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Transport through quantum dots with magnetic impurities

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We analyze the electronic transport through a quantum dot that contains a magnetic impurity. The coherent transport of electrons is governed by the quantum confinement inside the dot, but is also influenced by the exchange interaction with the impurity. The interplay between the two gives raise to the singlet-triplet splitting of the energy levels available for the tunneling electron. In this paper, we focus on the charge fluctuations and, more precisely, the height of the conductance peaks. We show that the conductance peaks corresponding to the triplet levels are three times higher than those corresponding to singlet levels, if electronic correlations are neglected (for non-interacting dots, when an exact solution can be obtained). Next, we consider the Coulomb repulsion and the many-body correlations. In this case, the singlet/triplet peak height ratio has a complex behavior. Usually the highest peak corresponds to the state that is lowest in energy (ground state), regardless if it is singlet or triplet. In the end, we get an insight on the Kondo regime for such a system, and show the formation of three Kondo peaks. We use the equation of motion method with appropriate decoupling.

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1. Introduction

The quantum dots are zero-dimensional structures (so called artificial atoms) that are possible to be experimentally obtained and investigated individually, for some years now. An incoming electron may tunnel through a quantum dot only if its energy matches a dot energy level, so we have a single electron transistor [1]. Another interesting potential application is quantum computing using the spin of the quantum dot as information carrier [2]. The parameters that influence transmittance through a quantum dot (like the size of the dot or the hybridization with the leads) are much more easy to control comparing to the parameters of a bulk system, so one can test quantum theories in a way that was not possible before. We give as example the observation of the two-channel Kondo effect, described theoretically many years ago [3, 4], but realized experimentally only very recently, in a double-dot system [5] .

In the last years, people have started to address the problem of quantum dots with magnetic impurities [6, 7, 8, 9, 10, 11, 12], which is also the subject of the present paper. The transient electron feels the exchange interaction with the impurity and forms singlet/triplet entangled states before tunneling out of the dot. We analyze the way in which this physics is seen in transport. It will be shown that the peaks corresponding to the triplet levels are three times higher than those corresponding to singlet levels, if we neglect the Coulomb repulsion and electronic correlations in the dot. In large dots, with low confinement, the Coulomb repulsion is reduced, and the approximation to neglect it may still capture some important physics. Such a case will be addressed in Section II.

The case of small quantum dots, with strong Coulomb repulsion is treated in Section III. If the Coulomb interaction and the many-body correlations are considered, the picture becomes more complex: the peak corresponding to the ground state is usually higher, regardless if it is singlet or triplet. The peaks height depends on the ratio J/Γ (exchange interaction versus coupling strength with the leads). An analytical formula is proposed for the transmittance and in particular for the singlet/triplet peak height ratio. In the end, we get an insight on the Kondo regime and show the formation of three Kondo peaks in the density of states.

2. Large quantum dots with magnetic impurities

In this section we shall neglect all electron-electron interactions, except for the interaction with the magnetic impurity. The correlations are neglected as well, and they will be considered in the next section. The problem presented in this section is a two-electron scattering problem (one of which -the impurity- is fixed and may only change its spin orientation).

If correlations are neglected, one can easily afford to consider a many-site dot, which is realistic from geometrical point of view. In (large) quantum dots with low confinement, the Coulomb repulsion is reduced, and this justifies the non-interacting model used here. A general lattice Hamiltonian, with a localized spin interaction, can be written as:

$$H_S = \sum_{\langle i,j\rangle,\sigma} w_{ij,\sigma} e^{i2\pi\Phi_{ij}} c^{\dagger}_{i,\sigma} c_{j,\sigma} + \sum_{i,\sigma} V_g c^{\dagger}_{i,\sigma} c_{i,\sigma} + \frac{1}{2} J(c^{\dagger}_{n\uparrow} c_{n\uparrow} - c^{\dagger}_{n\downarrow} c_{n\downarrow}) S^z_n + \frac{1}{2} J(c^{\dagger}_{n\uparrow} c_{n\downarrow} S^-_n + c^{\dagger}_{n\downarrow} c_{n\text{-}\uparrow} S^+_n); n \in QD, \quad (1)$$

