Suppression of the Kondo Effect in Quantum Dots by Even-Odd Asymmetry

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Abstract

We analyze here a model for single-electron charging in semiconductor quantum dots that includes the standard Anderson on-site repulsion (U) as well as the spin-exchange (\(J_d\)) that is inherently present among the electrons occupying the various quantum levels of the dot. We show explicitly that for ferromagnetic coupling (\(J_d > 0\)), an s-d exchange for an S=1 Kondo problem is recovered. In contrast, for the antiferromagnetic case, \(J_d < 0\), we find that the Kondo effect is present only if there are an odd number of electrons on the dot. In addition, we find that spin-exchange produces a second period in the conductance that is consistent with experimental measurements.

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When a gate voltage is applied to a nano-scale semiconductor inversion layer (or quantum dot), electrons will flow one at a time across this device provided that the applied voltage is an integral multiple of the capacitance charging energy of the quantum dot. Experiments illustrating the principle of charge quantization by virtue of the charging energy have been performed recently on numerous semiconductor [1] [2] [3] [4] [5] as well as superconducting [6] nano-structures. We focus here solely on the semiconductor devices. It is now well-accepted [5] that in semiconductor quantum dots, the dominant contribution to the capacitance charging energy, $E_c = \frac{e^2}{2C}$, arises from the on-site Coulomb repulsion. Here $C$ is the capacitance between the quantum dot, the tunnel junctions, and the electrical leads connected to the dot. Transport in quantum dots will be Coulomb limited if $k_B T < E_C$ and $k_B T > \Delta \epsilon$, where $\Delta \epsilon$ is the spacing between the single particle states of the dot.

Because of the central role played by on-site Coulomb repulsions in the transport properties of quantum dots, it is natural to model a quantum dot with a Hubbard-like model. So far as a quantum dot can be reduced to a single site [7] with a charging energy $U$, the Anderson model [8] for the interaction of a magnetic defect coupled to a non-interacting sea of conduction electrons is appropriate [7]:

$$H_A = \sum_{k,\sigma} \epsilon_k a_{k\sigma}^\dagger a_{k\sigma} + \sum_{\sigma} \epsilon_d a_{d\sigma}^\dagger a_{d\sigma} + \sum_{k,\sigma} V_{kd}(a_{k\sigma}^\dagger a_{d\sigma} + a_{d\sigma}^\dagger a_{k\sigma}) + U n_{d\uparrow} n_{d\downarrow}$$

$$= H_0 + \sum_{k,\sigma} V_{kd}(a_{k\sigma}^\dagger a_{d\sigma} + a_{d\sigma}^\dagger a_{k\sigma})$$

where $\epsilon_d$ is the defect energy of the magnetic impurity, $V_{kd}$ the overlap integral between a band state with momentum $k$ and the impurity, $a_k^\dagger$ creates an electron in the band states, $a_{d\sigma}^\dagger$ creates an electron with spin $\sigma$ on the impurity, and $n_{d\sigma} = a_{d\sigma}^\dagger a_{d\sigma}$ is the number operator for an electron of spin $\sigma$. As a consequence of the on-site repulsion, the single particle states on the impurity have energies, $\epsilon_d$ and $\epsilon_d + U$. At high temperatures, the density of states of this model has two Lorentzian peaks centered at these two energy levels. At low temperatures, however, the Anderson model displays a Kondo resonance [9] [10] at the Fermi level. Although the Kondo resonance is expected to occur for any value of the defect energy within the range $-U < \epsilon_d < 0$, it is most favourable at the defect energy corresponding
to the greatest stability of the local moment at the d-impurity, namely, \( \epsilon_d = -\frac{U}{2} \). At this energy \( H_A \) is particle-hole symmetric, and the Kondo resonance is pinned at \( \epsilon_F = 0 \). The single-particle states \( \epsilon_d \) and \( \epsilon_d + U \) lie symmetrically, then, around the Fermi energy. When the chemical potential of the source lead coincides with the energy of the Kondo resonance, a single electron should charge the dot. In the symmetric limit, this state of affairs should obtain at half-integer multiples of the charging energy. Thus far, no experimental hint of the Kondo resonance has been observed in quantum dots in zero bias voltage.

