Gutzwiller variational approach to the two-impurity Anderson model at particle-hole symmetry

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Abstract. We study Gutzwiller-correlated wave functions as variational ground states for the two-impurity Anderson model (TIAM) at particle-hole symmetry as a function of the impurity separation $R$. Our variational state is obtained by applying the Gutzwiller many-particle correlator to a single-particle product state. We determine the optimal single-particle product state fully variationally from an effective non-interacting TIAM that contains a direct electron transfer between the impurities as variational degree of freedom. For a large Hubbard interaction $U$ between the electrons on the impurities, the impurity spins experience a Heisenberg coupling proportional to $V^2/U$ where $V$ parameterizes the strength of the on-site hybridization. For small Hubbard interactions we observe weakly coupled impurities. In general, for a three-dimensional simple cubic lattice we find discontinuous quantum phase transitions that separate weakly interacting impurities for small interactions from singlet pairs for large interactions.
1. Introduction

The description of impurities in a metallic host poses a fundamental problem in solid-state theory. The single-impurity Anderson model, the s-d (or ‘Kondo’) model, and other single-impurity Hamiltonians are among the most studied many-particle problems because they can be treated analytically and numerically with a variety of methods and some of them can even be solved exactly; for an overview, see Refs. 1 \( \text{[1]–[3], and references therein. Nowadays, the ‘Kondo effect’ is well understood: the host electrons build a ‘Kondo cloud’ around the impurity so that the impurity spin-1/2 is screened into a ‘Kondo spin’. At zero temperature, the host electrons and the impurity spin eventually form a ‘Kondo singlet’, and the ground state of the system is non-degenerate.}

When there are two (magnetic) impurities present in the system, they interact via the RKKY mechanism, named after Ruderman and Kittel [5], Kasuya [6], and Yosida [7]. The electrons scatter off both impurities and thereby mediate an effective interaction between the impurities. For large enough couplings, two impurity spins can bind into a singlet, and the ground state is also non-degenerate. Apparently, the RKKY and Kondo mechanisms for singlet formation compete with each other. Consequently, as pointed out by Jones, Varma, and Wilkins [8] and by Jones and Varma [9], a quantum phase transition between Kondo singlet and spin-pair phases might occur, depending on the ratio of Kondo and RKKY couplings in the two-impurity Kondo model. This proposition was supported by a slave-boson mean-field study [10].

Subsequent numerical [11] and variational studies [12, 13, 14] questioned the existence of a quantum phase transition in the two-impurity Kondo and Anderson models. It was later shown analytically [15, 16] that the appearance of a quantum phase transition in the two-impurity Kondo model at particle-hole symmetry depends on the impurities’ lattice positions. To complicate matters, for impurities in the same bath of host electrons, the two competing energy scales prevent a straightforward mapping of the two-impurity Anderson model to the two-impurity Kondo model employing the Schrieffer-Wolff transformation [17]. Therefore, it is not obvious that the two models belong to the same universality class, and it remains interesting to investigate the competition between Kondo and RKKY interactions for the two-impurity Anderson model.

In this work, we investigate the ground state of the particle-hole symmetric two-impurity Anderson model at half band-filling [18]. In the Gutzwiller variational ground state, the Gutzwiller many-particle correlator is applied to a single-particle product state [19] that can be viewed as the ground state of an effective single-particle Hamiltonian. Since only the impurity electrons are correlated, the wave function can be evaluated without further approximations. Therefore, we derive upper bounds to the exact ground-state energy.

In contrast to previous variational studies [12, 13, 14], our single-particle product state for the Gutzwiller wave function is determined fully variationally as the optimal ground state of an effective non-interacting two-impurity Anderson model. In a previous article [20], referred to as MBG, we studied the non-interacting Hamiltonian. The solution in MBG parametrically depends on the effective electron transfer between the two impurities. The variational freedom to generate an inter-impurity electron transfer is decisive for the Gutzwiller ground-state phase diagram for the interacting two-impurity Anderson model. We find a (generically discontinuous) phase transition as a function of the Hubbard interaction between two impurity spins at large interactions. The transition appears generically in a parameter range where the two-impurity Anderson model cannot be described faithfully by an effective spin model.

Our work is organized as follows. In Sect. 2, we define the two-impurity Anderson model and rewrite it in the form of a two-orbital model. In Sect. 3, we recall the conditions for particle-hole symmetry at half band-filling, we define the various parameter limits of interest (atomic, spin-model, Kondo, itinerant), and we discuss the two phases that we expect to find. Next, in Sect. 4, we introduce and evaluate the Gutzwiller variational ground state. In particular, we identify the effective non-interacting two-impurity model and recover the exact results for the atomic limit. In Sect. 5, we investigate host electrons with nearest-neighbor transfers on a simple-cubic lattice and provide explicit expressions for the single-particle contribution to the variational ground-state energy for small hybridizations. In Sect. 6, we study the spin-model and Kondo limits where the Hubbard interaction is large. In this limit, the optimization of the Gutzwiller variational parameters can be done analytically to a far extent. This provides useful insights into the quantum phase transition from weakly coupled impurities to singlet pairs. In Sect. 7, we discuss the numerical results for the quantum phase transitions in the whole parameter space. Short conclusions, Sect. 8, close our presentation. We defer technical details to six extensive appendices.

2. Two-impurity Anderson model

We start our investigation with the definition of the Hamiltonian. Then, we rephrase the problem in terms of a two-orbital model.
2.1. Hamiltonian

Two impurities in a metallic host on a lattice are modeled by the Hamiltonian [18]
\[ H = \hat{T} + \hat{T}_d + \hat{V} + \hat{H}_{\text{int}} \equiv \hat{H}_0 + \hat{H}_{\text{int}}. \]  
(1)