where $c^{\dagger}_i$ (c_i) are creation (annihilation) operators in the dot sites indexed by i ; the index n is devoted to the site of the dot where the magnetic impurity is placed. $w_{ij,\sigma}$ are hopping parameters (actually the hopping parameters are spin-independent and in the following we give up the spin index; moreover, they are considered only between nearest neighbors $\langle i, j\rangle$) and Φ_{ij} are the phases associated with an (eventually applied) magnetic field. The last two term account for the exchange interaction with the magnetic impurity [13].

From this point on, we take S=1/2. One can define the fermionic operators $\{d^{\dagger}_{\uparrow}, d^{\dagger}_{\downarrow}, d_{\uparrow}, d_{\downarrow}\}$ for the localized spin (provided one projects out all the states with occupancy different from 1)

Then, $S^z_n, S^+_n and S^-_n$ in eq (1) can be written as:

$$S^z_n = \frac{1}{2}(d^{\dagger}_{\uparrow} d_{\uparrow} - d^{\dagger}_{\downarrow} d_{\downarrow}), S^+_n = d^{\dagger}_{\uparrow} d_{\downarrow}, S^-_n = d^{\dagger}_{\downarrow} d_{\uparrow}. \quad (2)$$

The natural formulation of the problem is in terms of singlet-triplet operators (see for instance [14]). Let us introduce the singlet operator Σ_i and the three triplet operators T^p_i (p=1,2,3):

$$\Sigma_i = \frac{1}{\sqrt{2}}(d_{\uparrow} c_{i\downarrow} - d_{\downarrow} c_{i\uparrow}), T^1_i = \frac{1}{\sqrt{2}}(d_{\uparrow} c_{i\downarrow} + d_{\downarrow} c_{i\uparrow}), \quad T^2_i = d_{\uparrow} c_{i\uparrow}, T^3_i = d_{\downarrow} c_{i\downarrow}. \quad (3)$$

The above operators may be used in order to write the Hamiltonian (Eq.1) in the following way (where we have used also $n_d = d^{\dagger}_{\uparrow} d_{\uparrow} + d^{\dagger}_{\downarrow} d_{\downarrow} = 1$):

$$H_S = \sum_{\langle i,j\rangle} (w^{\Sigma}_{ij} \Sigma^{\dagger}_i \Sigma_j + w^{T}_{ij} \sum_{p=1}^{3} T^{p\dagger}_i T^p_j),$$
$$\omega^{\Sigma}_{ij} = \omega_{ij} e^{i2\pi\phi_{ij}} + \delta_{ij} V_g - \frac{3}{4} J \delta_{ij} \delta_{in}$$
$$\omega^{T}_{ij} = \omega_{ij} e^{i2\pi\phi_{ij}} + \delta_{ij} V_g + \frac{1}{4} J \delta_{ij} \delta_{in} \quad (4)$$

In order to analyze transport properties, one needs to connect leads to the quantum dot. The leads are modeled by a one-dimensional chain, consistent with the dot tight-binding model. For the non-interacting dot (more precisely: the interaction is only with the magnetic impurity, and other electron-electron interactions and correlations are neglected), the introduction of leads is equivalent to the introduction of a complex selfenergy in the Hamiltonian, at the contact sites. The procedure is described in detail in [8]. It results the effective Hamiltonian:

$$H^{eff}_S = H_S + \sum_{\alpha,\sigma} t^2_{\alpha} e^{-iq} c^{\dagger}_{\alpha,\sigma} c_{\alpha,\sigma}$$
$$= \sum_{\langle i,j\rangle} [(\omega^{\Sigma}_{ij} + \delta_{i\alpha}\delta_{j\alpha}\tau^2_{\alpha} e^{-iq}) \Sigma^{\dagger}_i \Sigma_j + (\omega^{T}_{ij} + \delta_{i\alpha}\delta_{j\alpha}\tau^2_{\alpha} e^{-iq}) \sum_{p=1}^{3} T^{p\dagger}_i T^p_j]$$
$$= H^{Eff}_{\Sigma} + \sum_{p=1}^{3} H^{eff}_{T_p} \quad (5)$$

