Interplay between the mesoscopic Stoner and Kondo effects in quantum dots

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We consider electrons confined to a quantum dot interacting antiferromagnetically with a spin-$\frac{1}{2}$ Kondo impurity. The electrons also interact among themselves ferromagnetically with a dimensionless coupling $J$, where $J = 1$ denotes the bulk Stoner transition. We show that as $J$ approaches 1 there is a regime with enhanced Kondo correlations, followed by one where the Kondo effect is destroyed and impurity is spin polarized opposite to the dot electrons. The most striking signature of the first, Stoner-enhanced Kondo regime, is that a Zeeman field increases the Kondo scale, in contrast to the case for noninteracting dot electrons. Implications for experiments are discussed.

In the simplest version of the Kondo effect, a spin-$\frac{1}{2}$ magnetic impurity interacting antiferromagnetically (exchange coupling $J_K$) with delocalized conduction electrons forms a singlet with a cloud of conduction electrons. The nonperturbative Kondo energy scale $\Delta_K \simeq D \exp -1/J_K \rho_0$ (where $\rho_0$ is the density of states per spin per unit volume and $D$ is a high-energy cutoff) characterizes a host of properties of the system, including the reduction in ground state energy, the temperature dependence of the magnetic susceptibility, etc. Recent advances in nanofabrication have made new mesoscopic realizations of the Kondo problem possible. In one such realization, a small quantum dot with an odd number of electrons in a Coulomb Blockade valley plays the role of the impurity spin, while the conduction electrons live in the leads.

In this paper we consider a slightly modified setup in which the “conduction” electrons live in a large quantum dot with level spacing $\delta$, a variant of which has been realized. Such a model with noninteracting conduction electrons has been considered before, as has a model where the impurity spin interacts with a Luttinger liquid, but hitherto a treatment of realistic interactions between the “conduction” electrons in a quantum dot with the Kondo effect has been lacking.

Recently progress has been made in characterizing interactions in disordered quantum dots, where the Thouless energy $E_T = h/\tau_{\text{erg}}$ plays an important role ($\tau_{\text{erg}}$ is the time it takes for an electron to ergodize over the dot). In the limit where the Thouless number $g = E_T/\delta$ becomes large the following “Universal Hamiltonian” has been proposed for time-reversal invariant systems:

$$H_U = \sum_{\alpha,s} \epsilon_{\alpha,s} c_{\alpha,s}^\dagger c_{\alpha,s} + \frac{U_0}{2} N^2 - J S S^\dagger + \lambda T T^\dagger$$  \hspace{1cm} (1)

Here $\hat{N}$ is the total particle number, $\mathbf{S}$ is the conserved total spin, and $T = \sum c_{\beta,s}^\dagger c_{\beta,s}$. In addition to the charging energy, $H_U$ has an exchange energy $J$ and a superconducting coupling $\lambda$. For semiconductor quantum dots with $r_s \simeq 1$, $J$ is estimated to be $0.3\delta$, while $\lambda$ is negligible. Using the fermionic renormalization group, this Hamiltonian has been shown to be a stable fixed point at weak coupling (small $\tau_{\text{erg}}$), with other phases possible at strong coupling. We will use $H_U$ as prescribing the interactions among the electrons in the dot. As $J$ becomes stronger the system undergoes transitions to higher and higher total spin $S$, until at $J = \delta$ the dot becomes macroscopically polarized in a Stoner transition. Mesoscopic (sample-to-sample) fluctuations of the magnetization at a given $J$ due to variations of the energy levels have been theoretically characterized and observed.

The focus of this paper is the interplay between the Kondo and mesoscopic Stoner effects. Define $J = J/\delta$, $J_K = J_K/\delta$, and $E^0_S = S^0\delta$, where $S^0 = \text{total spin of the dot in the absence of the Kondo coupling}$, and denote $\Delta_{K0}$ as the Kondo scale for $J = 0$. Our central result is that in the limit when $g$, $S^0$, $\Delta_{K0}/\delta$ are large there are two regimes. In Regime I, $E^0_S \leq 2\Delta_{K0}$, the total spin $S$ is suppressed below $S^0$, while the Kondo scale $\Delta_K$ is enhanced over $\Delta_{K0}$. The most significant signature is that a Zeeman field increases $\Delta_K$ in this regime. In Regime II, the Kondo effect is destroyed by the mesoscopic Stoner effect, and the impurity is almost fully polarized opposite to the dot spin. There is a large jump in $S$ at the transition in our mean-field analysis, though a more accurate analysis may reveal a smooth crossover rather than a transition.

