Kondo screening regimes of a quantum dot with a single Mn ion

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(Dated: January 11, 2013)

We study the Kondo and transport properties of a quantum dot with a single magnetic Mn ion connected to metallic leads. By employing a numerical renormalization group technique we show that depending on the value of ferromagnetic coupling strength between the local electronic spin and the magnetic moment of the Mn, two distinct Kondo regimes exist. In the weak coupling limit, the system can be found in a completely screened Kondo state describing a local magnetic moment decoupled from the rest of the system. In contrast, in the strong coupling regime the quantum dot spin and the local magnetic moment form a single large-spin entity partially Kondo screened. A crossover between these two regimes can be suitably tuned by varying the tunnel coupling between the quantum dot electron and the leads. The model investigated here is also suitable to study magnetic molecules adsorbed on a metallic surface. The rich phenomenology of these systems is reflected in the conductance across the system.

PACS numbers: 72.20-l, 73.23.Hk, 75.50.Pp, 71.21.La, 72.10.-d, 73.21.La, 75.50.Xx

Spin manipulation of localized impurities is of great interest in spintronics and quantum computation[1]. In this context, diluted magnetic semiconductor quantum dots (DMSQDs) could play a prominent role as they allow the control of the spins of the magnetic ions[2, 3]. In general DMSQDs are grown in II-VI semiconductor composites with a few Mn atoms in each quantum dot (QD)[3]. In these systems, the coupling between the spins of the electrons in the QD and those of the manganese arises from the sp – d exchange interaction.

More recently, the successful fabrication of QDs doped with a single Mn$^{2+}$ ion[3–7] has stimulated many optical and transport measurements[8–10], demanding a great deal of theoretical efforts[11–14]. Recent investigations of these systems have uncovered many interesting physical phenomena[6–8, 10, 15–18]. For instance, the exchange interaction makes single photon emitters active at six different frequencies, thus serving as the basic framework for the six-state qubit[12]. In this context, a very exotic system composed of an “impurity” with spin degrees of freedom coupled to a QD containing electrons (the impurity is outside the QD) has been proposed and studied, recently[15]. Fewer theoretical works, however, have addressed the transport properties in these systems[12].

In this work we investigate the low temperature properties of a quantum dot with a single magnetic Mn ion connected to leads. The study could be applied as well to analyze a magnetic molecules containing sites connected to independent leads. The ideas have this general scope, to be concrete, we restrict our discussion to a system composed of a Mn$^{2+}$ ion implanted in a small QD, coupled to two metallic (source and drain) leads, schematically represented in Fig. 1. It is well known that a QD connected to leads possesses a Kondo ground state similarly to what happens in magnetic impurities embedded in metals under temperature below the characteristic Kondo temperature ($T_K$)[21]. At the same time the electrons in the QD couple to the Mn$^{2+}$ magnetic moment by a ferromagnetic exchange interaction, $J$, that can be optically or electrically tuned[14, 15]. The antiferromagnetic case will be discussed in detail elsewhere[23]. In our case, $T_K$ can be modified by tuning the hopping matrix element $V$ that connects the localized and the lead states, while $J$ in turn can be tailored by properly choosing the size of the QD[18].

Based on a numerical renormalization group (NRG) technique[24], our theoretical study shows two-distinct Kondo regimes: 1) $T_K/|J| \gg 1$, where the QD spin is completely screened by the conduction spins comprising a Kondo state and the Mn$^{2+}$ is decoupled from the rest of the system and 2) $T_K/|J| \ll 1$, in which the spins of the electrons in the QD strongly couples to the Mn$^{2+}$ spin, forming a large-spin local magnetic impurity that is partially screened by the conduction electrons: the underscreened Kondo state. A crossover between these two regimes is achieved by suitably tuning the parameters of the system. Our system is described by the Hamiltonian

$$H = H_{\text{imp}} + H_{\text{bands}} + H_{\text{F}}$$

where

$$H_{\text{imp}} = \sum_{\sigma} \varepsilon_{d\sigma} c_{d\sigma}^\dagger c_{d\sigma} + U n_{d\uparrow} n_{d\downarrow} + J \mathbf{M} \cdot \mathbf{s}$$