We describe briefly the next steps. First, we employ the Landauer-Buttiker formula to connect the transmission through the dot with the retarded Green function [8] :

$$T_{\sigma,\sigma' S,S'}(E) = 4t^4_0 \sin^2 q \, |\langle \alpha, S | G^{+}_{\sigma\sigma'}(E) | \alpha', S' \rangle|^2 . \quad (6)$$

The above formula assumes equal coupling strength to the leads (τ_0) connected with the sites (states) α, α'. "*S*" stands for the spin state of the magnetic impurity that is changed to "*S*'" after the scattering. The electron spin is also changed from σ to σ' and only the transitions that conserve the total spin give non-zero transmittances. The parameter "*q*" is the impulse of the incident electron (see [8] and references therein for details about the dispersion formula in the leads and connection with the Landauer-Buttiker formalism).

At this point we remember that, for a single electron case, the retarded Green function is equal to the resolvent, and can be calculated by:

$$G^{+}(E) = (E - H^{eff})^{-1}. \quad (7)$$

The following definitions are introduced :

$$G^{+}_{\Sigma}(E) = (E - H^{eff}_{\Sigma})^{-1},$$
$$G^{+}_{T_p}(E) = (E - H^{eff}_{T_p})^{-1} \quad (8)$$

All triplet Green functions are equal (we give up the index "*p*") and the transmittance can be written:

$$T_{total} = \sum_{\sigma,\sigma' S,S'} T_{\sigma,\sigma' S,S'}(E) = 4t^4_0 \sin^2 q (\frac{1}{4} |G^{+}_S(E)|^2 + \frac{3}{4} |G^{+}_T(E)|^2). \quad (9)$$

We show in Fig.1 the transmittance peaks for a quantum dot modeled by a 3x5 two-dimensional lattice. The

exchange interaction causes the singlet-triplet splitting of the spectrum and the conductance peaks. The triplet peaks have the hight 3/4 and the singlet peaks 1/4. For a realistic 2D dot, certain eigenstates may be *zero* at the impurity position. Obviously, the corresponding transmittance peaks will not split. This case is also captured in Fig.1 (the "degenerate peaks" do not undergo the singlet-triplet splitting).

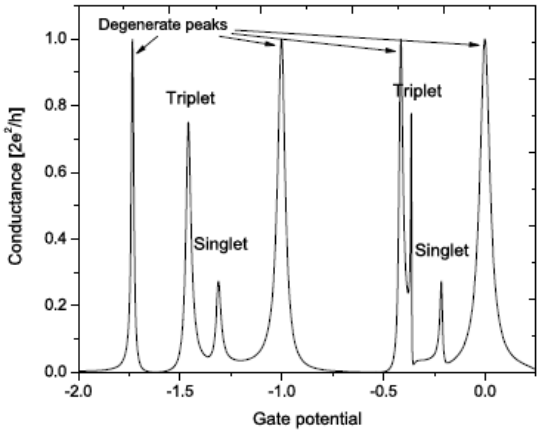

Fig. 1. The conductance through a many-site quantum dot with a magnetic impurity, in the absence of electron-electron interaction (only the interaction with the magnetic impurity is considered) . Some peaks split into triplet and singlet, with the heights 3/4 and 1/4, respectively. The peaks corresponding to trajectories that avoid the impurity do not split and remain degenerate. We show in Fig.1 the transmittance peaks for a quantum dot modeled by a 3×5 two-dimensional lattice. The exchange interaction causes the singlet-triplet splitting of the spectrum and the conductance peaks. The triplet peaks have the hight 3/4 and the singlet peaks 1/4. For a realistic 2D dot, certain eigenstates may be zero at the impurity position. Obviously, the corresponding transmittance peaks will not be splitted. This case is also captured in Fig. 1 (the "degenerate peaks" do not undergo the singlet-triplet splitting).