Here, \( \hat{T} \) is the kinetic energy of the non-interacting spin-1/2 host electrons (\( \sigma = \uparrow, \downarrow \)),
\[ \hat{T} = \sum_{\mathbf{R}, \mathbf{R}', \sigma} t(\mathbf{R} - \mathbf{R}') \hat{c}_{\mathbf{R}, \sigma}^+ \hat{c}_{\mathbf{R}', \sigma}, \]  
(2)

where the electrons tunnel between the sites \( \mathbf{R} \) and \( \mathbf{R}' \) of the lattice with amplitude \( t(\mathbf{R} - \mathbf{R}') \). The kinetic energy is diagonal in Fourier space. For \( \mathbf{k} \) from the first Brillouin zone we define
\[ \hat{c}_{\mathbf{k}, \sigma} = \frac{1}{\sqrt{L}} \sum_{\mathbf{R}} e^{-i\mathbf{k} \cdot \mathbf{R}} \hat{c}_{\mathbf{R}, \sigma}, \]
\[ \hat{c}_{\mathbf{R}, \sigma} = \frac{1}{\sqrt{L}} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{R}} \hat{c}_{\mathbf{k}, \sigma}, \]  
(3)

where \( L \) is the (even) number of lattice sites. With
\[ t(\mathbf{R}) = \frac{1}{L} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{R}} \epsilon(\mathbf{k}), \]
\[ \epsilon(\mathbf{k}) = \sum_{\mathbf{R}} t(\mathbf{R}) e^{-i\mathbf{k} \cdot \mathbf{R}}, \]  
(4)

the host electron kinetic energy becomes diagonal,
\[ \hat{T} = \sum_{\mathbf{k}, \sigma} \epsilon(\mathbf{k}) \hat{c}_{\mathbf{k}, \sigma}^+ \hat{c}_{\mathbf{k}, \sigma}, \]  
(5)

where \( \epsilon(\mathbf{k}) \) is the dispersion relation.

With \( \hat{T}_d \) we also permit a direct electron transfer with amplitude \( t_{12} \) between the impurity orbitals at sites \( \mathbf{R}_1 \) and \( \mathbf{R}_2 \),
\[ \hat{T}_d = \sum_{\sigma} t_{12} \hat{d}_{1, \sigma}^+ \hat{d}_{2, \sigma} + t_{12}^* \hat{d}_{2, \sigma}^+ \hat{d}_{1, \sigma}. \]  
(6)

For most of the paper, however, we shall focus on the case of vanishingly small direct electron transfer between the impurities, \( |t_{12}| \to 0 \).

Next, \( \hat{V} \) describes the hybridization between impurity and host electron states (\( b = 1, 2 \)),
\[ \hat{V} = \sum_{\mathbf{R}, \mathbf{k}, \sigma} V(\mathbf{R} - \mathbf{R}_b) \hat{c}_{\mathbf{R}, \sigma}^+ \hat{d}_{b, \sigma} + V^*(\mathbf{R} - \mathbf{R}_b) \hat{d}_{b, \sigma}^+ \hat{c}_{\mathbf{R}, \sigma}, \]
\[ = \frac{1}{\sqrt{L}} \sum_{\mathbf{k}, \mathbf{k}', \sigma} V_\mathbf{k} e^{-i\mathbf{k} \cdot \mathbf{R}} \hat{c}_{\mathbf{k}, \sigma}^+ \hat{d}_{b, \sigma} + V_\mathbf{k}^* e^{i\mathbf{k} \cdot \mathbf{R}} \hat{d}_{b, \sigma}^+ \hat{c}_{\mathbf{k}, \sigma}, \]
\[ V_\mathbf{k} = \sum_{\mathbf{R}} V(\mathbf{R}) e^{-i\mathbf{k} \cdot \mathbf{R}}, \quad V(\mathbf{R}) = \frac{1}{L} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{R}} V_\mathbf{k}. \]  
(7)

In Sects. [5, 7] we shall employ a local hybridization, \( V_\mathbf{k} \equiv V \). The non-interacting two-impurity Anderson model \( \hat{H}_0 = \hat{T} + \hat{T}_d + \hat{V} \) can be solved exactly using the equation-of-motion method [18], see MBG.

Last, \( \hat{H}_{\text{int}} \) represents the Hubbard interaction to model the Coulomb repulsion on the impurities,
\[ \hat{H}_{\text{int}} = U \sum_{b=1}^2 (\hat{n}_{b, \uparrow}^d - 1/2)(\hat{n}_{b, \downarrow}^d - 1/2), \]  
(8)

where \( \hat{n}_{b, \sigma}^d = \hat{d}_{b, \sigma}^+ \hat{d}_{b, \sigma} \) counts the number of impurity electrons. The two-impurity Anderson model \( \hat{H} = \hat{H}_0 + \hat{H}_{\text{int}} \) in eq. (1) poses a difficult many-particle problem that cannot be solved in general.

2.2. Single-site two-orbital model

As a second step, we map the two-impurity model onto an asymmetric two-orbital model. This step is equivalent to the introduction of even and odd parity channels.

2.2.1. Kinetic energy of d-electrons We introduce the new ‘h-basis’ for the impurity electrons using the unitary transformation
\[ \hat{d}_{1, \sigma}^+ = \frac{1}{\sqrt{2}} \left( \hat{h}_{1, \sigma}^+ + \alpha_{12} \hat{h}_{2, \sigma}^+ \right), \]
\[ \hat{d}_{2, \sigma}^+ = \frac{1}{\sqrt{2}} \left( -\alpha_{12} \hat{h}_{1, \sigma}^+ + \hat{h}_{2, \sigma}^+ \right), \]  
(9)

where
\[ \alpha_{12} = \frac{t_{12}^*}{|t_{12}|}, \quad \alpha_{21} = \frac{t_{12}}{|t_{12}|}. \]

(10)

The inverse transformation reads
\[ \hat{h}_{1, \sigma}^+ = \frac{1}{\sqrt{2}} \left( \hat{d}_{1, \sigma}^+ - \alpha_{12} \hat{d}_{2, \sigma}^+ \right), \]
\[ \hat{h}_{2, \sigma}^+ = \frac{1}{\sqrt{2}} \left( \alpha_{12} \hat{d}_{1, \sigma}^+ + \hat{d}_{2, \sigma}^+ \right). \]  
(11)

For a unitary transformation we have (\( \hat{n}_{b, \sigma} = \hat{h}_{b, \sigma}^+ \hat{h}_{b, \sigma} \))
\[ \hat{d}_{1, \sigma}^+ \hat{d}_{1, \sigma} + \hat{d}_{2, \sigma}^+ \hat{d}_{2, \sigma} = \hat{n}_{1, \sigma} + \hat{n}_{2, \sigma}, \]  
(12)

and the average number of \( d \)-electrons obviously equals the average number of \( h \)-electrons.