(1)

corresponds to the single-level QD and the Mn$^{2+}$ ion, in which the operator $c_{d\sigma}^\dagger (c_{d\sigma})$ creates (annihilates) an electron of spin $\sigma$ with energy $\varepsilon_d$, $U$ is the local Coulomb repulsion and $\mathbf{M}$ and $\mathbf{s}$ are the spin operators of the Mn$^{2+}$ ion and of the QD, respectively. The Hamilto-
H_{bands} = \sum_{\ell k \sigma} \varepsilon_{\ell k} c_{\ell k \sigma}^\dagger c_{\ell k \sigma}

describes the conduction band, where \(c_{\ell k \sigma}^\dagger\) (\(c_{\ell k \sigma}\)) creates (annihilates) an electron with momentum \(k\), energy \(\varepsilon_{\ell k}\) and spin projection \(\sigma\) in the \(\ell\)-th lead (\(\ell = L, R\)). The conduction bands are characterized by a constant density of states given by \(\rho_0(\omega) = (1/2D)\Theta(D - |\omega|)\), where \(D\) is the half bandwidth and \(\Theta(x)\) is the heaviside function. Finally, \(H_T = \sum_{\ell k \sigma} V_{\ell \ell' d\sigma} c_{\ell k \sigma}^\dagger c_{\ell' k' \sigma} + h.c.\) describes the coupling between electrons in the QD and reservoirs.

Within the NRG framework we are able to calculate the relevant physical quantities, such as the entropy, the local magnetic moment and the local retarded spectral function at the QD site. The latter is necessary to calculate the zero-bias conductance.

Here we mainly focus on investigating the particle-hole symmetric case \((\varepsilon_d = -U/2)\) and normal leads, unless otherwise stated. We take \(D\) as the energy unity. In order to illustrate clearly the underlying physics of the numerical results presented latter, we start off by briefly discussing a simplified model, i.e., an isolated impurity \((V = 0\) in our model\) which will help us to understand the results for the full system. In a single-electron QD, the spin is \(s = 1/2\). The coupling of the spin of the QD electron with the spin of the Mn\(^{2+}\) leads to a total angular momentum \(L = M + s\), with a total angular momentum quantum number, \(l = |M - s|, \ldots, |M + s|\). For the electron-Mn\(^{2+}\) complex, the possible values of \(l\) are 2 and 3. Assuming a ferromagnetic coupling \(J < 0\), the ground state has \(l = 3\) with degeneracy \(2l + 1 = 7\), corresponding to the projections of the total angular momentum \(L\) along the \(z\)-axis, \(l_z = -3, -2, \ldots, 2, 3\).

We start our numerical analysis by studying the impurity contribution to the entropy and the magnetic moment, defined, respectively, as \(S(T) = S_t(T) - S_0(T)\) and \(\mu^2(T) = k_B T [\chi_0(T) - \chi_0(0)]\), where \(\chi\) is the magnetic susceptibility. The subscripts \(t\) and \(0\) refer to the quantities calculated for the entire system and in the contribution of the conduction band alone, respectively. We calculate these quantities within the usual NRG methodology \([26]\). Similarly, we calculate the spin-spin correlation \(\langle M \cdot s \rangle\) an important quantity to characterize the system regime.

In this paper we assume \(U = 0.5\) and set \(k_B = \mu_B = h = 1\). In Fig. 2a and 2b we show, respectively, the effects of the ferromagnetic exchange interaction and temperature on the magnetic moment and entropy for \(J = -2.0 \times 10^{-5}\) and various values of \(V\). Since the Kondo temperature \(T_K\), evaluated in the absence of ferromagnetic exchange interaction, is strongly dependent on \(V\), and can be tuned by changing \(V\). As a reference, the \(J = 0\) curve is also depicted (dashed green lines).