3. Small quantum dots with magnetic impurities

We saw in the previous section that the non-interacting dot is easy to be modeled by a *n*×*m* lattice, but the problem becomes (technically) much more difficult when we include the Coulomb interaction. A general Hamiltonian, that includes the Coulomb interaction between any two electrons, can be written: : $H = H_S + \sum_{i,j,\sigma,\sigma'} U_{i,j} c^{\dagger}_{i,\sigma} c_{i,\sigma} c^{\dagger}_{j,\sigma'} c_{j,\sigma'}$. There are several ways to address such a complex Hamiltonian, that allows only approximate solutions with considerable difficulty. In this section, we shall restrict to the case of a single-site dot with Coulomb interaction and exchange interaction with a magnetic impurity. In comparison to the Hamiltonian Eq.1, we shall introduce the Coulomb interaction, but also we shall write explicitly, from the beginning, the Hamiltonian of the leads and the leads-dot hopping term. The reason is the following: in the previous section we used the effective Hamiltonian trick, meaning that leads are introduced by a selfenergy term in the coupling sites. But this is no longer possible if we consider electronic correlations or the Kondo effect, when the role of the leads is more complex.

We re-write the Hamiltonian, in a convenient notation:

$$H = \sum_{k,\sigma,\alpha} \varepsilon_k c^{\dagger}_{k\sigma,\alpha} c_{k\sigma,\alpha} + \varepsilon_0 \sigma \sum_{\sigma} c^{\dagger}_{0\sigma} c_{0\sigma} + U c^{\dagger}_{0\uparrow} c_{0\uparrow} c^{\dagger}_{0\downarrow} c_{0\downarrow} + \frac{1}{2} \mathrm{J}(\mathrm{c}^{\dagger}_{0\uparrow} \mathrm{c}_{0\uparrow} - \mathrm{c}^{\dagger}_{0\downarrow} \mathrm{c}_{0\downarrow}) \mathrm{S}^{z} + \frac{1}{2} \mathrm{J}(\mathrm{c}^{\dagger}_{0\uparrow} \mathrm{c}_{0\downarrow} \mathrm{S}^{-} + \mathrm{c}^{\dagger}_{0\downarrow} \mathrm{c}_{0\uparrow} \mathrm{S}^{+}) + + \sum_{\mathrm{k},\sigma,\alpha} \mathrm{t}_{\alpha} (\mathrm{c}^{\dagger}_{0\sigma} \mathrm{c}_{\mathrm{k}\sigma,\alpha} + \mathrm{h.c.}), \tag{10}$$

where the first term represents the electrons in the lead $\alpha=L, R$. The second term stands for the electronic level ε_0 in the dot. The next four terms describe interactions: the Coulomb interaction of electrons with the opposite spin orientation at the level ε_0 and exchange interactions with the magnetic impurity. The last term in the Hamiltonian (Eq.10) corresponds to the coupling between the quantum dot and the leads. It shall be considered, for simplicity, $t_L = t_R = t$.

