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.

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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 δ 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 task 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 δ, 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" $\mathbf{S}_K$). 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 $J_s \lesssim \delta$.

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 δ [12,13]). Standard numerical methods for the Kondo problem such as quantum Monte Carlo [8] and numerical renormalization group

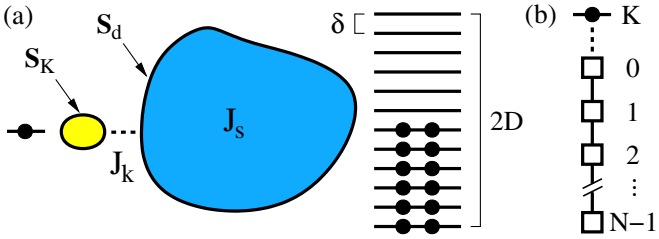

FIG. 1 (color online). (a) Schematic diagram of a small quantum dot with spin $\mathbf{S}_K$ (Kondo spin) that is coupled antiferromagnetically (coupling constant J_k) to a large quantum dot with spin $\mathbf{S}_d$. The large dot, described by the universal Hamiltonian, is characterized by a ferromagnetic exchange interaction (coupling constant J_s). We assume the large dot to have N equally spaced single-particle levels (with spacing δ) in a band of width $2D$ (half-filling). (b) The large dot is represented in the site basis (squares), in which $\mathbf{S}_K$ couples only to site 0.

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]

$$H = \sum_{\mu,\sigma=\pm} \varepsilon_\mu c^\dagger_{\mu\sigma} c_{\mu\sigma} - J_s \mathbf{S}_d^2 + J_k \mathbf{S}_K \cdot \mathbf{s}_d(0). \quad (1)$$

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 ε_μ and spin $\mathbf{S}_d = \frac{1}{2}\sum_{\mu\sigma\sigma'} c^\dagger_{\mu\sigma}\tau_{\sigma\sigma'}c_{\mu\sigma'}$ (τ are Pauli matrices). The coupling of the dot to the Kondo spin $\mathbf{S}_K$ (with $S_K = 1/2$) is mediated by its spin density $\mathbf{s}_d(0) = \frac{1}{2}\sum_{\mu\nu\sigma\sigma'} \phi_\mu(0) c^\dagger_{\mu\sigma}\tau_{\sigma\sigma'}\phi^*_\nu(0) c_{\nu\sigma'}$ at the tunneling position $\mathbf{r} = 0$ [$\phi_\mu(\mathbf{r})$ is the orbital wave function of level μ]. The parameters J_s 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 $\mathbf{S}_{\rm tot} = \mathbf{S}_d + \mathbf{S}_K$, so that $S_{\rm tot}$ and $S^z_{\rm tot} = M_{\rm tot}$ 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 $|\gamma S_d M_d\rangle$ (γ denotes orbital occupations n_μ 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 $|\gamma S_d S_{\rm tot} M_{\rm tot}\rangle$ (for simplicity the quantum number $S_K = 1/2$ is omitted). In this basis the UH is diagonal with energies $\sum_\mu \epsilon_\mu n_\mu - J_s S_d(S_d+1)$. The Kondo term $H_K = J_k \mathbf{S}_K \cdot \mathbf{s}_d(0)$ is a scalar product of vector operators in the uncoupled spaces. Thus, its matrix elements in the coupled basis conserve $S_{\rm tot}$, $M_{\rm tot}$ and are given by

$$\begin{aligned}&\langle \gamma' S'_d S_{\rm tot} M_{\rm tot}|H_K|\gamma S_d S_{\rm tot} M_{\rm tot}\rangle \\ &= J_k(-1)^{S_d+1/2+S_{\rm tot}}\sqrt{\frac{3}{2}}\begin{Bmatrix} S'_d & 1 & S_d \\ 1/2 & S_{\rm tot} & 1/2\end{Bmatrix} \\ &\quad\times(\gamma' S'_d \,\|\, \mathbf{s}_d(0) \,\|\, \gamma S_d),\end{aligned} \quad (2)$$