It is precisely the conditions under which the Kondo effect should be observed in quantum dots that we address here. While it may be premature to draw any conclusion from the lack of experimental confirmation of the Kondo effect, it is certainly appropriate to investigate the validity of the Anderson model to a quantum dot. It is in addressing this issue that we are 1) able to predict even-odd charging effects in semiconductor quantum dots as well as 2) a suppression of the Kondo effect when a quantum dot has an even number of electrons. The most obvious inadequacy of the Anderson model in the context of quantum dots is the truncation of the multiple electronic levels on a quantum dot to a single state. If multiple electronic levels are included on the dot, then other Coulomb interactions besides the on-site \( U \) become relevant. A key quantity that comes into play is the intrinsic Coulomb exchange energy, \( J_d \) between two levels. Consider for the moment a two-orbital model for a quantum dot. For two degenerate levels and \( U \approx U \), the direct Coulomb exchange integral, the energy of the two-body states predicted by this model are \( 2\epsilon_d + U - \frac{J_d}{4} \), \( 2\epsilon_d + U \), and \( 2\epsilon_d + U + \frac{3J_d}{4} \). The energy of the 3-electron state is \( 3(\epsilon_d + U) \). Consequently, the charging energy depends on \( J_d \). In fact, the general role of spin-exchange is to introduce a spin-dependent charging energy that is determined by the parity of the total number of electrons on the dot. It is worth pointing out that charging experiments on controlled-barrier atoms in strong magnetic fields display systematic oscillation in the peak heights, widths as well as in the separations that are consistent with a second period in the conductance as a function of the applied gate voltage. Such systematic deviations have been attributed to a splitting of the
energy between the up and down Landau levels, rather than to spin exchange. We propose here that such trends are also consistent with a spin-exchange model.

We explore then the simplest model of a quantum dot that includes the effects of spin-exchange. A natural way of including spin exchange is simply to introduce another level into the Anderson model. The only qualitative change this level is going to provide is the spin-interaction with the d-level. Consequently, we treat this level as a local spin, $S$. Because spin-exchange plays no role if $S = 0$, we will consider only the case in which the S-level is singly occupied, or equivalently, $S^z = \pm \frac{1}{2}$. Hence, large N expansion techniques are inappropriate to solving this problem. If we label the spin on the d-level of the impurity with $S_d$, we find that our Hamiltonian can be written as

$$H = H_A + H_J$$

$$H_J = -J_d S_d \cdot S$$

$$S_d^z = \frac{1}{2}(n_{d\uparrow} - n_{d\downarrow}), \quad S_d^+ = a_{d\uparrow}^\dagger a_{d\downarrow}, \quad S_d^- = a_{d\downarrow}^\dagger a_{d\uparrow}. \quad (5)$$

The first question we answer with this model is, does the Kondo effect still occur. Before rigorous calculations are performed, a heuristic answer can be put forth immediately. Without loss of generality, the defect energy can be taken to be $|\epsilon_d| \approx U >> |J_d|$, as $J_d$ is typically a fraction of U. The tunneling rate to the dot is determined by the matrix element $V_{kd}$. Because this quantity is an adjustable parameter determined by the width of the tunnel junction connecting the dot to the source lead, we can set $|J_d| > |V_{k}\cdot d|$. This inequality is crucial in the analysis of what follows. For example, in the absence of the $J_d$ coupling, the form of the antiferromagnetic interaction that gives rise to the Kondo effect scales as $|J_K| \approx \frac{|V_{k}\cdot d|^2}{U}$. However, in the limit that $|J_d| > |V_{k}\cdot d|$, $|J_d| >> |J_K|$. That is, the exchange coupling exceeds the Kondo coupling and could hence ultimately conspire to mask the Kondo effect. Consider the case in which the d-level and the S-level are singly occupied. In the ferromagnetic case, $J_d > 0$, the ground state of the dot is a triplet. A Kondo effect should result in this case that is determined by the total spin on the dot. However, in the antiferromagnetic case, $J_d < 0$, the ground state on the dot is a singlet. As a consequence,
there is no net spin to couple to the conduction electrons and the Kondo effect is suppressed. In the antiferromagnetic case, there must be an odd number of electrons on the dot for the Kondo effect to be observed. This is the essential physics of this model.