We introduced the h-basis because it diagonalizes \( \hat{T}_d \), eq. (6),
\[ \tilde{T}_d = |t_{12}| \sum_{\sigma} \left( \hat{h}_{2, \sigma}^+ \hat{h}_{2, \sigma} - \hat{h}_{1, \sigma}^+ \hat{h}_{1, \sigma} \right). \]  
(13)

In the h-basis representation, \( \tilde{T}_d \) has the form of a splitting of the two impurity levels. For \( |t_{12}| \to 0 \), the occupancies of both orbitals should be the same. A broken h-orbital symmetry in the ground state \( |\Psi_0\rangle \), \( \langle \Psi_0 | \hat{n}_{1, \sigma} | \Psi_0 \rangle \neq \langle \Psi_0 | \hat{n}_{2, \sigma} | \Psi_0 \rangle \), indicates that a finite electron transfer between the impurities is enhanced by the interplay of the electrons’ kinetic energy and their Coulomb interactions, \( \langle \Psi_0 | \hat{d}_{1, \sigma}^+ \hat{d}_{2, \sigma}^+ | \Psi_0 \rangle \neq 0 \).
2.2.2. Hybridization  In the h-basis, the hybridization \( V \), see eq. (7), takes the form
\[
\hat{V} = \frac{1}{\sqrt{L}} \sum_{k,\sigma} V_{k,\sigma} \hat{c}_{k,\sigma}^+ \hat{h}_{b,\sigma} + V_{k,\sigma} \hat{c}_{k,\sigma} \hat{h}_{b,\sigma}^+.
\]  (14)

The two impurity levels hybridize with the host electrons with the matrix elements
\[
V_{k,1} = V_{k,1}(\mathbf{R}_1, \mathbf{R}_2) = \frac{V_k}{\sqrt{2}} (e^{-i \mathbf{k} \cdot \mathbf{R}_1} - \alpha_{12} e^{-i \mathbf{k} \cdot \mathbf{R}_2}),
\]
\[
V_{k,2} = V_{k,2}(\mathbf{R}_1, \mathbf{R}_2) = \frac{V_k}{\sqrt{2}} (\alpha_{12}^* e^{-i \mathbf{k} \cdot \mathbf{R}_1} + e^{-i \mathbf{k} \cdot \mathbf{R}_2}).
\]  (15)

In the absence of a direct electron transfer between the impurities, \( t_{12} = 0 \), we may set \( \alpha_{12} = 1 \) for all \( \mathbf{R}_1, \mathbf{R}_2 \). Then, \( V_{k,1}(\mathbf{R}_2) \) describes the hybridization in the odd (even) parity channel, \( V_{k,1}(\mathbf{R}_2, \mathbf{R}_1) = -V_{k,1}(\mathbf{R}_1, \mathbf{R}_2) \) \( [V_{k,2}(\mathbf{R}_2, \mathbf{R}_1) = V_{k,2}(\mathbf{R}_1, \mathbf{R}_2)] \). For our study, we keep \( |t_{12}| \) infinitesimally small so that \( \alpha_{12} \) remains well defined by eq. (10).

2.2.3. Interaction  We write the interaction term in its eigenbasis \( |\Gamma\rangle \),
\[
\hat{H}_{\text{int}} = \sum_{\Gamma} E_\Gamma \hat{n}_\Gamma \otimes |\Gamma\rangle \langle \Gamma|,
\]  (16)
where \( \Gamma = 1, \ldots, 16 \) labels the 16 possible configurations on the two impurity sites. They are listed in the local h-basis in table 1 together with the atomic spectrum. The operator \( \hat{T}_d \) mixes the states \( |10\rangle \) and \( |11\rangle \). All other states \( |\Gamma\rangle \) in table 1 are also eigenstates of \( \hat{T}_d \) with energy \( E_d = 0, \pm |t_{12}| \).

3. Particle-hole symmetry at half band-filling

We are interested in the case where there is on average one electron on each of the impurities. This can be assured for a particle-hole-symmetric Hamiltonian [1] at half band-filling.

3.1. Conditions

We consider a bipartite lattice and assume that there exists half a reciprocal lattice vector \( \mathbf{Q} = \mathbf{G}/2 \) for which
\[
\epsilon(\mathbf{k} \pm \mathbf{Q}) = -\epsilon(\mathbf{k}), \quad e^{i \mathbf{Q} \cdot \mathbf{R}} = \begin{cases} 1 & \text{if } \mathbf{R} \in A\text{-lattice} \\ -1 & \text{if } \mathbf{R} \in B\text{-lattice} \end{cases}.
\]  (17)

We also assume inversion symmetry, \( \epsilon(-\mathbf{k}) = \epsilon(\mathbf{k}) \); recall that \( \epsilon(\mathbf{k} + \mathbf{G}) = \epsilon(\mathbf{k}) \). Note that the transfer matrix elements between sites on different sublattices are real and those between sites on the same sublattice are purely imaginary, see MBG. The same applies to the impurity transfer matrix element \( t_{12} \). In the main text we focus on the case that the two impurities are on different sublattices and consider the other case in the appendix.

Moreover, we demand that
\[
V_k = V_k^* \cdot \mathbf{R}_k.
\]  (18)

Note that a \( \mathbf{k} \)-independent hybridization, \( V_k \equiv V \), must necessarily be real. The conditions (17) and (18) make the Hamiltonian invariant under particle-hole transformation, \( \hat{H} = \hat{\phi}_p \hat{H} \hat{\phi}_p^{-1} \), see MBG.

3.2. Half-filled bands

In the following we consider paramagnetism at half band-filling where the number of electrons \( N = N_+ + N_- \) equals the (even) number of orbitals, \( N = L + 2 \), and \( N_+ = N_- = L/2 + 1 \). Note that there are \( L \) lattice sites for the host electrons and two additional impurity orbitals on the lattice sites \( \mathbf{R}_1 \) and \( \mathbf{R}_2 \).

