Strong-coupling limit of a Kondo spin coupled to a mesoscopic quantum dot: Effective Hamiltonian in the presence of exchange correlations

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We consider a Kondo spin that is coupled antiferromagnetically to a large chaotic quantum dot. Such a dot is described by the so-called universal Hamiltonian and its electrons are interacting via a ferromagnetic exchange interaction. We derive an effective Hamiltonian in the limit of strong Kondo coupling, where the screened Kondo spin effectively removes one electron from the dot. We find that the exchange-coupling constant in this reduced dot (with one less electron) is renormalized and that additional interaction terms appear beyond the conventional terms of the strong-coupling limit. The eigenenergies of this effective Hamiltonian are found to be in excellent agreement with exact numerical results of the original model in the limit of strong Kondo coupling.

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I. INTRODUCTION

The Kondo effect, which emerges when a localized impurity spin interacts antiferromagnetically with a delocalized electron gas, has generated considerable interest since it was first described in 1964.\textsuperscript{1,2} The observation that the Kondo resonance can be realized in the mesoscopic regime of quantum dots, in which many of the system parameters are experimentally tunable, has led to much renewed interest over the last decade.\textsuperscript{3–13} This experimental work has been accompanied by substantial theoretical progress on the mesoscopic aspects of the Kondo problem.\textsuperscript{14–27}

In the mesoscopic regime, the spin-1/2 Kondo impurity is typically represented by a small quantum dot with an odd number of electrons while the delocalized electron gas is realized by electrons in leads or in a large quantum dot. In this work we focus on the latter case, assuming two quantum dots of different size that are coupled antiferromagnetically, as in Fig. 1(a) (see Ref. 9 for an experimental realization of such a setup).

Certain features distinguish the mesoscopic Kondo regime from the bulk limit. While the conventional Kondo theory assumes a continuum band of energy levels in the electron gas, the single-particle energy levels in the large quantum dot are discrete. The discreteness and the dot-specific realization of these energy levels become important when the Kondo temperature $T_K$, the characteristic energy scale of the correlated Kondo resonance, is of the same order of magnitude as or smaller than the average level spacing $\bar{\delta}$.\textsuperscript{17,19,23,24,27} In the conventional bulk Kondo model the electron-electron interactions in the electron gas are often neglected. However, for the present double-dot setup, electron-electron interactions in the large dot can play an important role. In the following we assume the single-particle dynamics in the large quantum dot to be chaotic,\textsuperscript{28–31} in which case the dot is described by the so-called “universal Hamiltonian.”\textsuperscript{32} This Hamiltonian describes the low-energy physics in a Thouless energy interval around the dot’s Fermi energy. For a semiconductor quantum dot with a fixed number of electrons and in the limit of a large Thouless conductance, the electron-electron interaction is dominated by a ferromagnetic exchange interaction that is proportional to the square of the total dot’s spin. The universal Hamiltonian was shown to yield a quantitative agreement\textsuperscript{33} with experimental results measuring the statistics of the Coulomb-blockade peak heights and spacings.\textsuperscript{34}

The effect of ferromagnetic exchange correlations on the Kondo resonance was first addressed analytically in the bulk limit\textsuperscript{25} and, more recently, mean-field studies were carried out in the mesoscopic regime.\textsuperscript{23} In a recent work, we studied this problem numerically and provided analytical results for the weak and strong Kondo coupling limits.\textsuperscript{27} We found that for weak Kondo coupling, the Kondo spin acts like an external magnetic field, assisting the ferromagnetic polarization of

FIG. 1. (Color online) (a) Schematic illustration of the system under consideration: a small quantum dot with spin $S_K$ (Kondo spin) is coupled antiferromagnetically (coupling constant $J_k$) to a large quantum dot with spin $S_d$. The large dot is described by the universal Hamiltonian, characterized by a ferromagnetic exchange interaction (coupling constant $J_S$). The $N$ single-particle energy levels in the large dot are distributed within a band of width $2D$ (half filling). The average single-particle level spacing is fixed and given by $\bar{\delta}$. (b) The large dot is represented in the site basis (squares), in which $S_K$ couples only to site 0. In the strong-coupling limit, $J_k \rightarrow \infty$, it is useful to divide the Hamiltonian into three parts, $H_{K0}$, $H_{cp}$, and $H'_d$ [see Eq. (8)].
electrons in the large dot. In the case of strong Kondo coupling, the Kondo spin effectively removes one of the electrons of the large dot. We showed that this “reduced” dot with one less level and one less electron can again be described by a universal Hamiltonian but with a renormalized exchange constant.

A central issue that was not addressed in our previous work concerns the nature of residual interactions in the reduced dot beyond the renormalization of its exchange interaction term. From the work of Nozières we know that a noninteracting electron gas turns into a Fermi liquid when strongly coupled to a Kondo spin. The dominant effective interaction between the quasiparticles in this Fermi liquid is a repulsive interaction between spins of opposite orientation that are in close proximity to the Kondo spin. In the present case, the finite exchange interaction in the large dot leads to additional effective interaction terms in the strong-coupling limit. To identify these interaction terms, we follow a strategy that is similar to the one used by Nozières i.e., we perform an explicit perturbative expansion of the effective Hamiltonian of the reduced dot in the strong-coupling limit. In the presence of exchange interaction, this strong-coupling expansion is significantly more involved. However, the resulting effective quasiparticle interaction contains only a few new terms. The analytical expressions we derived for these effective exchange-like interactions are validated by a comparison with a full numerical diagonalization of the original two-coupled-dots model.

The outline of this paper is as follows: in Sec. II we present the model of a spin-1/2 quantum dot that is Kondo coupled to a large quantum dot (described by the universal Hamiltonian), and discuss the transformation from the single-particle orbital basis of the large dot to a chain site basis, commonly employed in the strong-coupling limit. In Sec. III we discuss the limit of strong Kondo coupling and use a projection method to derive an effective Hamiltonian for the reduced large quantum dot with one less electron. In Sec. IV we describe the evaluation of the eigenenergies of this effective Hamiltonian and in Sec. V we compare the results derived from this effective Hamiltonian with an exact numerical solution of the original model. In Sec. VI we conclude with a summary and discussion.

II. MODEL

We consider a chaotic quantum dot that is coupled antiferromagnetically to a Kondo spin as realized, e.g., by a small quantum dot with an odd number of electrons. A schematic illustration of such a system is shown in Fig. 1(a). In the following we will refer to the large quantum dot as the “dot” and to the small dot as the “Kondo spin.”

A. Hamiltonian

In the limit of a large Thouless conductance, a quantum dot whose single-particle dynamics is chaotic is described by the universal Hamiltonian

\[ \hat{H}_d = \sum_{n=0}^{N-1} \sum_{\sigma} e_n^\sigma \hat{a}_n^\dagger \hat{a}_{n,\sigma} - J \hat{S}_d^z. \]  

(1)

Here \( \hat{a}_{n,\sigma}^\dagger \) creates an electron with spin up/down (\( \sigma = \pm 1 \)) in level \( n \) in an orbital single-particle level with energy \( e_n^\sigma \). We assume \( N \) spin-degenerate single-particle levels spanning a bandwidth of \( 2D = (N-1) \times \delta \) (\( \delta \) is the mean level spacing). The second term on the right-hand side (rhs) of Eq. (1) represents a ferromagnetic exchange interaction \( (J > 0) \), where \( \hat{S}_d = \frac{1}{2} \sum_{n=0}^{N-1} \sigma \hat{a}_{n,\sigma}^\dagger \sigma n \sigma \hat{a}_{n,\sigma}^\dagger \) (\( \tau \) are Pauli matrices) is the total spin of the dot. In Eq. (1) we have ignored a constant charging energy term and a repulsive Cooper channel term.

The dot is coupled antiferromagnetically to a Kondo spin \( \hat{S}_K (S_K = 1/2) \) (Ref. 23)

\[ \hat{H} = \hat{H}_d + J \hat{S}_K \cdot \hat{s}_d(0), \]  

(2)

where \( J (J_K > 0) \) is the Kondo coupling constant and \( \hat{s}_d(0) \) is the spin density of the dot at the tunneling position \( r = 0 \). The dot spin density at position \( r \) is given by

\[ \hat{s}_d(r) = \frac{1}{2} \sum_{\sigma,\sigma'} \hat{\psi}_{\sigma'}^\dagger(r) \tau_{\sigma,\sigma'} \hat{\psi}_{\sigma}(r), \]  

(3)

where \( \hat{\psi}_{\sigma}(r) \) creates an electron with spin \( \sigma \) at position \( r \). In terms of the single-particle orbital wave functions \( \phi_n^\sigma(r) \), the field operator is given by \( \hat{\psi}_{\sigma}(r) = \sum_{n=0}^{N-1} \phi_{n}^\sigma(r) \hat{a}_{n,\sigma}^\dagger \) and the local density of states of the dot is given by \( \rho(r) = \sum_{n=0}^{N-1} |\phi_{n}^\sigma(0)|^2 \delta(r - r^\sigma_n) \), with an average value of \( \bar{\rho} = 1/(N\delta) \).