To prove the heuristic arguments given above, we diagonalize $H$ in the subspace of all singly-occupied states on the dot. We first note that because $[H_0, H_J] = 0$, we can work entirely with the eigenstates of the dot. Let us define a generalized eigenstate $|q\rangle = |N; S^z_{tot}, S^z_{tot}\rangle$, where $N$ refers to the number of electrons on the d-level of the dot. Recall, we have set $S^z = \pm \frac{1}{2}$. There are 8 eigenstates in the dot basis: $|0; \frac{1}{2}, S^z_{tot} = \pm \frac{1}{2}\rangle$ with total energy $E = 0$, the singlet $|1; 0, 0\rangle$ with energy $\epsilon_s = \epsilon_d + \frac{3J_d}{4}$, the triplets $|1, 1, S^z_{tot} = 0, \pm 1\rangle$, with energy $\epsilon_t = \epsilon_d - \frac{J_d}{4}$ and the doubly-occupied state, $|2; \frac{1}{2}, S^z_{tot} = \pm \frac{1}{2}\rangle$ with energy $2\epsilon_d + U$.

Each of the Fermion operators as well as the bi-linears such as $S_d \cdot S$ can be expressed in terms of these 8 basis states. Once this is done, matrix elements among these states can be determined straightforwardly. For an arbitrary wavefunction $|\psi\rangle$, we are interested in solving the Schrödinger equation

$$\sum_p \langle q|H|p\rangle \langle p|\psi\rangle = E \langle q|\psi\rangle \quad (6)$$

in the subspace of singly occupied states, $N = 1$. This is the relevant phase space for considering the Kondo effect. To reduce the full $8 \times 8$ to a $4 \times 4$, we rewrite the matrix elements involving the empty and doubly-occupied states in terms of the singly occupied states. The exact result is a $4 \times 4$ Hamiltonian matrix

$$\tilde{H} = \begin{pmatrix}
H_s & Q^{(-)}_{\uparrow\uparrow} - Q^{(-)}_{\downarrow\downarrow} & R_{\downarrow\downarrow} - R_{\uparrow\uparrow} \\
Q^{(-)}_{\uparrow\downarrow} - Q^{(-)}_{\downarrow\uparrow} & H_{t,0} & -R_{\uparrow\downarrow} - R_{\uparrow\uparrow} \\
R_{\uparrow\downarrow} & -R_{\downarrow\uparrow} & H_{t,1} \\
-R_{\uparrow\downarrow} & -R_{\downarrow\uparrow} & 0 & H_{t,-1}
\end{pmatrix} \begin{pmatrix}
\langle 1; 0, 0|\psi\rangle \\
\langle 1; 1, 0|\psi\rangle \\
\langle 1; 1, 1|\psi\rangle \\
\langle 1; 1, -1|\psi\rangle
\end{pmatrix} \quad (7)$$

where the singlet and triplet Hamiltonians are

$$H_s = H_c + \epsilon_S + \sum_\sigma Q^{(+)}_{\downarrow\sigma\sigma}, \quad H_{t,0} = H_c + \epsilon_t + \sum_\sigma Q^{(+)}_{\sigma\sigma\sigma}$$

$$H_{t,1} = H_c + \epsilon_t + 2Q^{(+)}_{\downarrow\uparrow}, \quad H_{t,-1} = H_c + \epsilon_t + 2Q^{(+)}_{\uparrow\downarrow}$$

(8)
and $Q$ and $R$ are the matrix elements

$$Q_{\sigma,\sigma'}^{(\pm)} = \frac{1}{2} \sum_{kk'} V_{k'} V_{k}^*[\pm (E + \epsilon_{k'} - H_c - 2\epsilon_d - U)^{-1} a_{k',\sigma}^+ a_{k\sigma} - (E - \epsilon_k - H_c)^{-1} a_{k',\sigma'}^+ a_{k\sigma'}]$$