Now, we want to determine the conductance for the model described by the Hamiltonian (Eq.10). The current can be expressed by means of the non-equilibrium Green functions as [15, 16] :

$$j = \frac{2e}{h} \Gamma \int dw [f_L(w) - f_R(w)] (-\mathrm{Im} \langle\langle c_{0\uparrow} \mid c^{\dagger}_{0\uparrow} \rangle\rangle), \tag{11}$$

where $\Gamma=2\pi t^2\rho$ and $\rho=1/2D$ is the DOS for the square band approximation; the half-width D will be taken as unity. f_L and f_R are the Fermi distribution functions in the left and right leads, respectively. $\langle\langle c_{0\uparrow} \mid c^{\dagger}_{0\uparrow} \rangle\rangle$ is the retarded single particle Green function for an electron with the spin $\sigma=\uparrow$ at the QD, which can be determined by the equation of motion (EOM). The generic EOM for the energy dependent retarded Green function is given by

$$\omega\langle\langle A|B\rangle\rangle = \langle\{A,B\}\rangle + \langle\langle [A,H]|B\rangle\rangle , \tag{12}$$

where $\langle\{A,B\}\rangle$ is the thermal average of the anticommutator between the operators A and B. The procedure for solving the EOM for the retarded green functions was described in detail in [12]. Basically, the equations of motion introduce higher order Green functions, which result by performing the commutation (with the Hamiltonian) required by the formula above. For a finite system, even in the presence of interactions, one can write a finite number of independent equations and the system closes (even if sometimes the number of equations

is very big, it is still solvable). But for our case, interacting system + infinite leads, one has an infinite set of equations, and approximations are needed to close the system.

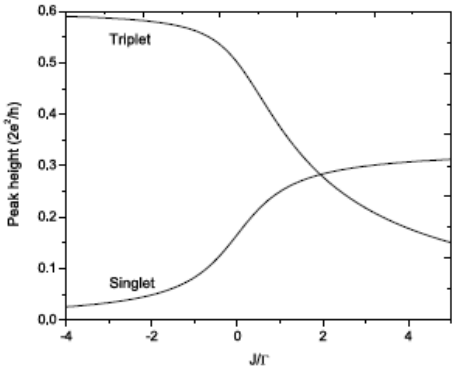

Fig. 2. The height of the singlet and triplet peaks, that depend on the ratio J/Γ (exchange interaction versus coupling strength with the leads), if the Coulomb interaction and electronic correlations are considered.

We propose the next approximation (that is valid at high temperatures [17])

$$2t\sum_k << c_{k,\sigma} A \,|\, c^{\dagger}_{0,\sigma} >> \approx -i\Gamma << c_{0,\sigma} A \,|\, c^{\dagger}_{0,\sigma} >> \tag{13}$$

It results a system of six equations (see [12]). The following solution is obtained for the conductance through the singlet and triplet peaks :

$$G = G_S + G_T = \frac{1+2\Phi_T}{2(3+4\Phi_S-4\Phi_S\Phi_T)} R_S + \frac{3(1+2\Phi_S)}{2(3+4\Phi_S-4\Phi_S\Phi_T)} R_T, \tag{14}$$

where $R_S = (2e^2/h)\Gamma^2/[\Gamma^2 + (E_F - e_0 + 3J/4)^2]$ and $R_T = (2e^2/h)\Gamma^2/[\Gamma^2 + (E_F - e_0 - J/4)^2]$ corresponds to the resonant conductance through the singlet and the triplet level, respectively, $\Phi_S = \arctan[(\varepsilon_0 - E_F - 3J/4)/\Gamma]/\pi$ and $\Phi_T = \arctan[(\varepsilon_0 - E_F + J/4)/\Gamma]/\pi$. The fractions that multiply the resonances $R_{S(T)}$ contain the information about the height of the conductance peaks. The ground and excited states role changes with the sign of J, and the dependence on the coupling also changes, being much more pronounced for the excited state. All these effects are incorporated in the above formula.

