Kondo tunneling through a biased double quantum dot

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Electron tunneling through a system formed by two coupled quantum dots in a parallel geometry is considered within a generalized Anderson model. The dots are assumed to have nearly equal radii but different (and tunable) gate voltages. In the absence of tunneling to and from the leads, the isolated two-dot system (with two electrons in it) resembles an hydrogen molecule within the Heitler London approximation. In particular, it has a singlet ground state and low lying excited triplet state. When tunneling is switched on, and when the gate voltages are properly tuned the ground state becomes a triplet and singlet and triplet states are intermixed. In the region where charge fluctuations are suppressed, the pertinent antiferromagnetic exchange interaction has the form \( (J^T S + J^{ST} P) \cdot s \). It is written in terms of the electron spin \( s \), the double dot spin 1 operator \( S \) and an additional vector operator \( P \). The operators \( S \) and \( P \) generate the algebra \( o_4 \) of a spin rotator. The related Kondo effect is similar to that of a vertical quantum dot, discussed and analyzed recently.

I. INTRODUCTION

Experimental discovery of resonance Kondo tunneling through quantum dots under strong Coulomb blockade is an impressive recent result in the physics of nanostructures. The observation of Kondo-like zero bias anomaly in the tunneling current through planar GaAs/GaAlAs quantum dot (QD) with odd electron occupation and a net spin \( S = 1/2 \) confirmed earlier theoretical predictions. A natural question then arose whether the Kondo tunneling is possible when the number of electrons in the dot is even, so that the nominal spin of an isolated QD is zero. Some experimental data were consistent with the occurrence of Kondo resonance also in that case. Of course, the simple reason for Kondo scattering in this case is the triplet ground state which can be realized provided the exchange interaction compensates the energy \( \delta_e \) of excitation of one electron from the last doubly occupied state. Another compensation mechanism is the Zeeman splitting of excited triplet state in external in-plane magnetic field \( B \). In this case Kondo resonance in electron tunneling should be observed at a specific value of Zeeman energy \( E_Z = g\mu_B B = \delta_e \). Unlike the conventional Kondo mechanism, this effect arises under a condition of broken rotational invariance in spin space, and the tunneling induced singlet-triplet mixing is shown to be the source of Kondo-like resonance. The experimental observation of Kondo tunneling in even Coulomb blockade windows at \( B = 1.36 \) T in QD formed in nanotube confirmed these theoretical predictions.

Another interesting possibility of realization of Kondo tunneling in QD with even electron occupation arises when the low-energy part of its spectrum consists of spin singlet, spin triplet and singlet charge transfer exciton. It was shown in the above paper that such spectrum arises in double quantum dots (DQD) provided this kind of nanostructure consists of two wells of different depth and the tunneling is allowed only through the "shallow" well with larger radius and smaller Coulomb charging energy \( Q \). In this case, the singlet-triplet crossover is an intrinsic property of nanoobject in a contact with metallic leads, which arises even at zero magnetic field. This artificial molecule has a natural prototype: the low-energy part of the electron spectrum of complex molecules in which a rare-earth ion is secluded in a carbon cage has the above mentioned structure provided the covalent chemical bonds exist between the \( f \)-electron of a rare-earth ion and \( p \)-electrons of carbon cage. Cerocene Ce(CsHs)2 is a known example of such double-shell molecule. Being adsorbed on a metallic substrate this molecule is expected to demonstrate the Kondo-type behavior.

II. EFFECTIVE SPIN HAMILTONIAN FOR BIASED DQD

In the present research we study the tunneling through DQD formed by two dots of nearly equal radii in a parallel geometry (Fig.1) with two separate gates generating voltages \( V_{g,r} \). Such setups were fabricated several years ago.
If only one of the tunnel channels (say, left) between the dot and the lead is open (the inter-dot tunneling $V = 0$, the tunnel coupling between the right dot and the leads $W_r = 0$), we are left with the "electrometer" configuration when the tunneling through the left well is controlled by the charge state in the right well. The Coulomb blockade windows between the resonances in the Coulomb energy of the dot $E_{\nu_r, \nu_l}(V_{rg}, V_{lg})$ form a honeycomb pattern where the vertices connect the windows with charge configurations $(\nu_r, \nu_l), (\nu_r, \nu_l - 1), (\nu_r + 1, \nu_l - 1)$. The lines $E_{\nu_r, \nu_l} \approx E_{\nu_r+1, \nu_l}$ are the regions where the Coulomb resonance induced by $V_{rg}$ allows tunneling through the left dot [9].