(10)

$$R_{\sigma\sigma'} = \frac{1}{\sqrt{2}} \sum_{kk'} V_{k'} V_{k}^* [(E + \epsilon_{k'} - H_c - 2\epsilon_d - U)^{-1} + (E - \epsilon_k - H_c)^{-1}] a_{k',\sigma}^+ a_{k\sigma'}$$

(11)

The matrix elements $R$ and $Q$ contain all powers of the coupling to the conduction electrons. To lowest order, they scale roughly as $\frac{|V_k|^2}{U}$. The Hamiltonian matrix can be partitioned into $1 \times 1$ singlet and $3 \times 3$ triplet subspaces provided that the differences between the diagonal elements exceeds the off-diagonal matrix elements $R$ and $Q$. The diagonal elements differ by the spin-exchange $J_d$. Consequently, the partitioning into singlet and triplet subspaces is valid provided that $|J_d| > \frac{|V_k|^2}{U}$. The effective Hamiltonian in each subspace that is valid to second order in the coupling to the leads can be obtained by setting $E = \epsilon_s + H_c$ and $E = \epsilon_t + H_c$ in the denominators of $R$ and $Q$ and transcribing the basis state representation back to the original Fermion operators. In the ferromagnetic case ($J_d > 0$), the reduced Hamiltonian in the triplet subspace is

$$H_{\text{eff}}^{\text{triplet}} = H_c + \epsilon_t + \sum_{kk'} W_{kk'}^t (\psi_{k'}^\dagger \psi_k) - \sum_{kk'} J_{kk'}^t (\psi_{k'}^\dagger \sigma \psi_k) \cdot S_{\text{tot}}$$

(12)

where $H_c$ is the Hamiltonian for the free conduction electrons, $\sigma$ is the Pauli spin matrix, $\psi_k$ is the two-component spinor

$$\psi_k = \begin{pmatrix} a_{k\uparrow} \\ a_{k\downarrow} \end{pmatrix}$$

the antiferromagnetic coupling constant is

$$J_{kk'}^t = 2V_{k'd} V_{kd}^* \left( \frac{1}{\epsilon_k' - 2\epsilon_d - U + \epsilon_t} - \frac{1}{\epsilon_k - \epsilon_t} \right)$$

(13)

and

$$W_{kk'}^t = \frac{1}{2} V_{k'd} V_{kd}^* \left( \frac{1}{\epsilon_k' - 2\epsilon_d - U + \epsilon_t} + \frac{1}{\epsilon_k - \epsilon_t} \right).$$

(14)
That $J_{kk'}^t$ is negative can be seen immediately because the largest energy scale in the denominator is $U$ which enters with a $-$ sign. The spin interaction obtained in this limit is identical to the usual Kondo coupling except in this case the total spin on the dot enters. For a triplet state $S_{tot} = 1$. Consequently, ferromagnetic exchange gives rise to a $S=1$ Kondo problem. The $S=1$ Kondo problem is an example of an undercompensated spin problem in which the conduction electrons only partially screen the spins on the dot. The remaining unscreened spin couples ferromagnetically to the conduction band. The only qualitative difference between the $S=1$ Kondo problem and $S=\frac{1}{2}$ is the behavior of the magnetic susceptibility. As a result of the undercompensation, the susceptibility does not vanish at $T=0$ in the $S=1$ problem.

