Interacting Quantum Dot Coupled to a Kondo Spin: A Universal Hamiltonian Study

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We study a Kondo spin coupled to a mesoscopic interacting quantum dot that is described by the “universal Hamiltonian.” The problem is solved numerically by diagonalizing the system Hamiltonian in a good-spin basis and analytically in the weak and strong Kondo coupling limits. The ferromagnetic exchange interaction within the dot leads to a stepwise increase of the ground-state spin (Stoner staircase), which is modified nontrivially by the Kondo interaction. We find that the spin-transition steps move to lower values of the exchange coupling for weak Kondo interaction, but shift back up for sufficiently strong Kondo coupling. The interplay between Kondo and ferromagnetic exchange correlations can be probed with experimentally tunable parameters.

A singly-occupied localized electron level (an “impurity” spin) interacting with a delocalized electron gas is a paradigmatic system in quantum many-body physics. It gives rise to the nonperturbative Kondo effect in which the localized electron’s magnetic moment is fully screened by the delocalized electrons below the Kondo temperature $T_K$. The Kondo effect was well studied in the context of an impurity moment embedded in a bulk metal [1]. Recently, Kondo physics has been the subject of much renewed interest [2], following its observation in quantum dots [3] in which key parameters may be tuned experimentally.

These experimental advances have been accompanied by progress in the theoretical treatment of the mesoscopic Kondo problem [4–9]. In mesoscopic systems, either the electrons in the leads or the electrons in a large dot play the role of the “electron gas” and a small spin-1/2 dot represents the Kondo spin. Here we focus on the latter case [see Fig. 1(a)], where the discrete spacing $\delta$ and the mesoscopic fluctuations of the single-particle levels in the large dot may alter the Kondo effect when $T_K \sim \delta$ [5–8].

A formidable challenge for both mesoscopic and bulk Kondo theory is to take into account electron-electron interactions in the electron gas. In the mesoscopic case this is simplified when the electron gas is confined to a large quantum dot in which the electron dynamics is chaotic [10,11]. When the dot’s Thouless energy $E_T$ is large compared with $\delta$, the effects of the electron-electron interaction are captured by the so-called universal Hamiltonian (UH) [12], valid in an interval $E_T$ around the dot’s Fermi energy $E_F$. For a fixed number of electrons, the dominant interaction in the UH is a ferromagnetic exchange interaction. Detailed comparison between theory and experiment for the statistics of Coulomb blockade peak heights and spacings shows that including the UH ferromagnetic exchange term is both necessary and sufficient to obtain quantitative agreement [13]. Thus, one can use this UH to obtain an experimentally relevant description of a large interacting dot (henceforth called the “dot”) that is Kondo coupled to a small dot with odd electron occupancy (henceforth called the “Kondo spin” $S_K$). Such a model was first discussed in the framework of a mean-field approximation [7], where Kondo correlations in a dot close to its ferromagnetic Stoner instability $J_s \sim \delta$ were investigated. A regime just below the instability was identified where the Kondo coupling substantially reduces the dot’s polarization. In contrast, studies in the bulk [14] found that a Kondo impurity enhances the polarization of a surrounding gas of electrons at similar high values of $J_s \leq \delta$.

The simplicity of the UH allows one to look for signatures of the Kondo interaction in the magnetic properties of the dot without necessarily assuming that the dot is very close to its bulk Stoner transition (in a quantum dot, $J_s$ is typically a fraction of $\delta$ [12,13]). Standard numerical methods for the Kondo problem such as quantum Monte Carlo [8] and numerical renormalization group...
techniques [9] are, however, not easily applied to this problem because the ferromagnetic exchange coupling introduces a sign problem in the former and nonlocal correlations in the latter. Here we apply a customized diagonalization method that takes advantage of the global spin-rotation invariance and uses the good-spin eigenstates of the UH as a basis [13,15].