We consider the tunneling through DQD where the source – drain current is allowed only through the left dot, but the tunnel coupling with the right dot controls the spin degrees of freedom of the DQD. The case of DQD occupied by two electrons in its ground state is studied assuming that both left and right dots are neutral at $\nu_r = \nu_l = 1$. Then the isolated DQD under strong Coulomb blockade conditions reminds, in some respect, the hydrogen molecule in the Heitler-London approximation. This system is described by a generalized Anderson tunneling Hamiltonian

$$H = \sum_{i=l,r} \sum_{\sigma} \epsilon_i n_{i\sigma} + V \sum_{i \neq j} d_{i\sigma}^\dagger d_{j\sigma} + \frac{1}{2} \sum_i Q_i n_i(n_i - 1)$$

$$+ \sum_{k\sigma\alpha} \varepsilon_{k\sigma\alpha} c_{k\sigma\alpha}^\dagger c_{k\sigma\alpha} + \sum_{k\sigma} \left(W_{ki} c_{k\sigma\alpha}^\dagger d_{i\sigma} + H.c.\right)$$

(1)

Here the quantized energy levels of electrons in the dots $\epsilon_i = \varepsilon_i + V_{gi}^2$ are biased by the gate voltages, the Coulomb blockade energies are assumed to be the same for both dots, $Q_l = Q_r = Q$, $\varepsilon_{k\sigma\alpha}$ are the band energies of the electrons in the leads, $\alpha = s, d$ stands for electrons from the source and drain electrodes, $c_{k\sigma} = 2^{-1/2}(c_{k\sigma, s} + c_{k\sigma, d})$, $W_{ki} = W_{k\alpha, i}/(W_{k\alpha, s}^2 + W_{k\alpha, d}^2)^{1/2}$.

We are interested in the case when the artificial Heitler-London molecule is strongly biased by the gate voltages so that $E_{2,2} - E_{1,2} \gg E_{1,2} - E_{1,1}$, but still $\beta \equiv V/(E_{1,2} - E_{1,1}) \ll 1$ (Fig.2).
FIG. 2. Electrostatic energy of the right ($r$) and left ($l$) dot as a function of electron occupation numbers $\nu_r$ and $\nu_l$.

In this case the dot Hamiltonian (first three terms in eq. (1)) may be easily diagonalized, and the lowest three levels are

$$E_S = \epsilon_l + \epsilon_r - 2\beta V, \quad E_T = \epsilon_l + \epsilon_r, \quad E_R = 2\epsilon_r + Q + 2\beta V$$

The small parameter is $\beta = V/(Q - \epsilon_l + \epsilon_r)$ expressed in terms of the coupling parameters entering the Hamiltonian (see Fig. 2). The gate voltage $V_g - V'_g$ is applied in such a way that the spin and charge degrees of freedom are nearly separate in isolated DQD, i.e. the energy of charge transfer exciton $E_R - E_S = Q - \epsilon_l + \epsilon_r + 4\beta V$ substantially exceeds the energy of spin exciton $E_T - E_S = 2\beta V$.

If the tunnel coupling between the DQD and the metallic leads is also small, $W_l/(E_R - E_S) \ll 1$, only the singlet-triplet excitations are involved in forming the tunnel transparency of the left dot. In spite of the occurrence of singlet ground state, the Kondo-like processes involving the triplet exciton are possible, and one can expect the corresponding anomalies provided the characteristic Kondo temperature $T_K$ is comparable with the energy $2\beta V$ of singlet-triplet excitation in DQD [7]. To study the low-energy anomalies we use the Haldane-Anderson renormalization group (RG) approach [10]. According to this method the Hamiltonian $\mathcal{H}$ diagonalized in accordance with (2) is rescaled by absorbing the high-energy excitations of order $D_0$ (where $D_0$ is the width of conduction band) in the renormalized parameters of this Hamiltonian at reduced $D = D_0 - \delta D$. It is known that one can neglect renormalization of the tunneling constant $W_l$ in comparison with rescaling of the energy levels $E_S$ and $E_T$. The scaling equations are

$$dE_\Lambda/d\ln D = \Gamma_\Lambda/\pi,$$

where $\Lambda = S, T$. The tunnel coupling constants $\Gamma_\Lambda = \pi\rho_0|\langle 0|c_{k\sigma}d_{k\sigma'}|\mathcal{H}|\Lambda \rangle|^2$ are different for singlet and triplet states because the singlet exciton ($\rho_0$ is the density of states in the leads). In other words, the doubly occupied right dot state $|ex_r\rangle = d_{r\uparrow}^\dagger d_{r\downarrow}^\dagger |0\rangle$ is admixed to the bare singlet $|s\rangle = \frac{1}{\sqrt{2}} \sum_\sigma d_{l\sigma}^\dagger d_{r\bar{\sigma}}^\dagger |0\rangle$ by inter-dot tunneling, whereas the triplet states $|t_0\rangle = \frac{1}{\sqrt{2}} \sum_\sigma d_{l\sigma}^\dagger d_{r\bar{\sigma}}^\dagger |0\rangle, |t_\pm\rangle = d_{l\sigma}^\dagger d_{r\sigma}^\dagger |0\rangle$ are not affected by this tunneling,

$$|S\rangle = (1 - \beta^2)|s\rangle + \sqrt{2}\beta|ex_r\rangle, \quad |T0\rangle = |t_0\rangle, \quad |T\pm\rangle = |t_\pm\rangle.$$