Consider now the more experimentally-relevant antiferromagnetic case [11]. We recover in this limit an effective Hamiltonian of the form

$$H_{eff}^{\text{singlet}} = H_c + \epsilon_s + \sum_{kk'} W_{kk'}^s (\psi_{k'}^\dagger \psi_k) + \frac{1}{8 J_d} \sum_{kk'112} J_{k1k_1'}^s J_{k2k_2'}^s (\psi_{k_1'}^\dagger \sigma \psi_{k_1}) \cdot (\psi_{k_2'}^\dagger \sigma \psi_{k_2})$$

(15)

where $W_{kk'}^s$ and $J_{kk'}^s$ are identical to their triplet counterparts with $\epsilon_s$ replaced by $\epsilon_t$. As is evident the spin coupling only involves the conduction electrons and is $O((V_{kd})^4)$. Further, the overall sign of this interaction is negative or ferromagnetic. Consequently, there is no antiferromagnetic exchange interaction that can produce a Kondo effect to fourth order in the coupling to the band electrons. The physical origin of the absence of the Kondo effect here is the stability of the singlet on an energy scale $J_d$. As a result, the Kondo coupling constant must be cut off at this energy scale. Consequently, it cannot diverge and give rise to a bound state at the dot. This result is consistent with the heuristic arguments of Ng and Lee [12] on the role of spin exchange in a quantum dot and a mean-field limit of the 2-impurity Anderson model in the presence of spin-exchange [13]. Jones, Kotliar and Millis [13] found that in the $N−>\infty$ limit of this model, a phase transition occurred which suppressed the Kondo effect if the bare exchange interaction exceeded a critical value. The critical condition is similar to the one used here, namely $|J_d| > \frac{|W_{kd}|^2}{U}$. There still remains one chance for the Kondo effect to be observed when $J_d < 0$. If the number of electrons on
the dot is odd, or equivalently we restrict ourselves to the N=0 subspace, the standard $S_{\frac{1}{2}}$ Kondo problem is recovered. If experiments are going to detect the Kondo effect, the total number of electrons on the dot must be carefully controlled.

In deriving the results in the Kondo regime, we have performed 2nd-order perturbation theory in the coupling to the leads. It is possible to construct a Schriefer-Wolff-type \cite{9} transformation that eliminates the coupling to the leads. The result

\[
\hat{S} = \frac{1}{2S + 1} \sum_{k,\sigma,\sigma'} V_k \left\{ \left( \frac{S + 1}{E_{t}^{(+)} - E_{s}^{(-)}} \right) \left( 1 - n_{d-\sigma} \right) + \left( \frac{S + 1}{E_{t}^{(+)} + E_{s}^{(+)}} \right) n_{d-\sigma} \right\} \delta_{\sigma\sigma'}
\]

\[
+ 2(S \cdot s_{\sigma\sigma'}) \left\{ \left( \frac{1}{E_{t}^{(+)} - E_{s}^{(-)}} \right) \left( 1 - n_{d-\sigma'} \right) + \left( \frac{1}{E_{s}^{(+)} - E_{t}^{(+)}} \right) n_{d-\sigma'} \right\} c_{k\sigma}^{\dagger} d_{\sigma'}^{\dagger}
\]

\[-h.c. \tag{16}\]

can be used to derive the effective Hamiltonians in the singlet and triplet subspaces. In the above, $E_{s,t}^{(-)} = \epsilon_k - \epsilon_{s,t}$ and $E_{s,t}^{(+)} = \epsilon_k - 2\epsilon_d - U + \epsilon_{s,t}$. In the singlet subspace, $S = 0$ and only those virtual transitions involving the triplet state survive. This transformation successfully eliminates the coupling to the leads in the limit $\frac{|V_k|^2}{U} << 1$ and hence is consistent with the perturbative treatment developed here.

We now calculate the conductance at finite temperature in the presence of spin-exchange. To facilitate this we need the average occupancy on the dot $\langle n_{d\sigma} \rangle$. This quantity is obtained by integrating the imaginary part of the d-electron Green function, $G_{d\sigma}(\omega) = \langle\langle a_{d\sigma}; a_{d\sigma}^{\dagger} \rangle\rangle$, weighted with the Fermi-Dirac distribution function. Standard equations of motion methods \cite{14} \cite{15} can be used to formulate an accurate expression for $G_{d\sigma}(\omega)$. Each level of iteration generates a new heirarchy of Green functions for which new equations of motion must be derived. To illustrate, the Heisenberg equations of motion for $G_{d\sigma}(\omega)$ generate two new Green functions, $\langle\langle n_{d-\sigma} a_{\sigma}; a_{d\sigma}^{\dagger} \rangle\rangle$ and $\langle\langle S \cdot S a_{d\sigma}; a_{d\sigma}^{\dagger} \rangle\rangle$. Equations of motion for this set of Green functions as well as for the new Green functions that appear at this level were derived and solved self-consistently for the impurity density of states by invoking the Hartree-Fock closure. The density of states obtained at the 3rd level of iteration is sufficient to describe the
Kondo effect. As we have already described the $T = 0$ phase, we focus on the experimentally-accessible high-temperature limit. The conductance