B. Chain site basis

The strong-coupling limit of the system in Fig. 1(a) is more clearly described when the Hamiltonian in Eq. (2) is rewritten in a different basis, known as the chain site basis. This chain site basis is obtained by a unitary transformation of the orbital basis

\[ \hat{c}_{n,\sigma}^\dagger = \sum_{n=0}^{N-1} U_{n,\sigma} \hat{a}_{n,\sigma}^\dagger, \]  

(4)

such that site \( \mu = 0 \) corresponds to the tunneling position \( r = 0 \), and the one-body site Hamiltonian of the dot is now tridiagonal, i.e., each site is coupled to its two nearest neighbors. Such a transformation is constructed by choosing \( \hat{c}_{n,\sigma}^\dagger = \hat{\psi}_{\sigma}(r = 0) \) and carrying out a Gram-Schmidt orthogonalization procedure. 2

The site single-particle energies \( e_n^\sigma \) are given by the diagonal elements of \( \hat{H}_0 = \sum_{n=0}^{N-1} e_n^\sigma \hat{c}_{n,\sigma}^\dagger \hat{c}_{n,\sigma}^\dagger \) when the latter is rewritten in the site basis. The off-diagonal matrix elements \( t_{\mu} = t_{\mu,\mu+1} \) and \( t_{\mu}^{\ast} = t_{\mu,\mu-1} \) describe the hopping amplitudes between neighboring sites. A spin \( \hat{s}_d(0) \) can be associated with each site, where the spin of site \( \mu = 0 \) is equal to the spin density at the tunneling position, i.e., \( \hat{s}_d(0) = \hat{s}_0 \). In the site basis, the Kondo spin couples only to a single site \( \mu = 0 \). The Hamiltonian in Eq. (2) is now given by

\[ \hat{H} = \hat{H}_0 - J \hat{S}_d^z + J \hat{S}_K \cdot \hat{s}_0, \]  

(5)

where the total spin of the dot is \( \hat{S}_d = \sum_{\mu=0}^{N-1} \hat{s}_\mu \). Here \( \hat{H}_0 \) is the one-body Hamiltonian of the dot in the site basis.

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we can ignore the coupling term quantities. For such operators, the summation over sites advantage of this symmetry, it is convenient to use a basis for given by expressions similar to Eqs. The site-basis formulation is particularly advantageous for the strong-coupling limit $J_s \to \infty$ when the site $\mu = 0$ effectively decouples from the rest of the chain. Accordingly, we decompose the Hamiltonian in Eq. (5) into three terms [see Fig. 1(b) for a schematic illustration]

$$\hat{H} = \hat{H}_K + \hat{H}_d' + \hat{H}_{cp},$$

where $\hat{H}_K$ describes the Hamiltonian of the Kondo spin plus site $\mu = 0, \hat{H}_d'$ is the Hamiltonian of a “reduced” dot with $N-1$ sites $\mu = 1, \ldots, N-1,$ and $\hat{H}_{cp}$ contains the remaining coupling terms. Writing $\hat{S}_d = \hat{S}_d' + \hat{s}_0,$ where $\hat{S}_d' = \sum_{\mu = 1}^{N-1} \hat{s}_\mu$ is the spin of the reduced dot, we have

$$\hat{H}_K = 4 \hat{s}_0 \cdot \hat{J}_d \hat{S}_d' + J \hat{S}_d \cdot \hat{s}_0,$$

$$\hat{H}_d' = \hat{H}_0' - J_s \hat{S}_d^2,$$

$$\hat{H}_{cp} = -2 J_s \hat{J}_d \cdot \hat{S}_d' + \sum_{\sigma} (t_{\sigma \sigma} \hat{c}_{\sigma 0, \sigma} \hat{c}_{\sigma 1, \sigma} + \mathrm{H.c.}).$$

$\hat{H}_d'$ in Eq. (10) is the “bare” Hamiltonian of the reduced dot given by expressions similar to Eqs. (6) and (7) but with the sums over $\mu$ starting at $\mu = 1.$ Here and in the following, operators in the reduced dot space of $N-1$ sites $\mu = 1, \ldots, N-1$ are denoted by primed quantities. For such operators, the summation over sites $\mu$ starts from $\mu = 1$ rather than $\mu = 0.$

C. Site basis with good spin quantum numbers

The Hamiltonian $\hat{H}$ in Eq. (8) is invariant under spin rotations and therefore conserves the total spin of the system (Kondo spin plus dot spin) $\hat{S}_{tot} = \hat{S}_K + \hat{S}_d = \hat{S}_K + \hat{S}_d'$ is the total spin of the system. To take advantage of this symmetry, it is convenient to use a basis for which both the total spin $\hat{S}_{tot}$ and the corresponding magnetic quantum number $M_{tot} = S_{tot}$ are good quantum numbers.

There are different ways to construct a basis with good total spin but one of them is particularly useful in the strong-coupling limit $J_s \gg J_d, J_s.$ To zeroth order in $J_d/J_s$ and $J_s/J_d,$ we can ignore the coupling term $\hat{H}_{cp},$ in which case the sub-system of Kondo spin plus site 0 decouples from the reduced dot. The Hamiltonian $\hat{H}_K$ is easily diagonalized by coupling the spins $\hat{S}_K$ and $\hat{s}_0$ to $\hat{S}_K = \hat{S}_K + \hat{s}_0,$ and using $\hat{S}_K \cdot \hat{s}_0 = (\hat{S}_K^2/2 - \hat{S}_d^2/2)/2.$ If site $\mu = 0$ is singly occupied, i.e., $n_0 = 1,$ this spin coupling will lead to either a singlet $S_{tot} = 0$ (lowest energy) or a triplet $S_{tot} = 1$ (highest energy). However, if site $\mu = 0$ is empty ($n_0 = 0$) or doubly occupied ($n_0 = 2,$ the spin at site 0 and the corresponding Kondo coupling term vanish.

This results in an unscreened Kondo spin in a doublet state ($S_{tot} = 1/2$), the energy of which is intermediate between the singlet and triplet states.

We now construct a basis of good total spin that also reflects the division into singlet/doublet/triplet manifolds. The eigenstates of $\hat{H}_K$ are characterized by $S_{tot}, M_{tot}$ with $M_{tot}$ being the magnetic quantum number of $\hat{S}_{tot}.$ The eigenstates of the reduced dot Hamiltonian $\hat{H}_d'$ with $N-n_0$ electrons are characterized by $|\gamma S_d'M_d'\rangle,$ where $S_d', M_d'$ are the spin and spin projection, respectively, of the reduced dot and $\gamma$ denotes all other quantum numbers distinguishing between states with the same $S_d'. We then couple the above eigenstates of $\hat{H}_K$ with the eigenstates of the reduced dot to form states with good total spin and spin projection quantum numbers $S_{tot}, M_{tot}. This basis of the coupled system is given by $|n_0, S_{tot}, \gamma S_d'M_d'\rangle.$ To keep the notation simple, we omitted the quantum numbers $S_d = 1/2$ and $n_0 = n_0(2-n_0)/2.$

Spin projection rules determine the allowed values of the reduced dot spin $S_d'$ for a given value of the total spin $S_{tot}$. We have $S_d' = S_{tot}$ for the singlet subspace, $S_d' = S_{tot} \pm 1/2$ for the doublet subspace, and $S_d' = S_{tot}, S_{tot} \pm 1$ for the triplet subspace.

III. STRONG-COUPLED HAMILTONIAN

The strong-coupling limit is defined by $J_s \gg \tilde{t}_0. Since \tilde{t}_0 \sim \hat{N} \approx \hat{N}, this limit corresponds to $J_s/\tilde{t}_0 \approx 1,$ where $\tilde{t}_0 = 1/(N\hat{N})$ is the average single-particle level density per site.

In the strong-coupling limit, the lowest eigenstates of $\hat{H}$ are dominated by the singlet manifold. The bare singlet Hamiltonian (in the limit when $\hat{H}_{cp}$ is ignored) is given by the reduced dot Hamiltonian $\hat{H}_d'$ with $N-1$ electrons (except for a constant shift). However, virtual transitions between the singlet and doublet/triplet manifolds add correction terms to this Hamiltonian. Our goal is to determine the resulting effective Hamiltonian of the reduced dot in the strong-coupling limit.