Our model Hamiltonian for a closed dot (not connected to leads) interacting with an impurity spin, ignoring the Coulomb term (for a constant number of particles), is

$$H = \sum_{k,s} \epsilon_k c_{ks}^\dagger c_{ks} + \sum_{kk', ss'} \tilde{\epsilon}_{k'} c_{ks}^\dagger \tilde{\tau}_{ss'} c_{k's'} + \sum_{kk', ss'} \tilde{\gamma}_{kk'} c_{ks}^\dagger \tilde{c}_{ss'} c_{k's'}$$  \hspace{1cm} (2)

Here $\mathbf{S}_f$ is the impurity spin and $\tilde{\tau}$ are the Pauli spin matrices. Since we are interested in the effects of competing interaction terms and not in mesoscopic fluctuations, we will make the level spacing uniform ($\epsilon_k = (k + \frac{1}{2}\delta)\delta$) with equal couplings to all levels. The high energy cutoff is $D = \epsilon_M$, and thus $k$ goes between $-M - 1$ and $M$. The local electronic spin at the impurity site $S_e = \sum_{kk', ss'} c_{ks}^\dagger \tilde{\tau}_{ss'} c_{k's'}/4M$, the impurity spin $\mathbf{S}_f$, and the
total electronic spin $S$ are not conserved, but the total spin of the system, $S_{\text{tot}} = S + S_r$ is conserved due to the spin-rotational invariance of $H$, as is $S_{\text{tot}}^z$. As $J$ increases, we expect transitions to successively higher values of $S_{\text{tot}}$, exactly as in the mesoscopic Stoner effect \[4, 10\]. If the dot has an even number of electrons, $S_{\text{tot}} = p + \frac{1}{2}$. We will work in the state $S_{\text{tot}}^z = S_{\text{tot}}$.

There are many ways to analyze the Kondo problem \[12\], with simplest way for our purposes being the large-$N$ approximation \[15, 16\]. In this approach, one writes the impurity spin in terms of an $f$-electron $S_r = \frac{1}{2} f^\dagger s f_s$, and extends the spin to a degeneracy quantum number $m$, with $-N/2 \leq m \leq N/2$. To represent the impurity spin properly a constraint on the number of $f$-electrons is imposed \[n_f = N/2 \] or $n_f = 1$ \[16\] which are identical for $N = 2$. Despite some subtle issues concerning the restoration of symmetry by quantum fluctuations \[17\], the leading large-$N$ results give a fairly good nonperturbative description of the physics, and are consistent with the results obtained by other methods \[14\]. We will take the leading large-$N$ approximation literally for $N = 2$, which should capture the physics of interest. One decouples the Kondo interaction by a Hubbard-Stratanovich transformation \[14, 19\]. We will also decouple the Stoner interaction $-JS^2$ by a Hubbard-Stratanovich transformation to get

$$Z = \int D\mathbf{h} Dc D\bar{c} Df D\bar{f} D\sigma D\lambda e^{-S}$$

$$S = \int dt \left( \frac{b^2}{4\pi} + 2\delta m_s + \sum_s \bar{f}_s \langle \partial_t + \epsilon_f + i\lambda \rangle f_s 
+ \sum_{kss'} \bar{c}_k (\langle \partial_t + \epsilon_k \rangle \delta_{ss'} - \frac{b}{2} \bar{s}_s \bar{s}_s') c_{kss'} 
- in_f \lambda + \sigma \delta \sum_{ks} \bar{c}_k f_s + \sigma \delta \sum_{ks} \bar{f}_s c_k \right)$$