$$\Gamma = \frac{-2e^2}{\hbar} \frac{\Delta}{k_B T} \int f_{FD}(\omega)(1 - f_{FD}(\omega)) ImG_{ds}(\omega + 0)d\omega$$

(17)

was calculated using the standard Landauer formula [7] [16]. In Eq.(16), $\Delta = \frac{-1}{\pi} \sum_k |V_k|^2 \delta(\omega - \epsilon_k)$ To illustrate the role of spin-exchange, we report here the infinite U limit of $G_d(\omega)$ at the second level equations of motion. We find that the second level Green function

$$G_d(\omega) = \frac{3}{4} \left( \frac{1 - \langle n_{d-\sigma} \rangle}{\omega - \epsilon_s - \Sigma_0 (1 - \langle n_{d-\sigma} \rangle)} + \frac{1}{2} \langle S \cdot S_d \rangle \right) + \frac{1}{4} \left( \frac{1 - \langle n_{d-\sigma} \rangle}{\omega - \epsilon_t - \Sigma_0 (1 - \langle n_{d-\sigma} \rangle)} - \frac{1}{2} \langle S \cdot S_d \rangle \right)$$

(18)

contains a contribution for the singlet and triplet states with differing spectral weights. In Eq. (18), the self energy is $\Sigma_0 = \sum_k |V_k|^2 (\omega - \epsilon_k)^{-1} \approx -i\Delta$. This expression clearly illustrates that the singlet (first term) and the triplet (second term) spectral weights differ. We expect that the conductance into the singlet and triplet levels should reflect the asymmetry in the spectral weights. The conductance calculated from Eq. (17) (with the 3rd level Green function) is shown in Figure 1 as a function of the chemical potential. Illustrated clearly is the asymmetry in the conductance peaks centered at $\epsilon_s$ (the first peak) and at $\epsilon_t$ (2nd peak). In figure 1, $J_d = -.1U$. Hence, the singlet is the ground state. In the absence of $J_d$, the singlet and triplet peaks would coalesce into a single peak as in the standard Anderson model. The higher peaks in the conductance occur at energies $2\epsilon_d - \epsilon_t + U$ and $2\epsilon_d - \epsilon_s + U$, respectively. The upper peaks appear inverted because for $J_d < 0$, $\epsilon_s < \epsilon_t$. In the ferromagnetic regime, the triplet peak dominates and it is the neighbouring singlet states that lead to the asymmetry in the peak heights in the conductance as illustrated in Figure 2. We conclude then that spin exchange in zero magnetic field leads to peak height alternation in the conductance that is identical in form to the experimental [2] trends seen in the presence of a magnetic field. Ultimately, the ferromagnetic and antiferromagnetic cases can be distinguished by a low temperature study of the Kondo phase.
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Figure Captions

Figure 1: Conductance (measured in units of $\frac{e^2}{h}$) as a function of the chemical potential (measured in units of $U$) as computed from Eq. (17) using the third-level decoupling of the equations of motion for the Green function for $J_d = -0.1U$ and $\Delta = 0.001U$. Each set of two peaks corresponds to a singlet and triplet pair, the singlet being lower in energy in the antiferromagnetic case.

Figure 2: Same as Figure 1 but for the ferromagnetic coupling, $J_d = 0.1U$. Each set of two peaks corresponds to a singlet and triplet pair, the triplet being lower in energy in the ferromagnetic case.