The Hamiltonian of our system, schematically illustrated in Fig. 1, is given by [7,12]

$$H = \sum_{\mu, \sigma = \pm} e_\mu c_\mu^\dagger c_\mu - J_1 S_d^2 + J_k S_K \cdot s_d(0). \quad (1)$$

The first two terms in (1) constitute the UH of the dot [12] (ignoring a constant charging energy and a Cooper channel term), described by $N$ spin-degenerate single-particle levels $e_\mu$ and spin $S_d = \sum_{\sigma}\epsilon_\sigma^\dagger c_\sigma c_\sigma$ ($\sigma$ are Pauli matrices). The coupling of the dot to the Kondo spin $S_K$ (with $S_K = 1/2$) is mediated by its spin density $s_d(0) = \frac{1}{2} \sum_{\sigma_\alpha \sigma_\beta} \phi_\alpha^\dagger \phi_\beta c_\alpha^\dagger c_\beta$ (for simplicity, the tunneling position $r = 0$) with orbital wave function of level $\mu$. The parameters $J_1$ and $J_k$ are the exchange and Kondo coupling constants, respectively. In this Letter we ignore mesoscopic fluctuations, taking equally spaced single-particle levels covering a band of width $2D = (N - 1)\delta$ and $\phi_\mu(0) = 1/\sqrt{N}$. We also assume half-filling of the band so that the number of dot electrons is $N$. The average local density of states of the dot is $\rho = 1/(N\delta)$ [6].

The spin-rotation invariance of the Hamiltonian (1) implies the conservation of the total spin $S_{tot} = S_d + S_K$, so that $S_{tot}$ and $S_{tot}^0 = M_{tot}$ are good quantum numbers. To take advantage of this symmetry, we construct a good total spin basis by coupling the eigenstates of the UH with those of the Kondo spin. The UH eigenstates with dot spin $S_d$ are characterized by $|\gamma S_d M_d\rangle$ ($\gamma$ denotes orbital occupations $n_\mu$ and other quantum numbers distinguishing between states of the same dot spin $S_d$). Thus a basis of the coupled system with good total spin is $|\gamma S_d S_K M_{tot}\rangle$ (for simplicity the quantum number $S_K = 1/2$ is omitted). In this basis the UH is diagonal with energies $\sum_\mu e_\mu n_\mu - J_1 S_d^2 + 1$. The Kondo term $H_K = J_k S_K \cdot s_d(0)$ is a scalar product of vector operators in the uncoupled spaces. Thus, its matrix elements in the coupled basis conserve $S_{tot}, M_{tot}$ and are given by

$$\langle \gamma' S_d' S_{tot}' M_{tot}' | H_K | \gamma S_d S_{tot} M_{tot} \rangle \gamma' S_d' S_{tot}' M_{tot}'$$

$$= J_k (-1)^{S_d + 1/2 + S_{tot}} \left[ \begin{array}{cc} S_d & 1 \\ \sqrt{2} & 1/2 \end{array} \right]$$

$$\times \langle \gamma' S_d' | s_d(0) | \gamma S_d \rangle, \quad (2)$$

in terms of a Wigner-$6j$ symbol and the reduced matrix element of the spin density $s_d(0)$ (known in closed form [15]). In this formulation, the full Hamiltonian $H$ has a block diagonal structure in $S_{tot}, M_{tot}$.

The problem is further simplified by transforming to the basis of sites $i (0 \leq i \leq N - 1)$, in which the one-body part of the dot’s Hamiltonian is tridiagonal and $H_K = J_k S_K \cdot s_i$ with $s$ the spin at site $i$ [11] [see Fig. 1(b)]. The exchange interaction is invariant under such transformation and has the same form as in Eq. (1) with $S_d = \sum_i S_i$. We can thus recast our formalism in this site basis, where only neighboring sites are coupled and the Kondo spin interacts solely with site 0. Because of these features the many-body Hamiltonian matrix in the site basis is more sparse than in the orbital basis, allowing for an efficient diagonalization in each subspace of good $S_{tot}$ using a Lanczos-Arnoldi algorithm. In this approach we can conveniently diagonalize (1) for dots with up to $N \sim 12$ levels, where the total Hilbert space contains $\sim 5.4 \times 10^6$ basis states.

We calculated the lowest many-body energy eigenvalue for each value of $S_{tot}$ and thereby determined the ground-state value of the total spin for different values of $J_1$ and $J_k$. This quantity has been studied theoretically [7,8] and can be probed experimentally [16]. As $J_1$ increases, the ground-state spin $S_{tot}$ is expected to undergo successive transitions to higher values (known as the Stoner staircase) until the dot becomes fully polarized at $J_1 \sim \delta$. For $J_1 \to 0$, the spin transitions occur at $J_{tot}^m = (m + 1)/(m + 2)$, with $m = 1, 3, 5, \ldots$ for an odd (even) number of dot electrons $N$. These transition steps in the Stoner staircase are shifted by the Kondo interaction. In Figs. 2(a) and 2(b) we show the transition curves (solid lines) separating regions of fixed ground-state spin $S_{tot}$ in the two-dimensional parameter space of $J_1, J_k$. We observe that these curves are monotonically decreasing for $J_1 \rho \leq 1$ and monotonically increasing for $J_1 \rho \geq 1$. In the strong-coupling limit, they converge to values of $J_1$ that are either lower (for smaller $S_{tot}$) or higher (for larger $S_{tot}$) than their corresponding weak-coupling values $J_1^m$.