A. Projection technique

At finite Kondo interaction, the above three manifolds (singlet, doublet, and triplet) are coupled to each other. Specifically, the exchange term in $\hat{H}_{cp}$ couples the singlet and triplet manifolds while the hopping term between sites 0 and 1 couples each of the singlet and triplet manifolds to the doublet manifold. To account for these couplings we define projection operators $\hat{P}_s, \hat{P}_d, \hat{P}_t$ on the corresponding singlet/doublet/triplet subspaces ($\hat{P}_s + \hat{P}_d + \hat{P}_t = 1$) and decompose the wave function $\psi = \psi_s + \psi_d + \psi_t$ accordingly. The Schrödinger equation for the coupled system $\hat{H}\psi = E\psi$ can then be written as

$$\sum_{\beta = s, d, t} \hat{H}_{\alpha \beta} \psi_\beta = E \psi_\alpha,$$

where each of the two indices $\alpha, \beta$ assumes any of three values $\{s, d, t\}$ and $\hat{H}_{\alpha \beta} = \hat{P}_\alpha \hat{H} \hat{P}_\beta.$
In the strong-coupling limit, our system is described to zeroth order by the singlet Hamiltonian $\hat{H}_{ss}$, which contains the bare reduced dot Hamiltonian $\hat{H}_{r}^{(0)}$ (for $N-1$ electrons) and $\hat{H}_{K0}$ (which assumes a constant value), decoupled from each other. Higher-order corrections arise from the coupling terms in $\hat{H}_{cp}$, which lead to an effective “dressed” Hamiltonian of the reduced dot. This effective Hamiltonian $\hat{H}^{\text{eff}}$ is formulated by eliminating $\psi_i$ and $\varphi_i$ in Eq. (12) and by writing a single equation in the singlet manifold $\hat{H}^{\text{eff}} \psi_s = E \psi_s$, where $\hat{H}^{\text{eff}}$ is given by

$$\hat{H}^{\text{eff}} = \hat{H}_{ss} + \hat{H}_{st}(E-\hat{H}_{s})^{-1}\hat{H}_{st} + [\hat{H}_{sd} + \hat{H}_{dt}(E-\hat{H}_{d})^{-1}\hat{H}_{td}]^{-1} \times [E - [\hat{H}_{dd} + \hat{H}_{dt}(E-\hat{H}_{d})^{-1}\hat{H}_{td}]]^{-1} \times [\hat{H}_{ds} + \hat{H}_{dt}(E-\hat{H}_{d})^{-1}\hat{H}_{ts}]$$. (13)

The diagonal components $\hat{H}_{ss}$ in the above equation are determined by evaluating $\hat{H}_{K0}$ [Eq. (9)] in each of the three subspaces $\alpha \in \{s,d,t\}$. The coupling terms in $\hat{H}_{st}$ [Eq. (11)] do not contribute to these diagonal components with the exception of $\hat{S}_0 \cdot \hat{S}_s$ which contributes to $\hat{H}_{ss}$ only. We find

$$\hat{H}_{ss} = \hat{P}_s (\epsilon_0^s - \frac{3J_k}{4} \cdot \hat{S}_0) \hat{P}_s$$, (14)

$$\hat{H}_{dd} = \hat{P}_d (\epsilon_0^d \hat{S}_0 + \hat{H}^d) \hat{P}_d$$, (15)

$$\hat{H}_{tt} = \hat{P}_t (\epsilon_0^t + \frac{J_k}{4} \cdot \hat{S}_0 - \frac{3J_s}{4} \cdot \hat{S}_d) \hat{P}_t$$, (16)

Contributions to off-diagonal components $\hat{H}_{sd}$ with $\alpha \neq \beta$ originate in $\hat{H}_{cp}$. The hopping term in $\hat{H}_{cp}$ changes the spin $S_{K0}$ by $\pm 1/2$ and can only couple the doublet to each of the singlet and triplet manifolds while the exchange term $\hat{S}_0 \cdot \hat{S}_i$ in $\hat{H}_{cp}$ only couples the singlet and triplet manifolds.

**B. Expansion in the strong-coupling limit**

The above effective Hamiltonian $\hat{H}^{\text{eff}}$ and the construction of a good spin basis are exact, in that no approximations were made beyond the original Hamiltonian $\hat{H}$ in Eq. (2). However, the form (13) of $\hat{H}^{\text{eff}}$ is not very useful for practical calculations. In the strong-coupling limit, $J_k \gg J_0 \sim N \delta$, we can expand $\hat{H}^{\text{eff}}$ in the two small dimensionless parameters $J_0/J_k \sim 1/(J_k \delta)$ and $J_1/J_k$36 We will do so up to fourth order in these parameters, where the expansion terms are measured in units of $J_k$ (this energy unit is set by the energy of the singlet).

The starting point for this expansion is the unperturbed singlet Hamiltonian $\hat{H}_{ss}$, the eigenbasis of which is given by $|n_0=1, S_{K0}=0; \gamma, S'_d; S_{tot}=S'_d, M_{tot}\rangle$. The corresponding eigenvalues are

$$E^{(0)}_m = \frac{3}{4}(J_d + J_s) + \epsilon_0^s + E_{0,m} - J_S S_{tot}(S_{tot} + 1),$$ (17)

where $E_{0,m}$ are the eigenvalues of $\hat{H}_{ss}$. These unperturbed eigenvalues, $E^{(0)}_m$, are the limiting solutions to which the eigenvalues $E_m$ of the full Hamiltonian in Eq. (13) converge in the limit $J_k \to \infty$. The differences between $E^{(0)}_m$ and $E_m$ at large but finite values of $J_k$ are induced by the virtual transitions from the singlet to the doublet or triplet subspaces. These virtual excitations, in turn, give rise to effective interaction terms in the reduced dot, denoted by $\delta \hat{H}^{\text{eff}}$. The full effective Hamiltonian in the singlet manifold is then given by $\hat{H}^{\text{eff}} = \hat{H}_{ss} + \delta \hat{H}^{\text{eff}}$.

The effective interaction terms in $\delta \hat{H}^{\text{eff}}$ must be consistent with charge and spin conservation. In particular, $\delta \hat{H}^{\text{eff}}$ must be a scalar operator in spin space (i.e., invariant under rotations in spin space) and invariant under time reversal. This restricts the possible terms that appear in the effective Hamiltonian.

Scalar one-body terms, i.e., $\hat{n}_i$ and $(\sum_{\sigma} \hat{c}_{i,\sigma} \hat{c}_{i,\sigma} + H.c.)$ lead to a renormalization of the one-body part of the reduced dot Hamiltonian $\hat{H}_{r}^{(0)}$. Two-body scalars that can be constructed from the spin $\hat{n}_i$ at site 1 and the total spin of the reduced dot $S_i$ are $\hat{s}_i, \hat{s}_i, \hat{s}_j$, and $\hat{s}_j$. The first, $\hat{s}_i = \frac{1}{2}(2 - \hat{n}_i)$ is the Nozières term known from the conventional Kondo problem in the absence of exchange ($J_k=0$) but the other two terms are new. The scalar triple product $\hat{s}_i \cdot (\hat{s}_i \times \hat{s}_j)$ (the imaginary $i$ is necessary for time-reversal invariance) does not lead to an additional term since $\hat{s}_i \cdot (\hat{s}_i \times \hat{s}_j) \equiv -\hat{s}_j \cdot \hat{s}_j$ while a fourth-order invariant is given by $\hat{s}_i \cdot \hat{s}_j \hat{s}_k \hat{s}_l$. Other invariants such as $\hat{s}_i \cdot \sum_{\sigma,\sigma'} (\hat{c}_{i,\sigma} \hat{c}_{i,\sigma'} \hat{c}_{i,\sigma} \hat{c}_{i,\sigma'} + H.c.)$, $\hat{s}_i \cdot \sum_{\sigma,\sigma'} (\hat{c}_{i,\sigma} \tau_{\sigma,\sigma'} \hat{c}_{i,\sigma'} \tau_{\sigma,\sigma'} + H.c.)$, and $\hat{s}_i \cdot \sum_{\sigma,\sigma'} (\hat{c}_{i,\sigma} \hat{c}_{i,\sigma'} + \hat{c}_{i,\sigma} \hat{c}_{i,\sigma'} + \hat{c}_{i,\sigma} \hat{c}_{i,\sigma'}) + H.c.$ are allowed but, as we shall see, they cancel out in $\delta \hat{H}^{\text{eff}}$.

We rewrite the effective Hamiltonian in Eq. (13) as $\hat{H}^{\text{eff}} = \hat{H}_{ss} + \sum_{i=1}^{4} \delta \hat{H}_i$, where

$$\delta \hat{H}_1 = \frac{1}{E - \hat{H}_{dd}} \frac{\hat{H}_{dd}}{E - \hat{H}_{tt}} \hat{H}_{td},$$

$$\delta \hat{H}_2 = \frac{1}{E - \hat{H}_{tt}} \hat{H}_{td},$$

$$\delta \hat{H}_3 = \frac{1}{E - \hat{H}_{tt}} \frac{\hat{H}_{td}}{E - \hat{H}_{tt}} \hat{H}_{ts} + H.c.,$$

$$\delta \hat{H}_4 = \frac{1}{E - \hat{H}_{tt}} \frac{\hat{H}_{td}}{E - \hat{H}_{tt}} \frac{\hat{H}_{ts}}{E - \hat{H}_{tt}} \hat{H}_{td}.$$ (18)

In the terms $\delta \hat{H}_3$ and $\delta \hat{H}_4$ above we have replaced $\hat{H}_{td}$ and $\hat{H}_{dd}(E - \hat{H}_{tt})^{-1}\hat{H}_{td}$ by $\hat{H}_{dd}$ (the neglected term gives contributions that are higher than fourth order in the expansion parameters).
We next expand each $\hat{\delta H}_1$ to fourth order in the parameters $t_0/J_z$ and $J_z/J_z$. Since the energy $E$ is of the order $J_z$, the fractions appearing in each term can be brought to a form $1/(1-\hat{X})$ with $\hat{X}$ being small in the expansion parameters. We then approximate $1/(1-\hat{X}) = 1 + \hat{X} + \hat{X}^2$. In the following we summarize the explicit calculation of each term.