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Table 1. Atomic eigenstates \( |\Gamma\rangle \) of \( \hat{H}_{\text{int}} \) with energy \( E_\Gamma \); all states apart from \( |10\rangle \) and \( |11\rangle \) are also eigenstates of \( \hat{T}_d \) with energy \( E_d = \pm |t_{12}| \).

| \( n \) | \( |\Gamma\rangle \) | \( E_\Gamma \) | \( E_d \) |
|---|---|---|---|
| 0 | \{0, 0\} \equiv \{\text{vac}\} | U/2 | 0 |
| 1 \* | \{\uparrow, \uparrow\} \equiv \{\text{up-up}\} | -|t_{12}| | 0 |
| 2 \* | \{\uparrow, \downarrow\} \equiv \{\text{up-down}\} | -|t_{12}| | 0 |
| 3 \* | \{\downarrow, \uparrow\} \equiv \{\text{down-up}\} | -|t_{12}| | 0 |
| 4 \* | \{\downarrow, \downarrow\} \equiv \{\text{down-down}\} | -|t_{12}| | 0 |
| 5 \* | \{\uparrow, \uparrow\} \equiv \{(1, 1)\} | 0 | t_{12} |
| 6 \* | \{\uparrow, \downarrow\} \equiv \{1, 0\} | 0 | t_{12} |
| 7 \* | \{\downarrow, \uparrow\} \equiv \{|0, 1\} | 0 | t_{12} |
| 8 \* | \{\downarrow, \downarrow\} \equiv \{|0, 0\} | 0 | t_{12} |
| 9 \* | \{\uparrow, \uparrow\} \equiv \{(1, 1)\} | -2U/2 | 0 |
| 10 \* | \{\uparrow, \downarrow\} \equiv \{1, 0\} | -2U/2 | 0 |
| 11 \* | \{\downarrow, \uparrow\} \equiv \{|0, 1\} | -2U/2 | 0 |
| 12 \* | \{\downarrow, \downarrow\} \equiv \{|0, 0\} | -2U/2 | 0 |
| 13 \* | \{\uparrow, \uparrow\} \equiv \{(1, 1)\} | 0 | t_{12} |
| 14 \* | \{\uparrow, \downarrow\} \equiv \{1, 0\} | 0 | t_{12} |
| 15 \* | \{\downarrow, \uparrow\} \equiv \{|0, 1\} | 0 | t_{12} |
| 16 \* | \{\downarrow, \downarrow\} \equiv \{|0, 0\} | 0 | t_{12} |

\( k \cdot q \)-basis in table 1, together with the atomic spectrum. The operator \( \hat{T}_d \) mixes the states \( |10\rangle \) and \( |11\rangle \). All other states \( |\Gamma\rangle \) in table 1 are also eigenstates of \( \hat{T}_d \) with energy \( E_d = 0, \pm |t_{12}| \).
At half band-filling, the non-degenerate ground state $|\Psi_0\rangle$ maps onto itself under the particle-hole transformation $\tau_{ph}$. Therefore, we find that each impurity level is exactly half filled for all hybridizations and interaction strengths,

$$\langle \Psi_0|\hat{d}_{1,\sigma}^\dagger \hat{d}_{1,\sigma}|\Psi_0\rangle = \langle \Psi_0|\hat{d}_{2,\sigma}^\dagger \hat{d}_{2,\sigma}|\Psi_0\rangle = 1/2.$$  \hspace{1cm} \text{(19)}

Moreover, it is readily shown that the bare density of states is symmetric,

$$D_{\sigma,0}(\epsilon) = \frac{1}{T} \sum_k \delta(\epsilon - \epsilon(k)) = D_{\sigma,0}(-\epsilon),$$ \hspace{1cm} \text{(20)}

so that the Fermi energy is $E_F = 0$ at half band-filling.

At half band-filling, eq. \text{(12)} implies

$$\langle \Psi_0|\hat{n}_{1,\sigma} + \hat{n}_{2,\sigma}|\Psi_0\rangle = 1.$$ \hspace{1cm} \text{(21)}

Moreover, as shown in MBG, particle-hole symmetry demands that there is no hybridization between the $h$-orbitals at half band-filling,

$$\langle \Psi_0|\hat{n}_{1,\sigma}^\dagger \hat{n}_{2,\sigma}|\Psi_0\rangle = 0.$$ \hspace{1cm} \text{(22)}

This relation considerably simplifies the evaluation of Gutzwiller-correlated wave functions.

### 3.3. Limiting cases

Before we proceed, we define some parameter limits of interest. We compare the Hubbard parameter $U$ with the bandwidth of the host electrons $W$ and the hybridization $V$. The hybridization is always assumed to be small compared to the bandwidth, $V \ll W$.

#### 3.3.1. Atomic limit

The atomic limit is defined by $V = 0$ so that the $d$-levels are singly occupied for all $U > 0$. In the presence of a direct electron transfer between the impurities, $t_{12} \neq 0$, the ground state of the corresponding two-site Hubbard model is a spin singlet. For $U \gg |t_{12}|$, the ground-state energy attains the familiar Heisenberg form,

$$E_{\text{Heis}} = -\frac{4|t_{12}|^2}{U} + O(1/U^2).$$ \hspace{1cm} \text{(23)}

If for $V \neq 0$ and large interactions the variational ground-state energy has a contribution proportional to $1/U$, eq. \text{(23)} indicates that the impurities are coupled by an effective electron transfer, see Sect.\text{\textsection}.

In the remainder of this section, we focus on the case of a vanishingly small direct coupling, $|t_{12}| \rightarrow 0$.