where the field $\lambda$ imposes the constraint. At the mean-field level this describes a set of electrons in the quantum dot hybridizing with the impurity site and subject to a Zeeman field $h$. The fermionic part of the action can be integrated out to yield the effective action, the parameters of which must be chosen to lie at a saddle point, and to satisfy the constraint \[12, 19\]. The saddle-point values of $\epsilon_f$ and $\lambda$ are zero for our case. In our maximally polarized state, $h_s$ has an expectation value, while $h_{\perp}$ fluctuates. In order to obtain the values of $\lambda$ where the total spin changes, we will need to keep terms of order $p^2$ and terms of order $p$ in the effective action. The analysis is particularly simple in the limit $D \rightarrow \infty$, $\lambda_K \rightarrow 0$, with $\Delta_K$ held fixed. In this limit, defining $b = h_s/2$ and $\Delta_K = |\sigma|^2 \delta$, the $j^{th}$ root of the single-particle Green's function is

$$\omega_{j\gamma} = j\delta - b - \left( \frac{b}{2\pi} - \tan^{-1}(j\delta - b)/\Delta_K \right)$$

with $b \rightarrow -b$ for the $\downarrow$ spin. The errors in this are of order $\delta/\Delta_K$, and can be neglected in Regime I, $\delta \ll E_S \leq \Delta_K$. The ground state has $-M \leq j \leq p$ filled for the $\uparrow$ spin, while states $-M \leq j \leq -p - 1$ are filled for the $\downarrow$ spin.

In the limit of $T \rightarrow 0$ the fermionic contribution to the effective action is the ground state energy, and we obtain for the static mean-field effective action at $S_{\text{tot}} = p + \frac{1}{2}$

$$S_{\text{eff}}^{MF} = \frac{b^2}{4\pi} + \delta p (p + 1) - \frac{2}{\pi} (b - E_S) \tan^{-1} \frac{b - E_S}{\Delta_K}$$

$$-2bs_{\text{tot}} + \Delta_K \left( \log \left( \frac{(E_S - b)^2 + \Delta_K^2}{\Delta_K^2} \right) - 2 \right)$$

where we have used $\Delta_K = D \exp\left(-1/\lambda_K\right)$ to eliminate references to $\lambda_K$. It can be seen that the minimum of $b$ will be close to $JE_S$. We still do not have all the terms in the effective action to order $p$, for which we have to address fluctuations in $h_{x,y}$. The action for $h_{x,y}$ is expressed in terms of the susceptibilities of the spin operators $S_{x,y}$. Since there is an average $S_z$, $h_x$ and $h_y$ will have cross terms (a consequence of $[S_x, S_y] = iS_z$). Fluctuations of $h_{x,y}$ of higher order than quadratic are suppressed by powers of $1/b$, which correspond to powers of $1/S_{\text{tot}}$ and can therefore be ignored for large $S_{\text{tot}}$ near $\lambda \approx 1$. Integrating out the quadratic fluctuations leads to an effective action correct up to terms of order $p$, which is

$$S_{\text{eff}}^{MF} = \frac{b^2}{4\pi} + \delta p (p + 1) - 2bs_{\text{tot}} - \frac{2}{\pi} (b - E_S) \tan^{-1} \frac{b - E_S}{\Delta_K}$$

$$+ \Delta_K \left( \log \left( \frac{(E_S - b)^2 + \Delta_K^2}{\Delta_K^2} \right) - 2 \right) - b|1 - 2\delta b F|$$

where the sum $F(p, b, \Delta_K)$

$$F = \frac{\Delta_K^2}{2b^2} \sum_p \frac{\left( \tan^{-1} \frac{b + m\delta}{\Delta_K} + \tan^{-1} \frac{b - m\delta}{\Delta_K} \right)^2}{(\Delta_K^2 + (b + m\delta)^2)(\Delta_K^2 + (b - m\delta)^2)}$$

arises from “diagonal” excitations $j \uparrow \rightarrow j \downarrow$ which dominate the susceptibilities. For $E_S \ll \Delta_K$, noting that the saddle point value of $b$ is very close to $E_S$, we get

$$F \approx \frac{S_{\text{tot}}}{2b^2} (1 - \frac{4b^2}{3\Delta_K^2} + \cdots)$$