1. Evaluation of $\delta\hat{H}_1$

For $\delta\hat{H}_1$ we find

$$\delta\hat{H}_1 \approx -\frac{4}{3\hat{J}_k} \hat{H}_{sd}(1 + \hat{A} + \hat{B} + \hat{C} + \hat{A}^2 + \hat{B}^2)\hat{H}_{ds}, \quad (19)$$

where

$$\hat{A} = \frac{4}{3\hat{J}_k} \left[ E_0^0 - \hat{H}_0^0 + \hat{\varepsilon}_0^0 (1 - \hat{n}_0^0) \right], \quad (20)$$

$$\hat{B} = \frac{4}{3\hat{J}_k} \left[ -3/4 - \hat{S}_{tot}(\hat{S}_{tot} + 1) + \hat{S}_d^2 \right], \quad (21)$$

$$\hat{C} = \frac{4}{3\hat{J}_k} \hat{H}_{ds}\hat{H}_{ds}. \quad (22)$$

In Eq. (19), we omitted the product terms $\hat{A}\hat{C}, \hat{B}\hat{C}, \hat{C}^2$ since their contribution is higher than fourth order, while the contribution of $\hat{A}\hat{B} + \hat{B}\hat{A}$ can be shown to vanish identically.

To keep track of the various contributions for each of the $\delta\hat{H}_1$, we label them in the following by $\delta\hat{H}_{1j}$. These terms are understood to act only in the space of the reduced dot while the Kondo spin and the spin on the dot are locked into a singlet. The operators $\delta\hat{H}_{1j}$ in the reduced dot space are obtained by taking a partial expectation value $\langle \ldots \rangle$, in the singlet state. The corresponding operators in the full space are given, respectively, by $P_\uparrow \langle \ldots \rangle, P_\uparrow \langle \ldots \rangle_c$. In the Appendix we list several relations that are useful in calculating the expectation values of various operators in the singlet space.

The most dominant contribution to Eq. (19) arises from the unity operator term (in the round brackets). We find

$$\delta\hat{H}_{1,1} = -\frac{4}{3\hat{J}_k} \langle \hat{H}_{sd}\hat{H}_{ds} \rangle = -\frac{4}{3\hat{J}_k} \langle \hat{H}_{\text{hop}}^{(0,1)} \hat{H}_{\text{hop}}^{(0,1)} \rangle = -\frac{4}{3\hat{J}_k} \langle t_{00}^{\uparrow \downarrow} \rangle^2, \quad (23)$$

where we have substituted $\hat{H}_{sd}$ by the hopping Hamiltonian between sites 0 and 1,

$$\hat{H}_{\text{hop}}^{(0,1)} = \sum_{\sigma} t_{00,\sigma} \hat{c}_{1,0,\sigma} + \text{H.c.}, \quad (24)$$

and used Eq. (A7). Alternatively, $\hat{H}_{sd}\hat{H}_{ds}$ describes the spin transitions illustrated in Fig. 2(a), and the result in Eq. (23) can be derived using Table I in the Appendix to sum up all the corresponding transition pathways.

The term containing $\hat{A}$ in Eq. (19) yields corrections that are second order in $t_0/J_z$. Using the difference in the values of $\hat{H}_0^0, \hat{n}_0^0$ in the singlet and doublet subspaces, and Eqs. (A11) and (A12), we find

$$\langle \hat{H}_{sd}\hat{H}_{ds} \rangle = \frac{|t_{00}|^2}{2} \sum_{\sigma} \langle \hat{c}_{1,0,\sigma} \hat{c}_{1,0,\sigma} + \hat{c}_{1,0,\sigma} \hat{c}_{1,0,\sigma} \rangle. \quad (28)$$

Using the identities (A5) and (A6), Eq. (28) can be simplified to give Eq. (A15) in the Appendix. Using $S_d^z = \hat{S}_{tot}$ in the singlet subspace, we obtain

FIG. 2. Spin-transition diagrams used to derive the effective strong-coupling Hamiltonian in Eq. (55). All transitions connect two singlet states, characterized by the quantum numbers $S_{K0} = 0, S_{K0,z} = 0$. The intermediate transition pathways involve combinations of doublet ($S_{K0} = 1/2, S_{K0,z} = \pm 1/2$) and triplet ($S_{K0} = 1, S_{K0,z} = \pm 1/2$) states.

$$\delta\hat{H}_{1,2} = -\frac{4}{3\hat{J}_k} \langle \hat{H}_{sd}\hat{A}\hat{H}_{ds} \rangle = -\frac{4}{3\hat{J}_k} \langle t_{01}^\uparrow \hat{c}_{1,0,0}^\uparrow \hat{c}_{1,0,0}^\uparrow \rangle \left[ (\varepsilon_0^c - \varepsilon_1^c) + (\varepsilon_0^c - \varepsilon_0^c)^2 \right] \left[ (\varepsilon_1^c - \varepsilon_1^c) + (\varepsilon_1^c - \varepsilon_0^c) \right]. \quad (25)$$

The constant shift in Eq. (25) can be incorporated into $\hat{H}_{ss}$ in Eq. (14) by redefining $\varepsilon_0^c$, while the one-body operator in Eq. (25) can be incorporated into the Hamiltonian $\hat{H}_0^0$ by redefining the site energy $\varepsilon_1^c$ and the hopping amplitude $t_1$. The term involving $\hat{B}$ in Eq. (19) contributes only for a finite exchange interaction ($J_x \neq 0$). We have

$$\langle \hat{H}_{sd}\hat{H}_{ds} \rangle = \frac{|t_{00}|^2}{2} \sum_{\sigma} \langle \hat{c}_{1,0,0} \hat{c}_{1,0,0}^\dagger + \hat{c}_{1,0,0} \hat{c}_{1,0,0} \rangle. \quad (29)$$

Using the identities (A5) and (A6), Eq. (29) can be simplified to give Eq. (A15) in the Appendix. Using $S_d^z = \hat{S}_{tot}$ in the singlet subspace, we obtain

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Finally, the contribution from $\delta H_{1,3}$ is a spin invariant in the reduced dot space.

The term involving $\hat{C}$ in Eq. (19) is given by

$$\delta H_{1,3} = - \frac{4}{3 J_k} \langle \hat{H}_{sd} \hat{d} \hat{H}_{dt} \rangle_s = 2 J_k \left( \frac{4 |t_0|^2}{3 J_k} \right) \hat{s}_1 \cdot \hat{S}_d. \quad (29)$$

We note that $\delta H_{1,3}$ is a spin invariant in the reduced dot space.

This term appears in the conventional Kondo problem (where $J_s = 0$) and is known as the Nozières term. Nozières found that this term yields an effective interaction in the singlet space that repels opposite spins on site 1. This term is induced by virtual transitions of the type singlet-doublet-triplet-singlet. Once we insert a triplet projection $\hat{P}_t$ in the rhs of Eq. (30), i.e., we write the corresponding singlet expectation value as $\langle \hat{H}_{sd} \hat{d} \hat{H}_{dt} \rangle_s$, we can replace both $\hat{H}_{sd}$ and $\hat{H}_{dt}$ by the hopping Hamiltonian $\hat{H}_{\text{hop}}$ between sites 0 and 1 [see Eq. (24)]. Using $\hat{H}_{\text{hop}}^{(1)} \hat{P}_t \hat{H}_{\text{hop}}^{(1)} = \hat{H}_{\text{hop}}^{(1)} \hat{P}_t \hat{H}_{\text{hop}}^{(1)} - \hat{H}_{\text{hop}}^{(1)} \hat{P}_t \hat{H}_{\text{hop}}^{(1)}$, we have

$$\langle \hat{H}_{sd} \hat{d} \hat{H}_{dt} \rangle = \langle \hat{H}_{\text{hop}}^{(1)} \hat{P}_t \hat{H}_{\text{hop}}^{(1)} \rangle_s - \langle \hat{H}_{\text{hop}}^{(1)} \hat{P}_t \hat{H}_{\text{hop}}^{(1)} \rangle_s. \quad (31)$$

With the help of Eqs. (A1), (A2), (A7), and (A11), we then find

$$\delta H_{1,4} = -3 \left( \frac{4}{3 J_k} \right)^3 \left| t_0 \right|^2 \left| s_1 \right|^2 = - \frac{16}{3 J_k} \left| t_0 \right|^2 \hat{n}_1 (2 - \hat{n}_1). \quad (32)$$

An alternative way to calculate $\delta H_{1,4}$ is to use the spin diagram in Fig. 2(e). It can be reduced to the transition diagram in Fig. 2(b) with the help of Table I in the Appendix.