#### 3.3.2. Spin-model, Kondo, and itinerant limits

When $U$ is the largest energy scale, $V \ll W \ll U$, the single-impurity Anderson model maps onto the $s$-$d$ (or Kondo) model \cite{1}

$$H_{\text{SIAM}} \Rightarrow H_K,$$ \hspace{1cm} \text{(24)}

$$H_{\text{SIAM}} = \hat{T} + \frac{V}{\sqrt{L}} \sum_{k,\sigma} \left( \hat{c}_{k,\sigma}^\dagger \hat{d}_{\sigma} + \hat{d}_{\sigma}^\dagger \hat{c}_{k,\sigma} \right)$$

$$+ U \left( \hat{n}_{1}^d - 1/2 \right) \left( \hat{n}_{2}^d - 1/2 \right),$$

$$H_K = \hat{T} + J_K \hat{S} \cdot \hat{S},$$ \hspace{1cm} \text{(25)}

where $\hat{S}$ is the impurity-spin operator and $\hat{s}$ denotes the host-electron spin at the impurity position at the origin. Here, the antiferromagnetic Kondo coupling is given by \text{[1]}

$$J_K = \frac{4V^2}{U}.$$ \hspace{1cm} \text{(26)}

A widely used generalization of \text{(25)} is the two-impurity Kondo model (TIKM),

$$\hat{H}_{\text{TIKM}} = \hat{T} + J_K \sum_\sigma \hat{s}_\sigma \cdot \hat{S}_\sigma + J_H \hat{S}_1 \cdot \hat{S}_2,$$ \hspace{1cm} \text{(27)}

see, e.g., Refs \text{[8, 9]}, it is not clear to us whether or not the TIKM can be derived from the TIAM rigorously. Therefore, the value of $J_H$ as a function of the TIAM parameters is not known. Consequently, $J_H$ is often taken as an independent model parameter \text{[8, 9] 10 11].

In the single-impurity Anderson model, the ‘spin-model limit’ $U \gg W$ is not a prerequisite to find effectively a spin on the impurities, i.e., to have the impurity levels almost exactly singly occupied. As is well known for the SIAM \text{[1]}, the relevant energy scale actually is

$$\Gamma = \pi d_0 V^2,$$ \hspace{1cm} \text{(28)}

where $d_0 = D_{\sigma,0}(0) \sim 1/W$ is the host-electron density of states at the Fermi energy. Even for $U \ll W$, the ‘Kondo limit’ $\Gamma \ll U \ll W$ guarantees that in the ground state there is basically a localized spin at the impurity site.

Lastly, for $U \lesssim \Gamma \ll W$, the system resembles the features of the non-interacting Anderson model where the occupation of the impurities is not integer. In this ‘itinerant limit’, the impurities experiences an effective RKKY interaction \text{[20].}

$$J_{\text{RKKY}}(R) = -\frac{\Gamma}{\pi} \left( \frac{d_R}{d_0} \right)^2,$$ \hspace{1cm} \text{(29)}

where $d_R$ can be expressed as an integral over the Fermi surface, see MBG and Sect.\text{\textsection}. The RKKY coupling strength vanishes as a function of the impurity separation $R = R_1 - R_2$.

The key advantage of our variational approach lies in the fact that we can study the TIAM on equal footing in the whole $(V, U, W)$ parameter space. In particular, we can treat the spin and Kondo limits analytically to a far extent, see Sect.\text{\textsection}.

#### 3.3.3. Singlet pairs versus weakly linked impurities

As pointed out by Jones and Varma \text{[8, 9]}, for the two-impurity Kondo model $\hat{H}_{\text{TIKM}}$ in \text{(27)}, there are two competing mechanisms for singlet formation in the ground state. For weakly linked impurities where $J_H \ll J_K$, the Kondo coupling between the impurities and the host electrons leads to individual singlets made from an impurity spin and its surrounding host electrons.
For strongly coupled impurity spins, \( J_H \gg J_K \) in the \( H_{\text{TICKM}} \), the two impurity spins form a (Heisenberg) spin singlet. Correspondingly, Varma and Jones found a quantum phase transition between these two phases at \( J_H \approx J_K \).

As we shall show in this work, such a transition generically also exists in the two-impurity Anderson model. Our analytical evaluation in the Kondo limit reveals a specific dependence of the variational ground-state energy on \( U/\Gamma \) in both phases,

\[
E_{\text{var}} \sim -\exp\left(-\frac{U\pi}{16\Gamma}\right) \quad \text{weakly coupled impurities} \quad U \ll U_c, \tag{30}
\]

\[
E_{\text{var}} \sim -\frac{\Gamma}{U} \quad \text{singlet pair} \quad U \gg U_c. \tag{31}
\]

The numerical minimization of our energy functional shows that the critical value for the transition is in the region \( \langle U/\Gamma \rangle_c \approx 12 \ldots 16 \) for all \( 2 \cdot 10^{-3}W \leq V \leq 2 \cdot 10^{-1}W \). This implies, however, that the transition cannot be found in the spin-model limit \( U \gg W \) because it implies \( V \gg W/\sqrt{U/\Gamma}\pi d_0 \) which contradicts our basic assumption \( V \ll W \).

Therefore it seems that our findings are in mild conflict with the work of Varma and Jones. Note, however, that \( J_H \) in \( H_{\text{TICKM}} \) is treated as an adjustable parameter so that the competition of the Kondo and Heisenberg singlet formation can be studied in the two-impurity Kondo model, independent of the existence and the form of a mapping of the two-impurity Anderson model to the two-impurity Kondo model.

### 4. Gutzwiller variational approach

For the two-impurity Anderson model \cite{1} we propose a Gutzwiller-correlated wave function as variational ground state that we evaluate without approximations. Therefore, the variational energies obtained in this work provide upper bounds to the exact ground-state energy. For comparison, we include results of the Gutzwiller variational approach for the symmetric single-impurity Anderson model in Appendix D.

#### 4.1. Variational state

In the Gutzwiller approach, we assume that the exact ground state can be approximated by a normalized single-particle product state \( |\varphi_0\rangle \) into which the so-called Gutzwiller correlator \( \hat{P}_G \) introduces many-particle correlations,

\[
|\Psi_G\rangle = \hat{P}_G |\varphi_0\rangle. \tag{32}
\]

For the non-interacting case, we recover the exact result by choosing \( |\varphi_0\rangle \equiv |\Phi_0\rangle \), and \( \hat{P}_G(U = 0) = 1 \). In contrast to other variational approaches to the two-impurity Anderson model \cite{2,3,4}, we determine \( |\varphi_0\rangle \) fully variationally.

For our two-orbital situation, we employ the most general Hermitian correlator

\[
\hat{P}_G = \sum_\Gamma \lambda_\Gamma \hat{m}_\Gamma + \lambda_m |10\rangle\langle 11| + \lambda_m^* |11\rangle\langle 10| \tag{33}
\]

that can be applied to the two-impurity subsystem and does not violate the symmetries. Here, \( \lambda_\Gamma \) are real variational parameters that control the occupation probabilities of the atomic configuration \( |\Gamma\rangle \) in the single-particle product state \( |\varphi_0\rangle \). Particle number conservation and spin/particle-hole symmetry permit only the states \( |10\rangle \) and \( |11\rangle \) to be coupled in the correlator, with the help of a complex parameter \( \lambda_m \).