In this regime, after ignoring the $\lambda_K$ term which is negligible, the effective action takes the form

$$S_{\text{eff}}^{MF} = b^2 \left( \frac{1}{4\pi} + \frac{4\lambda_K}{\Delta_K^2} + \delta p (p + 1) - 2bs_{\text{tot}} - JS_{\text{tot}} \right)$$

$$+ \Delta_K \left( \log \left( \frac{(E_S - b)^2 + \Delta_K^2}{\Delta_K^2} \right) - 2 \right)$$

The additional $b^2$ term in Eq. (9) suppresses $E_S$. Also, $S_{\text{tot}}^z = 0$ favors a larger $\Delta_K$. This term arises from a smaller gain in spin fluctuation energy at larger $E_S/\Delta_K$ (Eq. (10)). For $E_S \ll \Delta_K$, since the impurity spin is locked into a singlet, the entire spin $S_{\text{tot}}$ is carried by the dot electrons, with the corresponding energy gain $-JS_{\text{tot}}(S_{\text{tot}} + 1)$. With increasing $E_S/\Delta_K$ the spin is distributed between the dot electrons and the impurity spin, leading to a smaller gain in $-JS^2$. Since transitions
between states of different $S_{tot}$ are driven by the delicate balance between the increase in kinetic energy and gain in spin exchange energy, this physics is central to the interplay between the Kondo and mesoscopic Stoner effects.

In Fig. 1 we show the result of a numerical calculation of the minimum of Eq. (6) for $\Delta K_0 = 100\delta$ as a function of $J$. The enhancement of $\Delta_K$ over $\Delta K_0$, and the suppression of $E_S$ below $E_S^0$ are evident throughout.

![Fig. 1](image1.png)

**FIG. 1:** The variation of $p$, $\Delta_K$, and the ground state energy with $J$ for $\Delta K_0 = 100\delta$. The solid line represents the ground state energy of Regime I, while the dashed line represents that of Regime II. Their crossing, demarcated by the solid vertical line, is the transition. Note that $\Delta_K$ continuously increases in Regime I, and that there is a large jump in $p$ at the transition.

The clearest signature of this state lies in its response to a Zeeman field $E_Z$, which adds the term $-E_Z S_{tot}$ to Eq. (6). This term favors larger $S_{tot}$, which as we have seen in the previous paragraph, favors larger $\Delta_K$. This effect is displayed in Fig. 2 for $\Delta K_0 = 100\delta$, and $J = 0.99$. This is to be contrasted with the usual paramagnetic Kondo state, in which a Zeeman coupling suppresses $\Delta_K$. This signature can be seen experimentally as enhancement of the Kondo resonance in the conductance provided the large dot is weakly coupled to leads. It may already have been seen about which more below.

![Fig. 2](image2.png)

**FIG. 2:** The variation of $p$, $\Delta_K$, and the ground state energy with $E_Z$ for $\Delta K_0 = 100\delta$ and $J = 0.99$. Their values at $E_Z = 0$ have been subtracted out, and are $p(0) = 14$, $\Delta_K(0) = 109.5$, and $E_{tot}(0) = -81.67$. Note that $E_Z$ is in units of $\delta$, and even for $E_Z = 0.5\delta$ there is a 10% enhancement of $\Delta_K$.

Let us now turn to the competing state. For $S_{tot} = p + \frac{1}{2}$, the allowed values of $S$ are $p$ and $p + 1$. Taking into account only the ground state configurations of the dot electrons, one finds the fully polarized state to be

$$|\Omega\rangle = \frac{1}{\sqrt{2(p+1)}} [(p + 1)(p + 2) - \delta \frac{J_K}{2}(p + 2)]$$

(10)

which has the impurity spin almost fully polarized opposite to the dot spin. Consequently, there will be no Kondo coherence in this state. The coupling to the state with $S = p$ is negligible in the large-$p$ limit. The energy of this state is

$$E_{II} = \delta(p + 1)^2 - J\delta(p + 1)(p + 2) - \delta \frac{J_K}{2}(p + 2)$$

(11)

There are perturbative corrections to this state and its energy coming from particle-hole excitations (which can be taken into account as a static impurity problem since the impurity spin is effectively frozen), but the scale of these corrections can be shown to be $J_K E_S$. In the limit $D \to \infty$, $J_K \to 0$, keeping $\Delta K_0$ fixed, the last term of Eq. (11) and the perturbative corrections are negligible. Fig. 1 also shows the energy of this state (in the above limit) and its total spin. The vertical line denotes the first-order transition (which may be smoothed to a crossover in a more accurate calculation) at which there is a large jump in the spin. This transition is located at roughly $E_S^0 \approx 2\Delta K_0$ in our mean field model. The lowest collective excited state in Regime II flips the impurity spin, with an energy of order $J_K E_S$, and should appear as a resonance in the conductance.