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

The problem is further simplified by transforming to the basis of sites i ($0 \le i \le N-1$), in which the one-body part of the dot's Hamiltonian is tridiagonal and $H_K = J_k \mathbf{S}_K \cdot \mathbf{s}_0$ with $\mathbf{s}_i$ the spin at site i [1] [see Fig. 1(b)]. The exchange interaction is invariant under such transformation and has the same form as in Eq. (1) with $\mathbf{S}_d = \sum_{i=0}^{N-1} \mathbf{s}_i$. 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 $S_{\rm tot}$ 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 $S_{\rm tot}$ and thereby determined the ground-state value of the total spin for different values of J_s and J_k. This quantity has been studied theoretically [7,8] and can be probed experimentally [16]. As J_s increases, the ground-state spin $S_{\rm tot}$ is expected to undergo successive transitions to higher values (known as the *Stoner staircase*) until the dot becomes fully polarized at $J_s \sim \delta$. For $J_k \to 0$, the spin transitions occur at $J_s^m = \delta(m+1)/(m+2)$, with $m = 1, 3, 5, \ldots$ ($m = 2, 4, 6, \ldots$) 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 $S_{\rm tot}$ in the two-dimensional parameter space of J_s, J_k. We observe that these curves are monotonically decreasing for $J_k\rho \lesssim 1$ and monotonically increasing for $J_k\rho \gtrsim 1$. In the strong-coupling limit, they converge to values of J_s that are either lower (for smaller $S_{\rm tot}$) or higher (for larger $S_{\rm tot}$) than their corresponding weak-coupling values J_s^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

$$J_s^m = [(m+1)\delta - \alpha_m J_k]/[m+2], \quad (3)$$

where α_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 $J_k\rho \lesssim 0.1$.

The negative slope of the transition curves at weak coupling can be understood by considering that in this weak-coupling regime the Kondo spin $\mathbf{S}_K$ 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 $S_{\rm tot}$, hence the negative slope. As we approach the Stoner instability $J_s \to \delta$ (i.e., as m increases), the gain in exchange correlations $J_s S_d(S_d+1)$ 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 $J_k\rho \ll 1$,

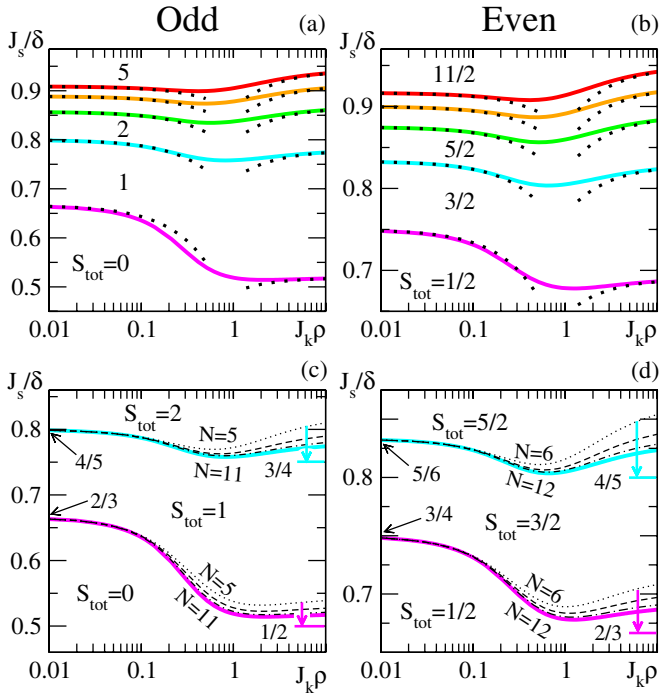

FIG. 2 (color online). Ground-state spin $S_{\rm tot}$ of the system in Fig. 1 at finite exchange J_s and Kondo coupling J_k for an odd (left column) and even (right column) number of electrons N. (a), (b) Transition curves for $N = 11$ (left) and $N = 12$ (right), separating regions of fixed $S_{\rm tot}$. Numerical results (solid lines) are compared with analytical estimates in the weak- and strong-coupling limits (dotted lines). (c), (d) Spin-transition curves for fixed bandwidth $2D$ but different N (top to bottom): $N = 5, 7, 9, 11$ (left) and $N = 6, 8, 10, 12$ (right). For increasing N (at $J_k\rho \gg 1$), the curves converge to the Stoner staircase of a dot with $N - 1$ electrons (vertical arrows).