The Nozières term (32) vanishes when site 1 is either empty ($n_1 = 0$) or doubly occupied ($n_1 = 2$) but is negative for $n_1 = 1$, thus favoring a singly occupied site 1.

The contribution from $A^2$ in Eq. (19) is evaluated using Eqs. (A13) and (A14) and leads to a constant shift

$$\delta H_{1,5} = - \frac{4}{3 J_k} \langle \hat{H}_{sd} \hat{d} \hat{d} \hat{H}_{dt} \rangle_s = - \left( \frac{4}{3 J_k} \right)^3 \left| t_0 \right|^2 \left| t_1 \right|^2 + \left( \epsilon_0 \right)^2 + \left( \epsilon_1 \right)^2 - 2 \epsilon_0 \epsilon_1. \quad (33)$$

Finally, the contribution from $B^2$ in Eq. (19) is found to be

$$\delta H_{1,6} = - \left( \frac{4}{3 J_k} \right)^3 \left| t_0 \right|^2 \left| t_1 \right|^2 \left( \hat{s}_d \cdot \hat{S}_d + 2 \hat{s}_d \cdot \hat{S}_d \right), \quad (34)$$

where we have used Eq. (A16).

2. Evaluation of $\delta H_2$

We next turn to the singlet-triplet transitions as described by $\delta H_2$ in Eq. (18). The corresponding expression for $\delta H_2$ is given by

$$\delta H_2 = - \frac{1}{J_k} \hat{H}_{st} \langle 1 + \hat{D} \hat{E} \hat{E}^2 \rangle \hat{H}_{st}, \quad (35)$$

where

$$\hat{D} = \frac{1}{J_k} \left[ E_0 - \hat{H}_0 \right], \quad (36)$$

$$\hat{E} = \frac{J}{J_k} \left[ - S_{tot}(S_{tot} + 1) + \hat{S}_d^2 + 2 \hat{s}_d \cdot \hat{S}_d \right]. \quad (37)$$

In Eq. (35) we omitted the terms $\hat{D}^2$ and $\hat{D} \hat{E}$, which can be shown to vanish.

The dominating term in Eq. (35) is the one involving the unity operator. The corresponding term induces a spin transition as in Fig. 2(b). Using $\hat{H}_{st} = -2 J_k \hat{P}_t \hat{s}_d \hat{S}_d \hat{P}_t$ and Eq. (A9) we find

$$\delta H_{2,1} = - \frac{1}{J_k} \langle \hat{H}_{st} \hat{D} \hat{H}_{st} \rangle_s = - \frac{J^2}{J_k} \hat{S}_d^2. \quad (38)$$

The same result can also be obtained with the help of Table II in the Appendix. The contribution $\delta H_{2,2}$ induced by the term involving $\hat{D}$ in Eq. (35) can be simplified using $\langle \hat{H}_{st} \hat{D} \hat{H}_{st} \rangle_s = \hat{f}_d \hat{s}_d \hat{S}_d \hat{S}_d$. Since $\hat{S}_d^2$ commutes with the scalar operator $\hat{H}_{st}$, we find

$$\delta H_{2,2} = - \frac{1}{J_k} \langle \hat{H}_{st} \hat{D} \hat{H}_{st} \rangle_s = 0. \quad (39)$$

The contribution $\delta H_{2,3}$ induced by the term $\hat{E}$ in Eq. (35) can also be simplified since $\hat{S}_d^2$ acts in the reduced dot space (and therefore has identical action in the singlet and triplet manifolds). The resulting expression gives rise to transition pathways as shown in Fig. 2(c), the sum over which is further simplified using Eq. (A10) to give

$$\delta H_{2,3} = - \frac{J^2}{J_k} \langle \hat{H}_{st} \hat{E} \hat{H}_{st} \rangle_s = \frac{J^2}{J_k} \hat{S}_d^2. \quad (40)$$

The last term $\delta H_{2,4}$ in Eq. (35), containing $\hat{E}^2$, is found to be $\delta H_{2,4} = - \langle 16 f_s^2 t_1^2 \rangle \langle \hat{s}_d \hat{S}_d \rangle^4$ and corresponds to the transition diagram in Fig. 2(g). Since $\hat{H}_{st}$ (a scalar operator) commutes with $\hat{S}_d^2$, all other terms vanish identically. Using Eq. (A3) this expression can be simplified to give

$$\delta H_{2,4} = - \frac{1}{J_k} \langle \hat{H}_{st} \hat{E} \hat{E} \hat{H}_{st} \rangle_s = - \frac{J^4}{J_k} \hat{S}_d^2. \quad (41)$$

3. Evaluation of $\delta H_3$

Following Eq. (18), the subsequent contribution, $\delta H_3$, involves transitions to both the doublet and the triplet subspaces.
\[ \delta H_3 = \frac{4}{3J_k^4} \tilde{\mathcal{H}}_d(1 + \tilde{D} + \tilde{E}) \tilde{\mathcal{H}}_{pd}(1 + \tilde{A} + \tilde{B}) \tilde{\mathcal{H}}_{ds} + \text{H.c.}, \]

where the operators \( \tilde{A} - \tilde{E} \) are the same as introduced above. Contributions involving products between \( \tilde{D}, \tilde{E} \) and \( \tilde{A}, \tilde{B} \) are higher than fourth order in the expansion parameters and are therefore not considered here.

The dominant contribution, \( \delta H_{3,1} \), originates in the transitions shown in Fig. 2(d). Using Table I we can simplify this transition diagram to the one of Fig. 2(b), for which we obtain

\[ \delta H_{3,1} = \frac{4}{3J_k^4} \langle \tilde{\mathcal{H}}_d \tilde{\mathcal{H}}_{pd} \tilde{\mathcal{H}}_{ds} \rangle + \text{H.c.} = \frac{16J^2}{3J_k^4} |t_0|^2 \tilde{S}_1 \cdot \tilde{S}_2. \] (43)

To simplify the term \( \delta H_{3,2} \), involving \( \tilde{D} \) in Eq. (42), we make use of \( \langle \tilde{H}_d \tilde{H}_d \rangle = 0 \) and find

\[ \delta H_{3,2} = \frac{4}{3J_k^4} \tilde{\mathcal{H}}_d \tilde{\mathcal{H}}_{pd} \tilde{\mathcal{H}}_{ds} + \text{H.c.} = \frac{8J^2}{3J_k^4} |t_0|^2 (E_0 - \tilde{H}_0) \tilde{S}_1 \cdot \tilde{S}_2' + \text{H.c.} \] (44)

The diagonal matrix elements of \( \delta H_{3,2} \) (when evaluated in the eigenstates of the reduced dot) vanish. Off-diagonal matrix elements enter in second-order perturbation theory and are related to be

\[ \delta H_{3,1 \ldots 3} = 0. \] (45)

The next term \( \delta H_{3,3} \), produced by \( \tilde{E} \) in Eq. (42), can be calculated using Table I and the diagram in Fig. 2(c), where a sum of all relevant terms can be identified with Eq. (44), yielding

\[ \delta H_{3,3} = \frac{4}{3J_k^4} \langle \tilde{\mathcal{H}}_d \tilde{\mathcal{H}}_{pd} \tilde{\mathcal{H}}_{ds} \rangle + \text{H.c.} = -\frac{8J^2}{3J_k^4} |t_0|^2 \tilde{S}_1 \cdot \tilde{S}_2 + \text{H.c.} \] (46)

The contribution \( \delta H_{3,4} \) induced by \( \tilde{A} \) in Eq. (42), is calculated to be

\[ \delta H_{3,4} = \frac{4}{3J_k^4} \langle \tilde{\mathcal{H}}_d \tilde{\mathcal{H}}_{pd} \tilde{\mathcal{H}}_{ds} \rangle + \text{H.c.} = \frac{32J^2}{9J_k^4} |t_0|^2 \tilde{S}_1 \cdot \tilde{S}_2 (E^{(0)} - \tilde{H}_0^2) + \text{H.c.} \] (47)

Following the same arguments used for the evaluation of \( \delta H_{3,2} \), we find \( \delta H_{3,4} = 0). \)

