from the following Hamiltonian:

$$H_{QD} = \sum_{\alpha,\sigma} \epsilon_\alpha d^\dagger_\alpha \sigma d_{\alpha \sigma} + H_{int}, \quad (A.1)$$

where $\alpha = 1, 2$ denotes the orbital single-particle levels and

$$H_{int} = \frac{1}{2} \sum_{\sigma_1, \sigma_2, \sigma_3} U_{\sigma_1 \sigma_2 \sigma_3 \sigma_4} d^\dagger_{\sigma_1 \sigma_1} d^\dagger_{\sigma_2 \sigma_2} d_{\sigma_3} d_{\sigma_4} \quad (A.2)$$

is the interaction part of the Hamiltonian. The matrix elements of the interaction are defined as

$$U_{\sigma_1 \sigma_2 \sigma_3 \sigma_4} = \int d\mathbf{r} d\mathbf{r}' \varphi^*_\sigma_1(\mathbf{r}) \varphi^*_\sigma_2(\mathbf{r}') U(\mathbf{r} - \mathbf{r}') \varphi^\dagger_\sigma_3(\mathbf{r}') \varphi^\dagger_\sigma_4(\mathbf{r}). \quad (A.3)$$

where $U(\mathbf{r}) = e^2/(\varepsilon \varepsilon r)$ is the Coulomb potential. Provided that the time-reversal invariance is conserved, only six matrix
elements $U_{1111}$, $U_{2222}$ and $U_{1212}$ describe the direct Coulomb interaction while $U_{1122}$ corresponds to the exchange energy.

Hamiltonian (A.1) commutes with the total number of electrons $\hat{N}$, the total spin square $\hat{S}_z$, and $\hat{S}_\perp$. Therefore, it is convenient to work in the basis of two-particle states $|N_S n_S\rangle$. Then $H_{QD}$ can be written as a $16 \times 16$ matrix. All the states except those with $N = 2$, $S = 0$, $S_z = 0$ and with $N = 3$, $S = \pm 1/2$, $S_z = \pm 1/2$ are eigenstates of Hamiltonian (A.1).

Their energies are

$$E_{0000} = 0, \quad E_{1\frac{1}{2} 0\frac{1}{2}} = \frac{1}{2} (\epsilon_1 + \epsilon_2), \quad E_{1\frac{1}{2} 1\frac{1}{2}} = \frac{1}{2} (\epsilon_1 + \epsilon_2), \quad E_{2\frac{1}{2} 0\frac{1}{2}} = \frac{1}{2} (\epsilon_1 + \epsilon_2), \quad E_{2\frac{1}{2} 1\frac{1}{2}} = \frac{1}{2} (\epsilon_1 + \epsilon_2).$$

The states $|3\frac{1}{2} 2\frac{1}{2}\rangle$ and $|3\frac{1}{2} 1\frac{1}{2}\rangle$ are also mixed and Hamiltonian (A.1) projected onto these states can be written as

$$H_2 = (\epsilon_1 + \epsilon_2) \mathbf{1} + V_2,$$

where $V_2$ is given by

$$V_2 = \left( \begin{array}{ccc} \epsilon_1 + U_{1111} & -U_{1122} & \epsilon_1 + U_{1212} \\ -U_{1111} & \epsilon_1 + U_{1122} & -U_{1212} \\ -U_{1122} & -U_{1222} & -\epsilon_1 + U_{1212} \end{array} \right).$$

As an example, let us consider a QD fabricated in a 2D electron gas in the Si(001)-MOSFET structure [48]. In such a QD the electrons can occupy the states in two valleys which remain from six-fold degeneracy of the bulk Si. Assuming the size-quantization level spacing to be large as compared with the valley splitting, we have only two low-energy orbital states (symmetric and anti-symmetric):

$$\psi_1(r) = \sqrt{2} \cos \frac{Qz}{2} \phi_0(z) \phi_\perp(\mathbf{p}),$$

$$\psi_2(r) = \sqrt{2} \sin \frac{Qz}{2} \phi_0(z) \phi_\perp(\mathbf{p}).$$

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