#### 4.2. Particle-hole symmetry

We demand that our variational state is invariant under particle-hole symmetry at half band-filling, i.e.,

\[
\hat{\tau}_\text{ph} \hat{P}_G \hat{\tau}_\text{ph} = |\Psi_G\rangle. \tag{34}
\]

For \( \Gamma = 1, \ldots, 5 \), the projectors \( \hat{m}_\Gamma \) obey \( \hat{\tau}_\text{ph}^+ \hat{m}_\Gamma \hat{\tau}_\text{ph} = \hat{m}_{10-\Gamma} \), and \( \hat{\tau}_\text{ph}^+ \hat{m}_\Gamma \hat{\tau}_\text{ph} = \hat{m}_{\Gamma} \). For \( \Gamma = 7, 9, 10, 11 \), the projectors \( \hat{m}_\Gamma \) are invariant under the particle-hole transformation \( \tau_{\text{ph}} \), and \( |10\rangle |11\rangle \) is equally invariant. Therefore, to ensure particle-hole symmetry, we must set \( \lambda_6 = \lambda_1 = \lambda_2 = \lambda_3 = \lambda_4 = \lambda_5 = \lambda_8 = \lambda_9 \). Moreover, due to symmetry under spin-flip \( \tau_{\uparrow \rightarrow \downarrow} \), we set \( \lambda_8 = \lambda_2 \), \( \lambda_4 = \lambda_5 \). We are left with eight variational parameters for the spin and particle-hole symmetric case whereby \( \lambda_1, \lambda_2, \lambda_3, \lambda_4, \lambda_5, \lambda_6, \lambda_7, \lambda_9, \lambda_{10} \), and \( \lambda_{11} \) are real and \( \lambda_m \) is complex. We subsume them in the real vector \( \lambda \),

\[
\lambda = (\lambda_1, \lambda_2, \lambda_3, \lambda_4, \lambda_5, \lambda_6, \lambda_7, \lambda_8, \lambda_9, \lambda_{10}, \lambda_{11}, x_m, y_m) \tag{35}
\]

with \( x_m = \text{Re}[\lambda_m] \), \( y_m = \text{Im}[\lambda_m] \). At half band-filling, the atomic states \( |1\rangle \), \( |9\rangle \), and \( |16\rangle \) belong to a charge-SU(2) triplet \cite{21}. Therefore, we can directly set \( \lambda_9 = \lambda_1 \). We did not implement this symmetry but verified it numerically to a high numerical accuracy so that the charge-SU(2) symmetry is indeed preserved.

#### 4.3. Constraints

To facilitate the evaluation of Gutzwiller-correlated wave functions, it is helpful to impose the constraints,

\[
\langle \varphi_0 | \hat{P}_G^+ \hat{P}_G \hat{h}^+_b \hat{h}_b | \varphi_0 \rangle = \delta_{\sigma,\sigma'} \langle \phi_0 | \hat{h}^+_b \hat{h}_b | \varphi_0 \rangle \tag{37}
\]

\[
\langle \varphi_0 | \hat{P}_G^+ \hat{P}_G \hat{h}^+_b \hat{h}_{b,\sigma} | \varphi_0 \rangle = \delta_{\sigma,\sigma'} \langle \varphi_0 | \hat{h}^+_b \hat{h}_{b,\sigma} | \varphi_0 \rangle. \tag{37}
\]
These constraints do not restrict the variational freedom. They simply insure that there are no Hartree bubbles in a diagrammatic evaluation of Gutzwiller-correlated wave functions [22, 23, 24].

Due to particle-hole symmetry, Eq. (22) also holds for the Gutzwiller variational state, i.e.,
\[
\langle \varphi_0| \hat{P}_G^{\dagger} \hat{P}_G \hat{h}_{b,\sigma} \hat{h}_{\bar{b},\sigma} | \varphi_0 \rangle = \langle \varphi_0| \hat{h}_{b,\sigma} \hat{h}_{\bar{b},\sigma} | \varphi_0 \rangle = 0
\]
with the notation \( I \equiv 2, \; 2 \equiv 1 \). The explicit conditions on \( \lambda \) are derived in Appendix A.

4.4. Calculation of expectation values

4.4.1. Host electrons For the host electrons we need to evaluate
\[
\langle \hat{c}_{k,\sigma} \dagger \hat{c}_{k,\sigma} \rangle_G = \frac{\langle \Psi_G | \hat{c}_{k,\sigma} \dagger \hat{c}_{k,\sigma} | \Psi_G \rangle}{\langle \Psi_G | \Psi_G \rangle},
\]
(39)

By construction, the denominator is unity because we normalized the Gutzwiller wave function, see eq. 36. The numerator can be cast into the form
\[
\langle \Psi_G | \hat{c}_{k,\sigma} \dagger \hat{c}_{k,\sigma} | \Psi_G \rangle = \langle \varphi_0| \hat{c}_{k,\sigma} \dagger \hat{c}_{k,\sigma} \hat{P}_G^{\dagger} \hat{P}_G | \varphi_0 \rangle,
\]
which can be written with the help of Wick’s theorem. All diagrams in eq. 40 with lines between \( k \) and the impurity system vanish because of the constraint 39 and the fact that the constraint is fulfilled due to symmetry. Consequently,
\[
\langle \Psi_G | \hat{c}_{k,\sigma} \dagger \hat{c}_{k,\sigma} | \Psi_G \rangle = \langle \varphi_0| \hat{c}_{k,\sigma} \dagger \hat{c}_{k,\sigma} | \varphi_0 \rangle.
\]
(41)

This relation is very useful because the correlations are seen to change only the impurity expectation values but not the host-electron energy,
\[
E_{\text{host}} = \sum_{k,\sigma} \epsilon(k) \langle \Psi G | \hat{c}_{k,\sigma} \dagger \hat{c}_{k,\sigma} | \Psi G \rangle = \sum_{k,\sigma} \epsilon(k) \langle \varphi_0| \hat{c}_{k,\sigma} \dagger \hat{c}_{k,\sigma} | \varphi_0 \rangle,
\]
(42)
as for a non-interacting symmetric two-impurity Anderson model with ground state \( |\varphi_0\rangle \).