No exchange interaction. Notice in Fig. 2a that at high temperature \(T > \varepsilon_d\) \(S \rightarrow 1.77 \log(6) \approx \log(24)\), which indicates that there are \(6 \times 4 = 24\) individual states, 6 from the Mn spin and 4 from the dot, (spin and charge degrees of freedom), that can be thermally activated at that temperature. As the temperature decreases, the entropy presents a plateau at \(\log(12)\), indicating that for \(T \ll U/2\) the dot charge degrees of freedom are frozen. When the temperature decreases below \(T_K\), the entropy for \(J = 0\) tends to \(\log(6)\) (Fig. 2a), indicating that the 6 degenerate spin states of Mn is the only contribution to the entropy because the QD electron and conduction electrons are locked into a Kondo singlet.

Finite ferromagnetic \(J\) case. Here the competition between the exchange interaction energy \(J\) and \(T_K\) determines the ground state of the system. The ground state is a Kondo singlet (KS) for \(|J| < T_K\) with an uncoupled Mn ion and it becomes a local ferromagnetic state (LFS) as \(|J| \gg T_K\) due to the Mn-dot spin coupling that creates a large spin underscreened by the conduction electrons. Here we use the expression \(T_K = \sqrt{\pi U} \exp[-\pi U/8T]\) to estimate the Kondo temperature of the system in the absence of the Mn atom (or \(J = 0\)), with \(\Gamma = \pi U^2/\bar{D}\) being the hybridization constant. In the intermediary case \((T_K \sim J)\) the system presents a crossover region between the regimes mentioned above. In the following we...
analyze in detail these several regimes. 

Small-\( J \) regime – \( |J| \ll T_K \). In the regime of very weak exchange-interaction, when \( T \ll T_K \), the regular full screened KS state emerges. When \( T < T_K \), as the temperature decreases, the KS ground state is formed by the strongly coupled dot and conduction electron spins. The characteristic of this regime is clearly illustrated in the curve for \( T_K/|J| = 10^2 \) (\( \square \) symbols in Fig. 2a, where almost no plateau at \( S = \text{Log}(7) \) is observed. The entropy goes directly to the \( S = \text{Log}(6) \) plateau. The screening of the QD-electron spin by the conduction electrons leaves the Mn-ion free. Hence the magnetic moment of the system is only due to the 6-fold degenerate state of the Mn atom, which at \( T = 0 \) gives \( \mu^2 = 2((-5/2)^2 + (-3/2)^2 + (-1/2)^2)/6 = 35/12 \approx 2.92 \), as clearly seen in Fig. 2b (\( \square \) curve). In order to confirm this observation, in Fig. 2c we show the spin-spin correlation \( \langle M \cdot s \rangle \) as a function of the temperature using the same parameters as in Fig. 2a. Notice that for a given \( T_K/|J| \), as the temperature decreases, the correlation rapidly increases, becoming constant for \( T \lesssim |J| \), indicating the formation of a large effective localized spin.

Intermediate regime – \( J \sim T_K \). In this case the system is in a crossover region between the two previous analyzed regimes. The amplitude of this region can be seen very clearly from Fig. 2d where we show \( \langle M \cdot s \rangle \) vs. \( |J|/T_K \) for three different values of \( T_K \). Notice that the correlation rapidly increases for \( |J|/T_K \lesssim 1 \) and saturates slowly for larger values of \( |J|/T_K \), achieving the value 5/4 for \( J/|T_K| \to \infty \). The region where the correlation changes rapidly corresponds in the parameter space to the crossover region. An inspection of the figure permits to conclude that the relevant parameter that controls the moment correlations is the quantity \( J/T_K \) as we obtain the same universal function for the different values of \( T_K \) taken.

The QD density of states, \( \rho_d(\omega) = -\pi^{-1} \text{ Im}[G_{dd}^r(\omega)] \), where \( G_{dd}^r(\omega) \) is the Fourier transform of the double time retarded Green’s function, is calculated adopting standard NRG procedures. Within the same framework the zero-bias conductance \( G \) across the QD is calculated using the Green function formalism with the Landauer-type formula

\[
G = \frac{2e^2}{h} \int_{-\infty}^{\infty} \text{ Im}[G_{dd}^r(\omega)] \cdot (-\partial f(\omega)/\partial \omega) \ d\omega ,
\]

where \( f(\omega) \) is the Fermi function.