As a result $\Gamma_S/\Gamma_T = 1 - 2\beta^2 < 1$. Due to this relation the scaling trajectories $E_\Lambda(D)$ determined by the scaling invariants for eq. (3) may intersect at a point $D_c$ estimated as

$$\frac{\Gamma_T - \Gamma_S}{\pi} \ln \frac{\pi D_0}{D_c} = E_T(D_0) - E_S(D_0) \equiv \delta_0.$$
According to numerical calculations of Ref. [4], this level crossing can occur either before or after the crossover to a Schrieffer-Wolff regime when the one-electron energies $E_{\alpha}(D) - E_{\beta b}$ exceed the half-width of reduced continuum band, $\bar{D} \sim |E_{\alpha}(\bar{D}) - E_{\beta b}|$. In both cases the charge degrees of freedom are quenched for the excitation energy within the interval $-\bar{D} < \varepsilon < \bar{D}$. Then, integrating out the residual charge excitations (tunneling transitions to the states with one and three electrons in the DQD) one comes to an effective Hamiltonian including only spin degrees of freedom,

$$\bar{H} = \sum_{\Lambda=S,T} \bar{E}_{\Lambda}X^{\Lambda\Lambda} + \sum_{(k)\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + \sum_{\Lambda\Lambda'kk'\sigma\sigma'} J^{\Lambda\Lambda'} X^{\Lambda\Lambda'} c_{k\sigma}^\dagger c_{k'\sigma'},$$

where $\bar{E}_{\Lambda} = E_{\Lambda}(\bar{D})$, $J^{\Lambda\Lambda'} \approx 2\sqrt{|\Lambda\Lambda'|}/(\varepsilon_F - \bar{\varepsilon}_l)$, $X^{\Lambda\Lambda'} = |\Lambda\rangle \langle \Lambda'|$ is a configuration change operator, $\bar{\varepsilon}_l = \varepsilon_l(\bar{D})$ includes the above RG operation, $(k)$ are the states inside the reduced conduction band.

Unlike conventional Schrieffer-Wolff Hamiltonian for a local spin that arises as a result of RG procedure \([14]\), the effective Hamiltonian \([8]\) contains also the singlet term with renormalized energy $\bar{E}_S$ and the effective interaction with coupling constant $J^{ST}$ intermixing singlet and triplet states of DQD. The coupling constants are connected by the relations $J^S = \alpha^2 J^T$, $J^{ST} = \alpha J^T$ ($\alpha = 1 - \beta^2$). This means that the Heitler-London type DQD possesses the symmetry of a spin rotator, i.e. that its algebra $o_4$ is described by two vectors $S$ and $P$ with spherical components

$$S^+ = \sqrt{2} (X^{10} + X^{01}), \quad S^- = \sqrt{2} (X^{01} + X^{-10}), \quad S_z = X^{11} - X^{-1, -1},$$

$$P^+ = \sqrt{2} (X^{1S} - X^{S, -1}), \quad P^- = \sqrt{2} (X^{S1} - X^{-1, S}), \quad P_z = - (X^{0S} + X^{S0})$$

obeying the commutation relations

$$[S_j, S_k] = i\epsilon_{jkl} S_l, \quad [P_j, P_k] = i\epsilon_{jkl} S_l, \quad [P_j, S_k] = i\epsilon_{jkl} P_l$$

($j, k, l$ are Cartesian coordinates). These vectors are orthogonal, $S \cdot P = 0$, and the Casimir operator is $S^2 + P^2 = 3$.

In terms of these operators the interaction term in the effective Hamiltonian acquires a symmetric form,

$$\bar{H}_{int} = J^T (S \cdot s) + J^{ST} (P \cdot s) + J^T 2 \sum_{\mu\nu} X^{\mu\nu}_{\mu} n_{\sigma},$$

where the local electron operators are defined as

$$n_{\sigma} = c_{\sigma}^\dagger c_{\sigma} = \sum_{kk'} c_{k\sigma}^\dagger c_{k'\sigma}, \quad s = 2^{-1/2} \sum_{kk'} \epsilon_{k\sigma}^\dagger \hat{\tau} c_{k'\sigma'},$$

($\hat{\tau}$ is the Pauli matrix).