4.4.2. Orbital occupancies Due to spin-flip symmetry, the orbital occupancies do not depend on the spin direction. Moreover, particle-hole symmetry leads to
\[
\langle \Psi G | \hat{h}_{\uparrow}\dagger \hat{h}_{\downarrow} | \Psi G \rangle = 1 - \langle \Psi G | \hat{h}_{\uparrow}\dagger \hat{h}_{\downarrow} | \Psi G \rangle,
\]
(43)

cf. eq. 21. We are left with the task to calculate
\[
\langle \Psi G | \hat{h}_{\uparrow}\dagger \hat{h}_{\downarrow} | \Psi G \rangle = \langle \varphi_0| \hat{P}_G^{\dagger} \hat{n}_{\uparrow}\dagger \hat{P}_G | \varphi_0 \rangle.
\]
(44)
The matrix element is evaluated in Appendix B. As a function of \( \lambda \) and \( n_{\uparrow}, \downarrow \), it becomes
\[
\langle \hat{n}_{\uparrow}\dagger \rangle_G = \left( \frac{\lambda_{10} + \lambda_{11} + 2x_m}{4} \right)^2 (n_{\uparrow,\downarrow})^4 + \frac{3\lambda_2^2(n_{\uparrow,\downarrow})^2}{2} n_{\uparrow,\downarrow}^2 + \frac{2}{2} (\lambda_{11} - \lambda_{10} + 2y_m)^2 (n_{\uparrow,\downarrow})^4 + \frac{2}{4} \lambda_4^2(n_{\uparrow,\downarrow})^3 + \frac{2}{4} \lambda_4^2(n_{\uparrow,\downarrow})^4 + \frac{2}{4} \lambda_4^2(n_{\uparrow,\downarrow})^5
\]
where \( n_{\uparrow} = \langle \varphi_0| \hat{h}_{\uparrow}\dagger \hat{h}_{\uparrow} | \varphi_0 \rangle \) and \( n_{\downarrow} = 1 - n_{\uparrow} \).

4.4.3. Hybridization Due to spin-flip symmetry, the hybridization matrix elements do not depend on the spin direction. Moreover, particle-hole symmetry leads to
\[
\langle \Psi G | \hat{c}_{k,\uparrow} \dagger \hat{h}_{\downarrow} | \Psi G \rangle = \langle \Psi G | \hat{c}_{\bar{k},\uparrow} \dagger \hat{h}_{\downarrow} | \Psi G \rangle.
\]
(46)

Therefore, we are left with the task to calculate
\[
\langle \Psi G | \hat{c}_{k,\uparrow} \dagger \hat{h}_{\downarrow} | \Psi G \rangle = \langle \varphi_0| \hat{c}_{k,\uparrow} \dagger \hat{P}_G^{\dagger} \hat{h}_{\downarrow} | \varphi_0 \rangle.
\]
(47)

This matrix element is evaluated in Appendix B with the result
\[
\langle \Psi G | \hat{c}_{k,\uparrow} \dagger \hat{h}_{\downarrow} | \Psi G \rangle = q \langle \varphi_0| \hat{c}_{k,\uparrow} \dagger \hat{h}_{\downarrow} | \varphi_0 \rangle.
\]
(48)

As a function of the Gutzwiller variational parameters \( \lambda \) and of \( n_{\uparrow} \), we find
\[
q(\lambda, n_{\uparrow}) = \frac{\lambda_2(\lambda_{10} + \lambda_{11} + 2x_m)(n_{\uparrow})^3 - 2}{2} + \frac{\lambda_2(2\lambda_{10} + 3\lambda_6 + \lambda_9)(n_{\uparrow})^2}{2} n_{\downarrow} + \frac{\lambda_4(2\lambda_{10} + 3\lambda_6 + \lambda_9)(n_{\uparrow})}{2} n_{\downarrow}^2 + \frac{\lambda_4(\lambda_{10} + \lambda_{11} - 2x_m)(n_{\downarrow})}{2} (n_{\uparrow})^3,
\]
(49)

where \( n_{\downarrow} = 1 - n_{\uparrow} \).

4.4.4. Interaction For the interaction on the impurity we need to evaluate
\[
\langle \hat{H}_{\text{int}} | G \rangle = \sum_{\Gamma} E_{\Gamma} \langle \varphi_0| \hat{P}_G^{\dagger} \hat{n}_{\uparrow}\dagger \hat{P}_G | \varphi_0 \rangle \equiv E_{\text{int}}.
\]
(50)
The matrix element is evaluated in Appendix B. As a function of \( \lambda \) and \( n_{\uparrow} \), it becomes
\[
\frac{2E_{\text{int}}}{U} = \left( \frac{2\lambda_{11}^2 - 3\lambda_{11}^2 + \lambda_8^2}{2} \right)(n_{\uparrow})^2 + \left( \frac{2\lambda_{11}^2 - 3\lambda_{11}^2 + \lambda_8^2}{2} \right) (n_{\uparrow})^4 + \left( \frac{2\lambda_{11}^2 - 3\lambda_{11}^2 + \lambda_8^2}{2} \right) (n_{\uparrow})^6 + x_m(\lambda_{10} - \lambda_{11})((n_{\uparrow})^4 - (n_{\downarrow})^4).
\]
(51)

4.5. Optimization of the single-particle state

To determine the variational parameters, we must minimize the variational ground-state energy,
\[
E_{\text{var}}(\lambda, |\varphi_0\rangle) = \langle \Psi_G | \hat{H} | \Psi_G \rangle
\]
\[
= \sum_{k,\sigma} \epsilon(k) \langle \varphi_0| \hat{c}_{k,\sigma} \dagger \hat{c}_{k,\sigma} | \varphi_0 \rangle + E_{\text{int}} + 2|t_{12}| (1 - 2) n_{\uparrow}\dagger G
\]
\[
+ \sum_{k,\bar{k},\sigma} [q(V_{k,\bar{k}} \langle \varphi_0| \hat{c}_{k,\sigma} \dagger \hat{h}_{\bar{k},\sigma} | \varphi_0 \rangle) + c.c.]
\]
where explicit expressions for \( q(\lambda, n_{\uparrow}) \) and various other expectation values can be found in eqs. (45), (49), and (51). In the following we consider the case where
there is no direct coupling between the impurities, \(|t_{12}| \to 0\).