Let us now tie up some loose ends. Our mean-field approximation gives an accurate picture of the electronic spectrum deep in Regime I, and there the physics described after Eq. (6) is robust. However, close to the mean-field transition, our approximation may be inaccurate, and the transition found in mean-field may be smoothed into a crossover. We have considered an even number of dot electrons. For large $S_{tot}$ there is no qualitative difference between even and odd numbers of dot electrons. While we have carried out the calculation with $E_S^0$, $\Delta K_0 \gg \delta$, no qualitative difference is expected with $E_S^0$, $\Delta_K$ are a few times $\delta$. There should still be a sharp crossover from a regime with Kondo coherence to one without as $J$ increases. Finally, we have assumed equal spacings and couplings to the Kondo spin, whereas in reality both of these are controlled by Random Matrix Theory. For large $\Delta K_0$ and $J$ close to 1, the Kondo part of the physics is much the same. The main change will be that there are large mesoscopic fluctuations of $S$ (Kurland et al in ref. [8]), which may result in large mesoscopic fluctuations of the transition point. For smaller $\Delta K_0$, $E_S^0$, a numerical calculation along the lines of refs. [9] [10] needs to be carried out.

Consider now the experimental signatures. Kondo correlations, which can be seen by their conductance
signatures\(^{6}\) when the dot is weakly coupled to leads, are present in Regime I, and absent in Regime II. The total spin of the state can be measured by tracking the movement of conductance peaks as a function of parallel magnetic field \(B_{∥}\)\(^{13}\), and a large change should be seen in the total spin at the transition/crossover. Experimentally, the clearest signature is the strong enhancement of \(\Delta_K\) with the Zeeman coupling \(E_Z\) in Regime I, as shown in Fig.4. Since the spin of the Regime II state is much larger, a large enough Zeeman coupling will eventually push the system over into Regime II. In a recent experiment on a system with a large dot and two smaller dots serving as the impurity spins\(^{4}\), the authors observe that the zero-bias Kondo peak grows stronger with \(J_{K}\) (a signature of Regime I) before disappearing at zero bias by splitting into two peaks at finite bias (possibly the ±\(J_{K}\) \(E_S\) resonances of Regime II). In that experiment\(^{4}\), both the dots were strongly coupled to leads, so these observations are suggestive (but not conclusive) evidence for our physical picture.

In summary, we have analyzed a model in which electrons on a large quantum dot interact with themselves with a Universal Hamiltonian ferromagnetic exchange, while also interacting antiferromagnetically with an impurity spin. We find two regimes: In Regime I (the Stoner-enhanced Kondo regime) there is a robust Kondo scale which is enhanced as either \(J\) or the Zeeman coupling increases. For large enough \(J\) or \(E_Z\) the system will make a transition/crossover into Regime II, in which the Kondo coherence is destroyed in favor of polarizing the impurity spin opposite to the total spin of the dot electrons. There is a large change in the total spin at the transition/crossover. These are both mesoscopic regimes, in which the magnetization per particle can be made as small as one wishes.

There are a number of directions in which this work can be extended, the most natural and experimentally relevant being the investigation of mesoscopic fluctuations\(^{10}\). Another theoretically interesting system is the two-impurity Kondo problem\(^{21}\), which for noninteracting conduction electrons has an unstable non-fermi-liquid critical point. Since such a triple-dot system has already been realized experimentally\(^{14}\), the study of the effects of the Universal Hamiltonian exchange on two-impurity Kondo physics would be very interesting and timely. Finally, the case when the dots are strongly coupled to the leads demands closer scrutiny.

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