The term \( \tilde{B} \) in Eq. (42) gives rise to a transition diagram of the type shown in Fig. 2(d) which can be reduced to a diagram as in Fig. 2(b) with the help of Table III. Further simplifications involving several of the equations in the Appendix yield

\[ \delta H_{4,5} = \frac{4}{3J_k^4} \langle \tilde{\mathcal{H}}_d \tilde{\mathcal{H}}_{pd} \tilde{\mathcal{H}}_{ds} \rangle + \text{H.c.} = -\frac{J^2}{3J_k^4} |t_0|^2 \tilde{S}_0' + 2S_1 \cdot \tilde{S}_2 + \text{H.c.}. \] (48)

4. Evaluation of \( \delta H_4 \)

To determine the terms contributing to \( \delta H_4 \) (up to fourth order), we make the following approximations in Eq. (18): \( (E - \tilde{H}_0)^{-1} = -1/J_k \) and \( (E - \tilde{H}_{ds})^{-1} = -4/(3J_k) \). The resulting expression for \( \delta H_4 \) corresponds to a transition diagram as in Fig. 2(f). Using Eqs. (A1), (A4), (A9), and (A10) we find

\[ \delta H_4 = -\frac{4J^2}{3J_k^4} \langle \tilde{\mathcal{H}}_d \tilde{\mathcal{H}}_{pd} \tilde{\mathcal{H}}_{ds} \rangle = -\frac{4J^2}{3J_k^4} |t_0|^2 \tilde{S}_0' + 2S_1 \cdot \tilde{S}_2 + \text{H.c.}. \] (49)

Alternatively, we can obtain this result using Table I to get a transition diagram as in Fig. 2(c), which can then be simplified using Eq. (A4).

5. Additional terms

In the above calculations, we have replaced the energy eigenvalue \( E \) in Eq. (13) by its unperturbed value \( E^{(0)} \) in Eq. (17). However, additional terms to the effective Hamiltonian are found when corrections to \( E^{(0)} \) are included self-consistently. To the order we are interested in, it is sufficient to consider \( \delta H_{1,1} \) and \( \delta H_{2,1} \) [see Eqs. (23) and (38), respectively] as corrections to \( E^{(0)} \)

\[ E \approx E^{(0)} - \frac{4|t_0|^2}{3J_k^4} S_{0 \perp} (S_{0 \perp} + 1). \] (50)

Adding these correction terms inside the square brackets in Eq. (20) gives the following term

\[ \delta H_{1,7} = \left( \frac{4}{3J_k^4} \right)^3 |t_0|^4 + \left( \frac{4J}{3J_k^4} \right) |t_0|^2 \tilde{S}_0' + \text{H.c.}. \] (51)

A similar correction inside the square brackets of Eq. (36) yields

\[ \delta H_{2,5} = \frac{4J^2}{3J_k^4} |t_0|^2 \tilde{S}_0' + \frac{J^2}{J_k^4} \tilde{S}_0'' + \text{H.c.}. \] (52)

Up to fourth order, the corrections from Eq. (50) do not lead to additional terms in \( \delta H_3 \) and \( \delta H_4 \).
\[ H_{\text{eff}} = \eta + H_d - \left( \frac{4}{3J_k} \right)^3 |t_1|_0^4 (3S^2_d - 1) \]
\[ + J_k \left( \frac{80J_d}{9J_k} - \frac{536J^2_d}{27J^2_k} |t_0|_0^2 \right) \hat{\mathbf{S}}_d^\dagger \right) + \frac{J^4_d}{J^2_k} \hat{S}^4_d, \]

where \( \eta \) is a constant
\[ \eta = \frac{e^0_s - \frac{3(J_k + J_s)}{4} |t_0|^2}{3J_k} - \left( \frac{4}{3J_k} \right)^3 |t_1|^2 |t_0|^2 + (\epsilon^0_s - \epsilon^1_s)^2 (\epsilon^0_s - \epsilon^1_s), \]

and \( \tilde{H}_d \) describes a renormalized universal Hamiltonian for the reduced dot
\[ \tilde{H}_d = \tilde{H}_s - \bar{J}_s \bar{S}_d^2. \]

\( \tilde{H}_s \) is a renormalized one-body Hamiltonian of the reduced dot, obtained from \( H_s \) by redefining \( e^0_s \) and \( t_1 \) according to Eqs. (26) and (27), respectively. This tridiagonal Hamiltonian can be rediagonalized \( \tilde{H}_s = \sum_{\alpha=\uparrow,\downarrow} \tilde{\epsilon}^{\alpha}_s \tilde{a}^\dagger_{\alpha n} \tilde{a}_{\alpha n} \) to define new effective single-particle orbitals \( \tilde{a}^\dagger_{\alpha n} = 0 \) and energies \( \tilde{\epsilon}^{\alpha}_s \) of the reduced dot. \( \bar{J}_s \) in Eq. (55) is a renormalized exchange constant
\[ \bar{J}_s = J_k \left( 1 + \frac{J_d}{J_k} - \frac{J^2_d}{J^2_k} + \frac{112J_d |t_0|^2}{27 J^2_k} \right). \]

The most dominant contributions in Eq. (56) are positive and thus lead to a stronger exchange interaction in the reduced dot than in the original dot, \( \tilde{J}_s > J_s \). Since the Kondo spin and the spin at site 0 are coupled to a singlet, the spin of the reduced dot \( \bar{S}_d = \bar{S}_\text{tot} \), and is thus conserved (i.e., \( S'_d = S_\text{tot} \) and \( M'_d = M_\text{tot} \) are good quantum numbers).

The effective Hamiltonian of the reduced dot contains several additional interaction terms, see Eq. (53). The term proportional to \( (3S^2_d - 1) \) is the Nozières term, known from the conventional Kondo problem in the absence of exchange \( (J_s=0) \). The term proportional to \( \hat{\mathbf{S}}_d^\dagger \hat{\mathbf{S}}_d \) is an effective interaction in the reduced dot that is induced by the finite exchange interaction \( (J_s \neq 0) \) and describes an exchange interaction between the spin at site 1 and the spin of the reduced dot. This exchange interaction is to leading-order antiferromagnetic but depending on the particular values of \( J_k \) and \( J_s \) it can become ferromagnetic. The last term in Eq. (53) is a four-body term but it can be easily evaluated in terms of the conserved total spin \( \bar{S}_\text{tot} \) (since \( \bar{S}_d = \bar{S}_\text{tot} \) in the singlet space). It can be combined with the renormalized exchange interaction in the reduced dot by defining an exchange coupling that depends on the total spin.

IV. EIGENVALUES OF THE EFFECTIVE HAMILTONIAN

The effective Hamiltonian of the reduced dot is valid to fourth order in \( |t_0|/J_k \) and \( J_d/J_k \) (when its terms are measured in units of \( J_k \)). To determine its eigenenergies to this fourth order, it is sufficient to solve \( \tilde{H}_{\text{eff}} \) in first-order perturbation theory. Both \( \tilde{H}_d \) and \( \tilde{S}_d^\dagger \) are diagonal in a basis of good orbital occupations and good total spin of the reduced dot while a second-order perturbation theory of the remaining interaction terms leads to terms that are higher than fourth order in the combined power of \( t_0/J_k \) and \( J_d/J_k \).

As required by first-order perturbation theory, we evaluate the expectation value of \( \tilde{H}_{\text{eff}} \) in the unperturbed basis, i.e., in the eigenbasis of the renormalized universal Hamiltonian \( \tilde{H}_d \) of the reduced dot. For simplicity, we will denote the good spin eigenstates of \( \tilde{H}_d \) by \( \left| \xi \right\rangle \) and the corresponding expectation values by \( \langle \left\langle \cdots \right\rangle_{\xi} \).

The calculation of \( \langle \xi^2 \rangle_{\xi} \) in Eq. (53) simplifies for the lowest eigenstate of \( \tilde{H}_d \) at each given spin value \( S'_d = S_\text{tot} \). Those eigenstates of \( \tilde{H}_d \) with \( M'_d = S'_d \) have a maximal spin projection with only spin-up electrons in singly occupied levels and thus have \( \bar{n}_\alpha \) as good quantum numbers (in contrast to a general eigenstate of \( \tilde{H}_d \) where only the orbital occupation numbers \( \bar{n}_\alpha \) are well defined). For these states we have
\[ \langle \xi^2 \rangle_{\xi} = \frac{3}{4} \sum_{\sigma=\uparrow,\downarrow} \langle \hat{n}_{1\sigma} (1 - \hat{n}_{1-\sigma}) \rangle_{\xi} = \frac{3}{4} \sum_{\sigma=\uparrow,\downarrow} \langle \hat{n}_{1\sigma} (1 - \hat{n}_{1-\sigma}) \rangle_{\xi}. \]

The occupation \( \langle \hat{n}_{1\sigma} \rangle_{\xi} \) can be calculated from \( \langle \hat{n}_{1\sigma} \rangle_{\xi} = \sum_{\mu=1}^{N-1} \langle U_\sigma | \hat{a}^\dagger_{\mu} \hat{a}_{\mu} \rangle_{\xi} \) where \( U_\sigma \) is the unitary matrix (of order \( N-1 \)) transforming between the renormalized single-particle orbitals of the reduced dot (with creation operators \( \hat{a}^\dagger_{\mu} \)) and the site basis states \( \mu = 1, \ldots, N-1 \).