To gain insight into the behavior of the transition curves, we evaluate them for weak Kondo coupling in first-order (degenerate) perturbation theory. We find

$$J_1^m = [\left( m + 1 \right) \delta - \alpha_m J_k]/[m + 2]. \quad (3)$$

where $\alpha_m$ are positive constants of order one. These perturbative results (dotted lines in Figs. 2(a) and 2(b)) agree well with the numerical calculations for $J_1 \rho \ll 0.1$.

The negative slope of the transition curves at weak coupling can be understood by considering that in this weak-coupling regime the Kondo spin $S_K$ plays the role of an effective magnetic field, polarizing the dot in the direction opposite to its own spin. This will favor larger values of $S_d$ and thus also larger values of $S_{tot}$, hence the negative slope. As we approach the Stoner instability $J_1 \to \delta$ (i.e., as $m$ increases), the gain in exchange correlations $J_k s_d(S_d + 1)$ dominates over the gain in Kondo correlations, hence the flattening of the slope in this limit.

A perturbative analysis can also be carried out for the limit of strong Kondo coupling, which, at zero temperature, is characterized by $T_K \gg \delta$. For dots with sufficiently large $N$, this limit can be reached already for $J_k \rho \ll 1$, but we have not explored it here.
where the Kondo temperature is of the order $T_K \sim D e^{-1/J_{K}}$ (at $J_s = 0$) [1]. However, for the present case of $N \leq 12$, the limit $T_K \gg \delta$ requires $J_{K}\rho \gtrsim 1$ or, equivalently, $J_{K} \approx 2D$. The latter condition represents the bare strong-coupling limit for which a perturbative solution is available (without renormalizing the band width) [17]. In this limit the Kondo spin $S_K$ and the spin $s_0$ at site 0 form a strongly bound singlet $S_{K,0} = 0$ ($S_{K,0} = S_K + s_0$) that is effectively decoupled from the rest of the spin-chain with sites $i \geq 1$. The tridgonal one-body Hamiltonian of sites $1 \leq i \leq N-1$ can be rediagonalized to give $\sum_{\mu} \tilde{e}_{\mu}(r)$ and single-particle energies $\tilde{e}_\mu$. This dot has one less level and one less electron than the original dot. While the original dot levels are equally spaced, the level spacings in the reduced dot are given by $\tilde{e}_{\mu+1} - \tilde{e}_\mu \approx \delta + \beta_\mu/N > \delta$, $\beta_\mu > 0$ are of order 1 and increase monotonically from the new band center towards the band edges).

To explore how the strong-coupling limit is modified in the presence of exchange interaction in the dot, we rewrite the latter as $-J_s S^2_A = -J_s \mathbf{S}_d \cdot \mathbf{S}_d - 2J_s s_0 \cdot \mathbf{S}_d/2$, where $\mathbf{S}_d = \sum_{i=0}^{N-1} \mathbf{s}_i$ is the spin of the reduced dot. The cross term $-2J_s S_0 \cdot \mathbf{S}_d$ has vanishing matrix elements in the singlet subspace $S_{K,0} = 0$ but induces virtual transitions to the triplet subspace $S_{K,0} = 1$ that renormalize the exchange coupling constant $J_s \to J_s(1 + J_s/\mu_B + \cdots)$ (details will be presented elsewhere). To lowest order in $1/J_s$, our system is thus described by an effective Hamiltonian $\sum_{\mu} \tilde{e}_\mu \tilde{e}_\mu + J_s \tilde{S}_d^2$ that has the form of a UH for the reduced dot with single-particle energies $\tilde{e}_\mu$ and exchange constant $J_s$. The spin-transition curves of this reduced dot [dotted lines in Figs. 2(a) and 2(b)] are found to be in good agreement with the exact numerical curves when $J_s \rho \gg 1$. These exact numerical results further demonstrate that the spin-transition curves make a smooth crossover between the weak and strong Kondo coupling limits.