### III. KONDO TUNNELING

If the inequality $\bar{D} < D_c$ is satisfied, the triplet becomes the lowest state of the DQD, i.e., $\bar{\delta} = \bar{E}_T - \bar{E}_S < 0$, and a Kondo screening for $S = 1$ should emerge. However, the Anderson-type scaling equations for the parameters $J(D)$ involve both $J^T$ and $J^{ST}$. In dimensionless variables $j_1 = \rho_0 J^T, j_2 = \rho_0 J^{ST}, d = \rho_0 D$ these equations are written as

$$dj_1/d\ln d = - [(j_1)^2 + (j_2)^2], \quad dj_2/d\ln d = -2j_1j_2.$$ 

Such quasi degeneracy of triplet and singlet states was discussed previously in relation to the physics of tunneling through vertical quantum dots [11], planar QD occupied by even number of electrons [3] and strongly asymmetric DQD containing two dots of essentially different radii [4]. In the first two cases the singlet-triplet degeneracy is induced by an external magnetic field, while in the third case the asymmetry is an intrinsic property of the DQD. In the case under consideration here, the desired asymmetry is induced by the asymmetric gate voltage. Analysis of the scaling equations [9] shows that both $S$ and $P$ operators are involved in anomalous Kondo scattering in a case when $\bar{\delta} < T_K$. Then the scaling equations [3] are reduced to a single equation for the reduced exchange parameter

$$j_+ = j_1 + j_2$$

$$dj_+/d\ln D = -(j_+)^2$$

with a fixed point at $j_+ = \infty$ and the Kondo temperature $T_{K0} = \bar{D} \exp(-1/j_+)$. This is a Kondo temperature of a spin rotator, having the rotational symmetry $SO(4)$. The spin symmetry of DQD is reduced together with the Kondo
temperature in the case $\delta \gg T_K$. Then the singlet-triplet coupling $j_2$ is quenched at energies $D \sim \tilde{\delta}$, so the low-energy spectrum is determined by the $S=1$ triplet state. The Kondo temperature at large $\delta$ is a function of $\delta$ which obeys the law $T_K/T_{K0} = (T_{K0}/\bar{\delta})^\lambda$, where $\lambda$ is a universal numerical constant [5].

The Kondo-type zero bias anomaly in conductance arises when $\delta < 0$ and the DQD is in a state with $S=1$. It grows with temperature as $G \sim \ln^{-2} [T/T_K(\bar{\delta})]$ for $|\delta| \gg T \gg T_K(\bar{\delta})$ and as $G \sim \ln^{-2} [T/T_{K0}]$ for $T \gg T_{K0} \gg \bar{\delta}$. At $T \to 0$ the conductance tends to the unitarity limit $G_0 = 2e^2/\pi\hbar$.

Thus one arrives at the following picture of rearrangement of the low-energy spectrum of DQD occupied by two electrons in a parallel geometry (Fig. 1) under external bias. In a symmetric case $V^l_g = V^r_g$ the isolated system possesses an axial symmetry and a singlet ground state (Heitler-London type artificial molecule). Its spin spectrum consists of singlet-triplet excitations, and the singlet state is stabilized by the indirect exchange energy $E_T - E_S = V^2/Q$ (Fig. 3a). Two singlet charge excitons (even and odd) corresponding to symmetric and asymmetric combination of the polar states $d_{i\uparrow}^\dagger d_{i\downarrow}^\dagger |0\rangle$ ($i = l, r$) are separated by a large energy gap $Q$ from the spin exciton. The level renormalization [6] is the same for the singlet and triplet states because the charge transfer excitations are axially symmetric in this case. The only possibility to open a Kondo channel is achievable by a trivial switching off the interdot tunneling ($V \to 0$). Then the singlet-triplet gap also tends to zero, and when it becomes less than $T_K$ for the left dot, the latter behaves as conventional $S=1/2$ QD with odd occupation. The axial symmetry is broken when $V^l_g - V^r_g > 0$, and the "right" exciton $E_R$ then softens (Fig. 3b).

The system acquires the symmetry of a spin rotator described by the semi-simple Lie group $SO(4)$ with triplet exciton mixed with the singlet ground state due to the tunneling interaction with the leads. When the condition [5] and the inequality $\tilde{D} < D_c$ are valid, the ground state of DQD becomes a triplet and it behaves as an under-screened $S=1$ Kondo center with zero bias anomaly of the conductance. Thus, the charge excitations in the right dot that is not involved directly in the source-drain tunneling can drive the spin excitations in the left dot and induce the Kondo resonance in a DQD with even occupation.

FIG. 3. Electron levels of double quantum dot at zero (a) and non-zero (b) value of $V^r_g - V^l_g$.

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