For the good spin eigenstates of \( \tilde{H}_d \) with \( M'_d = S'_d \), the expectation value of \( \hat{\mathbf{S}}_d^\dagger \hat{\mathbf{S}}_d \) is given by
\[ \langle \hat{\mathbf{S}}_d^\dagger \hat{\mathbf{S}}_d \rangle_{\xi} = \frac{1}{2} \sum_{\sigma=\uparrow,\downarrow} \sigma \langle \hat{\mathbf{S}}_d^\dagger \hat{\mathbf{S}}_d \rangle_{\xi} = \frac{1}{2} \sum_{\sigma=\uparrow,\downarrow} \sigma \langle \hat{n}_{1\sigma} \rangle_{\xi}^2. \]

Using Eqs. (57) and (59), we can calculate the lowest many-body eigenenergy for each total spin value of the Kondo Hamiltonian (2) up to fourth order in \( |t_0|/J_k \) and \( J_d/J_k \).

V. COMPARISON WITH EXACT NUMERICAL DIAGNOLIZATION

To validate our expression (53) for the effective Hamiltonian, we compare our analytical results for the many-body energies in the strong-coupling limit with an exact numerical diagonalization of the Hamiltonian (2) in a good spin basis scheme we developed previously.37

As a first test, we compare results for the lowest energy of a given total spin (e.g., \( S_\text{tot} = 3 \)). In Fig. 3, we show the difference \( \Delta E \) (in units of \( J_k \)) between the energy determined...
the effective Hamiltonian to be given by $H_{9257}$.

versal Hamiltonian of the reduced dot, i.e., when we assume to include interaction terms beyond the renormalized
gram in the two-dimensional parameter space of $S_{20849}$.

The exact spin transition curves can be probed experimentally.40 The ground-state spin undergoes successive transitions to higher values $J_{\text{tot}}=0$ and $Js/\hbar=0.52$ chosen from an arbitrary random matrix realization but with nonfluctuating orbital wave functions $\phi(0)=1/\sqrt{N}$. The results, shown for an arbitrary but fixed value $J_{\text{tot}}/\hbar=0.52$ (red dots), behave like $1/(J_{\bar{\rho}}\hbar)^2$ (black solid line), expected for a strong-coupling expansion up to fourth order in $1/(J_{\bar{\rho}}\hbar)$ in the limit $J_{\bar{\rho}}\hbar \to \infty$.

from the effective Hamiltonian (53) and the energy found from exact numerical diagonalization of Eq. (2) as a function of $J_{\bar{\rho}}\hbar$ (at an arbitrary but fixed value $J_{\text{tot}}/\hbar=0.52$). This energy difference $\Delta E$ should scale as $1/(J_{\bar{\rho}}\hbar)^2$, which is the next order correction beyond the accuracy considered here. The results shown in Fig. 3 confirm this scaling behavior and thereby the accuracy and completeness of our effective Hamiltonian. Similar results (not shown here) are found for other values of $S_{\text{tot}}$ and for both even and odd number of electrons in the dot.

It is interesting to study the ground-state value of the total spin $S_{\text{tot}}$. This quantity was studied theoretically21,24,27 and can be probed experimentally.48 The ground-state spin $S_{\text{tot}}$ undergoes successive transitions to higher values (known as the Stoner staircase) when the exchange coupling constant $J_s$ is varied between $J_s=0$ and $J_s \sim \delta$. The transition steps in the Stoner staircase are shifted by the Kondo interaction. In Fig. 4 we show numerical results for the ground-state spin diagram in the two-dimensional parameter space of $J_s/\delta, J_{\bar{\rho}}\hbar$ for a particular mesoscopic realization of the single-particle Hamiltonian of the dot. The exact spin transition curves (colored lines) that separate regions of fixed ground-state spin $S_{\text{tot}}$ are monotonically decreasing for $J_{\bar{\rho}}\hbar \leq 1$ and monotonically increasing for $J_{\bar{\rho}}\hbar \geq 1$. Note also that for the particular mesoscopic realization chosen in Fig. 4, certain values of $S_{\text{tot}}$ (e.g., $S_{\text{tot}}=1, 3$) never become the ground-state values of the total spin in the weak-coupling limit. In contrast, the ground-state spin assumes these values in the strong-coupling limit. Our analytical results in this limit, shown by the dashed lines in Fig. 4, are in very good agreement with the exact numerical results down to values of $J_{\bar{\rho}}\hbar=2$. The dotted black lines in Fig. 4 are the corresponding transition lines when we do not include interaction terms beyond the renormalized universal Hamiltonian of the reduced dot, i.e., when we assume the effective Hamiltonian to be given by $H_{\text{eff}}=\eta+H_{\text{d}}^{\gamma}$ [see Eq. (53)]. These dotted curves converge much slower to the full numerical solutions (colored lines) than the dashed curves determined from the effective Hamiltonian (53). However, both the dashed and the dotted lines reproduce the monotonic increase of the exact transition curves with $J_{\text{tot}}$ for $J_{\bar{\rho}}\hbar \geq 2$. We conclude that this monotonic increase originates in the renormalization of the effective exchange-coupling constant in the reduced dot, $\bar{J}_{\text{tot}}=J_{\text{tot}}(1+J_{\text{tot}}/J_{\bar{\rho}}\hbar+\cdots)$, which is contained in the approximations used for both the dashed and the dotted lines. The renormalized exchange constant $\bar{J}_{\text{tot}}$ decreases with increasing $J_{\text{tot}}$ causing the spin-transition curves to move upward with increasing $J_{\text{tot}}$.

VI. DISCUSSION AND CONCLUSION

We have investigated the strong-coupling limit of the Kondo problem when the screening electrons reside in a large quantum dot that is described by the universal Hamiltonian. Unlike the conventional Kondo problem, the model considered here has a discrete single-particle spectrum and includes electron-electron interactions in the form of a ferromagnetic exchange interaction.

We have followed here a procedure that was originally proposed in Ref. 36 for the conventional Kondo problem in the absence of exchange correlations in the electron gas. As pointed out there, one can find the effective Hamiltonian at strong Kondo coupling ($T\ll T_K$) by considering the bare the absence of exchange correlations in the electron gas. As pointed out there, one can find the effective Hamiltonian at strong Kondo coupling ($T\ll T_K$) by considering the bare
strong-coupling limit $J \gg t_0$. This bare strong-coupling limit is the one for which we have now provided a closed expression of all effective interaction terms up to fourth order in $t_0/J$ and $J/J$, when the electron gas is described by the universal Hamiltonian. However, if the bandwidth $D$ of this electron gas is very large $D \gg \tilde{D}$, the limit of strong coupling can be effectively reached at much smaller values of $J_0$ than those of the bare limit. For such a system with large bandwidth, the strong-coupling limit corresponds to a Kondo temperature $T_K$ that is larger than the system’s temperature and average level spacing, $T_K \gg T, \tilde{D}$. An important insight in Kondo theory is that the effective Hamiltonians of both strong-coupling limits are related by a scaling analysis\(^{41}\) in which the Kondo Hamiltonian is renormalized by successive truncations of the bandwidth $D$, leaving the low-energy physics unchanged. As the reduced bandwidth $\tilde{D} \rightarrow 0$ (or equivalently $T \rightarrow 0$), the renormalized Kondo coupling constant diverges.\(^{42}\) The coupling constants of the various terms in the effective strong-coupling Hamiltonians are typically determined by fitting the lowest excitations of the effective Hamiltonian with those obtained by a numerical solution of the full problem. For a detailed review of this procedure see, e.g., Refs. 2, 42, and 43.

By deriving the effective Hamiltonian $\hat{H}^\text{eff}$ for the bare strong-coupling limit [see Eq. (53)], we have completed successfully the first step in our goal to understand the strong-coupling limit of the Kondo problem in the presence of exchange correlations in the mesoscopic electron gas. We found that the exchange interaction in the universal Hamiltonian gives rise to two additional terms in $\hat{H}^\text{eff}$: a four-body contribution $\hat{S}^{d_1}_{\alpha}^{d_4}$ that can be absorbed into a spin-dependent exchange coupling in the reduced dot, and an interaction term $\hat{s}_1 \cdot \hat{S}_{d}^{d_4}$ that describes an exchange interaction between an electron in the vicinity of the Kondo spin and the total spin of the reduced dot. This term is induced by the virtual polarization of the Kondo singlet involving excitations to both the doublet and triplet subspaces. Unlike the conventional Kondo problem ($J_0 = 0$), this interaction is nonlocal as it involves the total spin of all sites of the electron gas (after the removal of an electron at site 0).