FIG. 3: (color online) Spectral function vs. \( \omega \) near the Fermi level for various values of \( T_K/|J| \) showing how the Kondo peak is affected due to the coupling \( J \). The line corresponds to \( J = 0 \) and the same \( T_K \) as in the dashed (black) line. The inset shows a zoom-in around the Fermi level.

In Fig. 3 we show \( \rho_d(\omega) \) for various values of \( T_K/|J| \) near the Fermi level. Notice that even for \( T_K > |J| \) (e. g., \( T_K/|J| = 12.1 \)) line the Kondo peak split into a three-peak structure (including the very sharp peak at zero) due to the coupling \( J \). The characteristic energy of this regime is given by the width of the complete three peak structure. When compared to the solid curve for \( J = 0 \) we see that the change in the density of states is restricted to the Kondo peak (\( \sim T_K \)) and there is a clear collapse of the two curves for \( |\omega| \gtrsim T_K \). As \( T_K/|J| \) decreases we see a dramatic distortion in the Kondo peak: the three-peak structure tends to disappear and essentially the density of states is dominated by the central sharp peak at the Fermi level. It is interesting to observe that despite the strong modification of the spectral function, the height of the peak at the Fermi level remains \( 1/\pi \Gamma \), as predicted by the Friedel sum rule. As a
result, no effect would be expected for the conductance at $T = 0$. However, as we discuss below, the presence of this central peak has essential consequences on the interesting behavior of the conductance as the temperature is increased.

In Fig. 4 we show the conductance as function of temperature for various values of $J$. We notice two distinct regimes: i) for $J \ll T_K$ the conductance drops at $T \sim T_K$, where the effect of the temperature is to take the system out of the standard Kondo regime and ii) for $|J| \gg T_K$ the conductance drops for much lower temperature. For intermediate values of $J$, such as in $|J| = 7 \times 10^{-7} < T_K$, we observe that the behavior corresponding to the two regimes is contained in the same curve. The shape of the conductance differs completely whether $T < |J|$ or $T > |J|$. The appearance of these two regimes with temperature can be understood as following: by decreasing $T$, in the interval $|J| < T < T_K$, the system restores the behavior of the completely screened Kondo state at that temperature, as thermal excitations destroys the dot-magnetic atom spin-spin correlation (see Fig. 2c). However, reducing $T$, after a crossover region, when $T < J$, the dot-magnetic atom spin degrees of freedom are coupled and the system enters an underscreened Kondo state, characterized by a conductance that is significantly dependent upon temperature.

In summary, we have investigated the Kondo regime of a system composed of a single Mn$^{2+}$ ion in a QD coupled to metallic leads. Our numerical approach shows two distinct low temperature regimes, depending on how the ferromagnetic exchange interaction $J$ between the electrons in the QD and the magnetic moment of the Mn$^{2+}$ compares with $T_K$. In the weak regime ($T_K \gg |J|$), the QD is locked in a Kondo state singlet while the magnetic moment of the Mn$^{2+}$ decouples from the rest of the system, while in the strong coupling regime ($T_K \ll J$), the QD and the Mn$^{2+}$ forms a spin $I = 3$ impurity that couples to the conduction band. In this case, the impurity is underscreened by the Kondo correlation with the conduction band electrons. From the experimental point of view, Fig. 4 shows that the nature of the Kondo regime is reflected very significantly on the conductance permitting, through a transport measurement, to fully characterize the spin configuration of the system with very interesting quantum information implications. We expect that our results will stimulate experimental studies of these systems and contribute to the understanding of Kondo effect and transport in magnetic quantum dots.

We would like to thank the Brazilian agencies CNPq, CAPES, FAPERJ, FAPEMIG and FAPESP for financial support.

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