The spin-transition curves in the crossover from weak to strong coupling (see Fig. 2) are determined by two counteracting effects. (i) The effective removal of an electron from the dot shifts down the Stoner staircase according to $J_s \to J_s^{m}$. Since the reduced dot has one less electron, the shifted Stoner staircase is associated with the opposite number parity of electrons. (ii) The effective removal of a level from the dot stretches the step size in the staircase due to the larger level spacing in the reduced dot (i.e., $\delta_\mu > \delta$), and thus increases the spin-transition values of $J_s$. The downward shift in (i) is independent of $N$, but weakens for increasing $m$ (where the step values $J_s^m$ are more densely spaced). The upward shift in (ii) is a finite-size correction $-1/N$ that decreases with increasing $N$, but increases with increasing $m$ because of the nonuniform $\delta_\mu$. For smaller values of $J_s$, effect (i) dominates over (ii), resulting in an overall downward shift of the transition values in the strong-coupling limit (as compared to the weak-coupling values $J_s^0$). Close to the Stoner instability, however, finite-size effects (ii) dominate over (i), leading to transition values larger than $J_s^m$.

To investigate the interplay between effects (i) and (ii) more closely, we compare the spin-transition curves of our original systems ($N = 11, 12$) with systems of equal band width $2D$, but different values of $N$. Results shown in Figs. 2(c) and 2(d) demonstrate that finite-size effects (ii) decrease with increasing $N$, leading to a convergence of the strong-coupling transition curves towards $J_s^{m-1}$.

For dots with a large band width ($D \gg \delta$), truncation of the band to a size below $T_K$ leads to a strong-coupling Hamiltonian that includes additional interaction terms. It would be interesting to study the effects of these terms.

Signatures of the interplay between the intradot exchange and the Kondo coupling are revealed by applying an in-plane field $B$ [16], adding a Zeeman term $g\mu_B B S_0$ to the Hamiltonian $H$ in Eq. (1) ($g$ is the gyromagnetic factor and $\mu_B$ the Bohr magneton). This term favors a parallel configuration of dot spin ($\uparrow$) and Kondo spin ($\uparrow$), increasing $S_{tot}$ at $J_s = 0$. The addition of the Kondo interaction opposes such a parallel alignment ($\uparrow\uparrow$); correspondingly.
we find [see Fig. 3(a)] the spin-transition values of $B$ to increase monotonically with $J_k$.

A more subtle behavior appears for $J_s \neq 0$ [see Fig. 3(b)], where nonmonotonic spin-transition curves may arise. The behavior in weak Kondo coupling can again be understood in perturbation theory, for which the dot spin may arise. The behavior in weak Kondo coupling can again and giving a positive slope to $J_k$ further one reaches the point at which the antiparallel configuration is also favored in the subspace with higher $S_{\text{tot}}$. This decreases the slope of the spin-transition line, making it negative in some cases [see, e.g., the lowest curve in Fig. 3(b)]. For even larger $J_k$ the perturbative picture breaks down and the transition curves make a smooth crossover to the strong-coupling limit. Here the effective exchange constant $J_s$ decreases with increasing $J_k$, favoring lower $S_{\text{tot}}$ again and giving a positive slope to the transition curves. The nonmonotonicity of the lower spin-transition curves becomes more pronounced for larger values of $J_s$.

The ground-state spin of quantum dots (in the absence of Kondo coupling) has been measured in a number of experiments by varying an in-plane (Zeeman) field $B$ [16]. A Zeeman field can be used to map the spin-transition diagram in the following manner. A point where the lowest energy states with different spin are degenerate at fixed $J_k$ can be determined by observing a change in the slope of the Coulomb blockade peak positions vs. an in-plane field $B$ [18]. Tuning $J_k$ (by means of a pinch-off gate) at fixed $J_s$ will cause these slope changes to shift to higher or lower values of $B$ in a manner predictable from our calculations.

In summary, the interplay of Kondo and ferromagnetic (Stoner) correlations in large quantum dots leads to an interesting ground-state spin diagram. At weak Kondo coupling the Kondo spin acts as an external field to assist ferromagnetic polarization. At strong coupling the system is described again by a universal Hamiltonian, but with a renormalized exchange constant for a reduced dot with one less level and one less electron. Ferromagnetic polarization can be either enhanced or reduced in this limit, depending on how close the dot is to the bulk Stoner instability. The weak and strong-coupling limits are described well by perturbation theory and our exact numerical solutions find a smooth behavior in the nonperturbative crossover region.

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