By employing further the simplified form Eq.(14), one can estimate the ratio between the heights of the conductance for the singlet and triplet peaks. We notice that in Eq.(14), the parameter Φ_S vanishes at the singlet resonance (found with the condition $\varepsilon_0 - E_F - 3J/4 = 0$). Φ_T vanishes at the triplet resonance. Therefore, one can straightforward calculate the ratio of the conductance peaks

$$\frac{\max[G_S]}{\max[G_T]} = \frac{(1+2\Phi_0)(3-4\Phi_0)}{9(1-2\Phi_0)}. \tag{15}$$

where $\Phi_0 = \arctan(J/\Gamma)/\pi$. Fig. 2 gives a graphical image of the peaks height dependence on the parameter J/Γ. For the antiferromagnetic coupling, the electronic transmission through the singlet state dominates and, in the limit $J/\Gamma >> 1$, this ratio goes to infinity and the triplet peak disappears. In the case of the ferromagnetic coupling and $|J|/\Gamma >> 1$, we have opposite situation - the singlet peak disappears from transport and only the peak corresponding to the triplet ground state is visible.

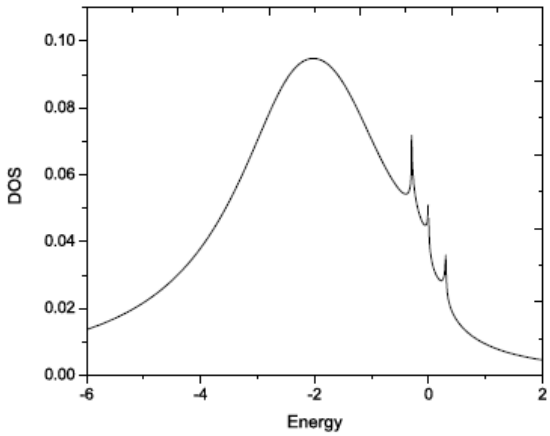

Fig. 3. The density of states for a quantum dot with a magnetic impurity, in the Kondo regime. Notice the formation of three Kondo peaks, one at the Fermi energy (E_F=0) and two side peaks at a distance equal to t he exchange interaction (J=0.3) .

In the last part of the paper, we give a very brief analysis of the Kondo regime for the impurity dot. It is known that a Kondo peak in the density of states, at the Fermi energy, is present only if a degenerate level exists below the Fermy energy. This would allow for fluctuations (in the "classical" Kondo one has spin fluctuations), that involve also the electrons from the leads which, for this reason, can pass through the dot with increased probability. In our case, such a degenerate level would be the triplet and the system shows the central Kondo peak (at the Fermi energy). This central peak is important for transport, because at low temperature and low bias, practically only the electrons around the Fermy level contribute to the conductance.

On the other hand, the singlet-triplet level structure should allow also the observation of side Kondo peaks, similar to the case of an applied magnetic field [20]. We

obtain a three-peaks Kondo structure and the typical density of states is plotted in Fig.3. The calculations were performed following the decoupling proposed in [18, 19] (see [12] for calculations details) for temperatures closed to the Kondo temperature.

4. Conclusions

In this paper, we present two models of quantum dots with magnetic impurities. The first model is a many-site lattice model (large quantum dots). It neglects the Coulomb interaction and many-body correlations, but can be used to model realistic geometries. In the absence of the magnetic impurity, the transmittance shows peaks with the height 1 (perfect transmittance). If a magnetic impurity is placed in the dot, one has triplet peaks with height 3/4 and singlet peaks with height 1/4. Some eigenstates inside the dot may avoid the impurity position, and the corresponding peaks do not split into singlet and triplet.

The second model we propose is a single-site dot with Coulomb and exchange interactions. Here we give up the geometrical aspects, in order to focus on the correlations effects. Such a model is realistic for small quantum dots, where the Coulomb is strong and the level-spacing is large. The singlet and triplet conductance peaks are shown to depend on the ratio J/Γ (the exchange interaction versus the coupling strength with the leads). In particular, the transmission through the excited states depends strongly on the coupling to the leads.

In the Kondo regime, the presence of the S=1/2 magnetic impurity generates three Kondo peaks in the density of states.

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