where the Kondo temperature is of the order $T_K \sim De^{-1/J_k\rho}$ (at $J_s = 0$) [1]. However, for the present case of $N \lesssim 12$, the limit $T_K \gg \delta$ requires $J_k\rho \gtrsim 1$ or, equivalently, $J_k \gtrsim 2D$. 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 $\mathbf{S}_K$ and the spin $\mathbf{s}_0$ at site 0 form a strongly bound singlet $S_{K,0} = 0$ ($\mathbf{S}_{K,0} = \mathbf{S}_K + \mathbf{s}_0$) that is effectively decoupled from the rest of the spin-chain with sites $i \geq 1$. The tridiagonal one-body Hamiltonian of sites $1 \leq i \leq N - 1$ can be rediagonalized to give $\sum_{\mu\sigma}\bar{\varepsilon}_\mu \bar{c}^\dagger_{\mu\sigma}\bar{c}_{\mu\sigma}$, describing a "reduced" dot with new orbital wave functions $\bar{\phi}_\mu(\mathbf{r})$ and single-particle energies $\bar{\varepsilon}_\mu$. 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 $\bar{\varepsilon}_{\mu+1} - \bar{\varepsilon}_\mu \equiv \delta_\mu \approx \delta + \beta_\mu/N > \delta$, ($\beta_\mu > 0$ 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 $-J_s\mathbf{S}_d^2 = -J_s\bar{\mathbf{S}}_d^2 - 2J_s\mathbf{s}_0 \cdot \bar{\mathbf{S}}_d - 3J_s/4$ where $\bar{\mathbf{S}}_d = \sum_{i=1}^{N-1}\mathbf{s}_i$ is the spin of the reduced dot. The cross term $-2J_s\mathbf{s}_0 \cdot \bar{\mathbf{S}}_d$ 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 $J_s \to \bar{J}_s = J_s(1 + J_s/J_k + \cdots)$ (details will be presented elsewhere). To lowest order in $1/J_k$, our system is thus described by an effective Hamiltonian $\sum_{\mu\sigma}\bar{\varepsilon}_\mu \bar{c}^\dagger_{\mu\sigma}\bar{c}_{\mu\sigma} - \bar{J}_s\bar{\mathbf{S}}_d^2$ that has the form of a UH for the reduced dot with single-particle energies $\bar{\varepsilon}_\mu$ and exchange constant $\bar{J}_s$ ($\bar{J}_s \to J_s$ for $J_k \to \infty$). 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 $J_k\rho \gg 1$. 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 $J_s^m \to J_s^{m-1}$. 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 $\sim 1/N$ that decreases with increasing N, but increases with increasing m because of the nonuniform δ_μ. 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^m). 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 $g\mu_B B S^z_{\rm tot}$ to the Hamiltonian H in Eq. (1) (g is the gyromagnetic factor and μ_B the Bohr magneton). This term favors a parallel configuration of dot spin ($\uparrow$) and Kondo spin ($\Uparrow$), increasing $S_{\rm tot}$ at $J_k = 0$. The addition of the Kondo interaction opposes such a parallel alignment ($\uparrow\Uparrow$); correspondingly

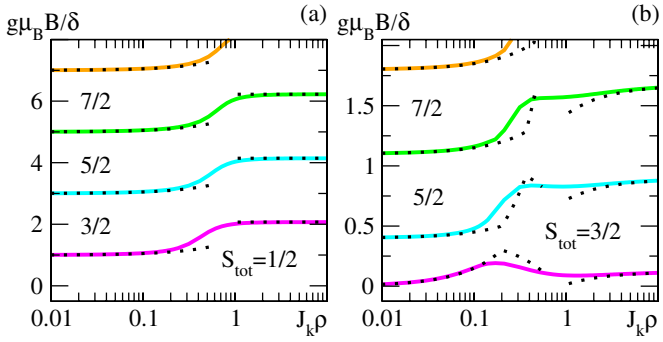

FIG. 3 (color online). Spin-transition curves for $N = 12$ at Zeeman field B and Kondo coupling J_k. The exchange coupling is (a) $J_s = 0$ and (b) $J_s/\delta = 0.65$. Solid (dotted) lines show the numerical (perturbative) solutions.

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 can still be regarded as a good quantum number. In the limit $J_k \rightarrow 0$ the ground state will always be the parallel configuration ($\uparrow\uparrow$) aligned with the external field, so that the spin-transition lines slowly increase with J_k (as for $J_s = 0$). At larger values of J_k the energy of the antiparallel ($\uparrow\downarrow$) configuration (with rearranged orbital occupancies) becomes lower in each spin subspace. This happens at lower J_k for the lower value of S_{tot}, leading to a marked increase in the slope of the transition line. Increasing J_k further one reaches the point at which the antiparallel configuration is also favored in the subspace with higher S_{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 $\bar{J}_s$ decreases with increasing J_k, favoring lower S_{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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