It would be of interest to identify similar interaction terms in the low-temperature behavior of a correlated Kondo state with a large bandwidth $D$. Our numerical diagonalization method of Ref. 27 is limited to a rather small bandwidth, e.g., $N=11$ levels for the results shown in Figs. 3 and 4. For such small bandwidths, the bare and renormalized strong-coupling limits essentially coincide, and we could use our numerical diagonalization method to validate the analytical derivations. For a numerical solution at larger bandwidths, a numerical renormalization group (NRG) technique\(^{42}\) might be useful. The challenge for NRG is the inclusion of nonlocal correlations induced by the exchange interaction in the universal Hamiltonian. With numerical solutions at hand, it would be interesting to investigate whether the renormalization of the large bandwidth Kondo problem induces any other “leading irrelevant” interaction terms around the strong-coupling fixed point beyond those we have identified here.

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**APPENDIX**

In this appendix we provide various expressions that are useful for deriving the effective Hamiltonian. The notation used here follows the convention of Ref. 44.

The product of a hopping operator $\Sigma_{\alpha} c_{1,\alpha} c_{0,\alpha}$ between sites 0 and 1 and its Hermitian conjugate can be expressed in terms of the occupation number $(n_0, n_1)$ and spin $(s_0, s_1)$ operators at these sites,

\[
\sum_{\alpha, \alpha'} c_{1,\alpha} c_{0,\alpha}^\dagger c_{0,\alpha'}^\dagger = n_0 (1 - n_1/2) - 2s_0 \cdot s_1, \quad (A1)
\]

\[
\sum_{\alpha, \alpha'} c_{1,\alpha} c_{0,\alpha}^\dagger c_{0,\alpha'} c_1 = n_1 (1 - n_0/2) - 2s_0 \cdot s_1. \quad (A2)
\]

The spin raising and lowering operators, $\hat{S}_\pm = \hat{S}_x \pm i\hat{S}_y$, satisfy, together with $\hat{S}_z$, the usual su(2) commutation relations

\[
[\hat{S}_-, \hat{S}_+] = \pm \hat{S}_z, \quad [\hat{S}_+, \hat{S}_-] = 2\hat{S}_z. \quad (A3)
\]

\[
\hat{S}_x^2 = \hat{S}_y^2 + (\hat{S}_z)^2. \quad (A4)
\]

Other operator relations between $\hat{S}_d'_{d'}$ and operators on site 1 are

\[
\hat{S}^2_{1,d'} \hat{c}_{1,z} = \hat{c}_{1,z} \hat{S}^2_{d'} + 3\hat{c}_{1,z} \hat{S}^2_{d'z} \mp \hat{c}_{1,z} \hat{S}^2_{d'z}, \quad (A5)
\]

**TABLE I.** Matrix elements $\langle \eta_{\phi} | \hat{O}_1 | \phi_\psi \rangle$ of the operator $\hat{O}_1 = \hat{H}_0^{(0,1)} | \phi_\psi \rangle \langle \eta_{\phi} |$. The corresponding states $\phi_\psi$ ($\eta_{\phi}$) are listed in the left column (top row), and are characterized by the quantum numbers $S_{K0} = S_{K0}^{(1,1)}$.

| $\langle \eta_{\phi} | \phi_\psi \rangle$ | $|0, 0\rangle$ | $|1, -1\rangle$ | $|1, 0\rangle$ | $|1, +1\rangle$ |
|-----------------------------|----------------|----------------|----------------|----------------|
| $|0, 0\rangle$              | 1              | $\sqrt{2} \hat{S}_{1z}$ | 2$\hat{S}_{1z}$  | $-\sqrt{2} \hat{S}_{1z}$ |
| $|1, -1\rangle$             | $\sqrt{2} \hat{S}_{1z}$ | 1 + 2$\hat{S}_{1z}$ | $-\sqrt{2} \hat{S}_{1z}$ | 0             |
| $|1, 0\rangle$              | 2$\hat{S}_{1z}$ | $-\sqrt{2} \hat{S}_{1z}$ | 1              | $-\sqrt{2} \hat{S}_{1z}$ |
| $|1, +1\rangle$             | $-\sqrt{2} \hat{S}_{1z}$ | 0              | $-\sqrt{2} \hat{S}_{1z}$ | 1 + 2$\hat{S}_{1z}$ |
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TABLE II. Matrix elements $\langle \psi_0 | \hat{S}^i_1 | \psi_0 \rangle$. Notation as in Table I.

| $\langle \psi_0 | \hat{S}^i_1 | \psi_0 \rangle$ | (0,0) | (1,−1) | (1,0) | (1,+1) |
|-----------------------------------|-------|-------|-------|-------|
| $\langle 0,0 \rangle$             | 0     | $-\hat{S}_{d,z}^z/\sqrt{2}$ | $-\hat{S}_{d,r}^z/\sqrt{2}$ | $\hat{S}_{d,s}^z/\sqrt{2}$ |
| $\langle 1,−1 \rangle$            | $-\hat{S}_{d,r}^z/\sqrt{2}$ | $-\hat{S}_{d,s}^z/\sqrt{2}$ | $0$ | $\hat{S}_{d,r}^z/\sqrt{2}$ |
| $\langle 1,0 \rangle$             | $-\hat{S}_{d,s}^z/\sqrt{2}$ | $\hat{S}_{d,r}^z/\sqrt{2}$ | 0 | $\hat{S}_{d,s}^z/\sqrt{2}$ |
| $\langle 1,+1 \rangle$            | $\hat{S}_{d,s}^z/\sqrt{2}$ | 0 | $-\hat{S}_{d,r}^z/\sqrt{2}$ | $+\hat{S}_{d,s}^z/\sqrt{2}$ |

\[
\hat{S}_{d}^{z}(\frac{1}{2}) \hat{c}_{d}^{z} = \hat{c}_{d}^{z} + \frac{3\hat{c}_{d}^{z}}{4} + \hat{c}_{d}^{z} \hat{S}_{d}^{z} \pm \frac{1}{2} \hat{S}_{d}^{z}.
\] (A6)

Useful relations involve the expectation values in the singlet state of observables at site 0

\[
\langle \hat{c}^\dagger_{0,a'}\hat{c}_{0,a} \rangle = \langle \hat{c}^\dagger_{0,a'}\hat{c}_{0,a} \rangle = \frac{1}{2} \delta_{a,a'}, \quad \langle \hat{n}_{0} \rangle = 1, \quad \langle \hat{n}_{0} \rangle = 0, \quad \langle \hat{n}_{0} \rangle = 0,
\] (A7)

\[
\langle \hat{S}_{0}\hat{n}_{0} \rangle = 0, \quad \langle \hat{S}_{0}\hat{n}_{0} \rangle = \frac{1}{2} \delta_{ij}, \quad \langle \hat{S}_{0}\hat{n}_{0} \rangle = \frac{1}{8} \epsilon_{ijk},
\] (A8)

where $\hat{S}_{0}^{z}$ is the $i$th cartesian component of $\hat{S}_{0}$ and $\epsilon_{ijk}$ is the third rank antisymmetric tensor.

We can also derive the following expressions for singlet expectation values of the form $\langle \hat{H}_{dd} \ldots \hat{H}_{dd} \rangle$,

\[
\langle \hat{H}_{dd}\hat{n}_{dd}\hat{H}_{dd} \rangle = |t_{0}|^{2} \hat{n}_{1}, \quad \langle \hat{H}_{dd}\hat{n}_{dd}\hat{H}_{dd} \rangle = |t_{0}|^{2}, \quad \langle \hat{H}_{dd}\hat{n}_{dd}\hat{H}_{dd} \rangle = 2|t_{0}|^{2} \hat{n}_{1}, \quad \langle \hat{H}_{dd}\hat{n}_{dd}\hat{H}_{dd} \rangle = |t_{0}|^{2}(1 + \hat{n}_{1} - 2\hat{n}_{1}\hat{n}_{1}),
\] (A11)

and

\[
\langle \hat{H}_{dd}\hat{S}_{d}^{2}\hat{H}_{dd} \rangle = |t_{0}|^{2} \left( \frac{3}{4} + \hat{S}_{d}^{2} - 2\hat{s}_{1} \cdot \hat{S}_{d} \right), \quad \langle \hat{H}_{dd}\hat{S}_{d}^{2}\hat{H}_{dd} \rangle = |t_{0}|^{2} \left( \hat{S}_{d}^{4} - 4\hat{s}_{1} \cdot \hat{S}_{d} \hat{S}_{d}^{2} - \hat{s}_{1} \cdot \hat{S}_{d} + \frac{5}{2} \hat{S}_{d}^{2} + \frac{9}{16} \right).
\] (A12)

Matrix elements of various observables within and between the singlet and triplet manifolds are listed in Tables I–III.

39 Since the exchange constant $J_s$ is typically below $\sim \bar{\delta}$, the condition $J_k \gg J_s$ is automatically satisfied in the strong-coupling limit.
44 A. Messiah, Quantum Mechanics (North-Holland, Amsterdam, 1961), Vol. II, Appendix C.