To facilitate the optimization with respect to the normalized single-particle state \(|\varphi_0\rangle\), we consider the Lagrange functional

\[
\mathcal{L} = \mathcal{L}(|\varphi_0\rangle, \hat{t}_{12}, n_{1,\uparrow}) \quad \text{and} \quad \mathcal{L} = E_{\text{var}}(\lambda; |\varphi_0\rangle) \\
- \hat{t}_{12} \sum_\sigma (1 - 2n_{1,\uparrow} + \langle \varphi_0 | \hat{n}_{1,\sigma} - \hat{n}_{2,\sigma} | \varphi_0 \rangle), \tag{53}
\]

where we consider the Gutzwiller parameters \(\lambda\) fixed. Here, we introduced the Lagrange parameter \(\hat{t}_{12}\). It guarantees that \(n_{1,\uparrow} = \langle \varphi_0 | \hat{n}_{1,\uparrow} | \varphi_0 \rangle\) holds for the optimal \(|\varphi_0\rangle\); the other impurity occupancies follow from particle-hole and spin-flip symmetry.

The minimization of \(\mathcal{L}\) with respect to the single-particle product state \(|\varphi_0\rangle\) shows that \(|\varphi_0\rangle\) must be a normalized eigenstate of an effective, non-interacting particle product state that fixes \(\hat{\nu}_0\) (or appendix A of [26], normalized eigenstate of an effective, non-interacting Lagrange functional normalized single-particle state \(|\varphi_0\rangle\).

\[
E = \int \phi^\dagger \phi \d x. \tag{54}
\]

It is natural to choose \(|\varphi_0\rangle\) as the normalized ground state of \(H_0^{\\text{eff}}\). Moreover, the derivative of the Lagrange function \(\mathcal{L}\) with respect to \(\hat{t}_{12}\) gives back the condition \(n_{1,\uparrow} = \langle \varphi_0 | \hat{n}_{1,\uparrow} | \varphi_0 \rangle\) (55) that fixes \(\hat{t}_{12}(q, n_{1,\uparrow})\).

4.6. Atomic limit

In the atomic limit, \(V \equiv 0\), we must project onto the atomic eigenstates with minimal energy, \(E_0 = -U/2\). Therefore, as seen from table L1, we must set \(\lambda_1 = \lambda_0 = \lambda_{11} = \lambda_{n0} = 0\). Furthermore, \(\lambda_2 = \lambda_A = 0\) guarantees \(\bar{q} = 0\). The two constraints (A.6) and (A.8) reduce to

\[
\begin{align*}
1 &= C_1 = 3(\lambda_{00})^2 n^2 \bar{n}^2 + (\lambda_{10})^2 (n^4 + \bar{n}^4)/2, \\
2n &= 2C_2 = 3(\lambda_{00})^2 n^2 \bar{n}^2 + (\lambda_{10})^2 n^4.
\end{align*} \tag{56}
\]

This gives

\[
(\lambda_{00})^2 = \frac{1}{3n^2 \bar{n}^2} \left(1 - \frac{(n - \bar{n})(n^4 + \bar{n}^4)}{n^4 - \bar{n}^4}\right), \tag{57}
\]

\[
(\lambda_{10})^2 = \frac{2(n - \bar{n})}{n^4 - \bar{n}^4}. \tag{57}
\]

From eq. (54) it follows that the interaction energy is \(E_{\text{int}} = -U/2\) for all \(n\), as it must. The occupation probabilities for the spin triplet and the spin singlet are given by \(p_n = 3(\lambda_{00})^2 n^2 \bar{n}^2\) and \(p_{\bar{n}} = (\lambda_{10})^2 (n^4 + \bar{n}^4)/2\), respectively. For \(n = \bar{n} = 1/2\) we find \(\lambda_{00} = \lambda_{10} = 2\) so that \(p_n = 3/4\) and \(p_{\bar{n}} = 1/4\), as it should for two uncoupled spins on the impurity sites.

5. Tight-binding host electrons

To obtain explicit results, we consider host electrons on a simple cubic lattice with nearest-neighbor hopping of band width \(W \equiv 1\),

\[
\epsilon(k) = -\frac{1}{6} (\cos(kx) + \cos(ky) + \cos(kz)). \tag{58}
\]

We address the case of a small local hybridization, \(V_k \equiv V \ll 1\). In addition, we assume that the impurities are on different sublattices so that \(R = R_1 - R_2 \in B\)-lattice. In this case, the hybridization functions between odd and even channels vanish, \(H_{12}(\omega; R) = H_{21}(\omega; R) = 0\), see MBG. The case where \(R_1\) and \(R_2\) belong to the same sublattice is addressed briefly in Appendix C.

In MBG we derived the single-particle energy, eq. (54), and the local particle density, eq. (55). Here, we summarize the results for the non-interacting case with a purely local hybridization. For the interacting case, \(V\) must be replaced by \(qV\).

5.1. Hybridization functions and density of states

With the abbreviations

\[
R_1(\omega; R) = \Lambda_0(\omega) - \Lambda_B(\omega; R),
\]

\[
R_2(\omega; R) = \Lambda_0(\omega) + \Lambda_B(\omega; R),
\]

\[
I_1(\omega; R) = D_0(\omega) - D_B(\omega; R),
\]

\[
I_2(\omega; R) = D_0(\omega) + D_B(\omega; R)
\]

the hybridization functions are given by

\[
H_{b,b}(\omega; R) = V^2 R_0(\omega; R) - i\pi V^2 I_0(\omega; R). \tag{60}
\]

For electrons with nearest-neighbor transfers on a simple-cubic lattice at half band-filling the densities \(D_{A,B}(\omega; R)\) and their Hilbert transforms \(\Lambda_{A,B}(\omega; R)\) are calculated from

\[
\Lambda_A(\omega; R) = \delta_{R \in A} (-1)(R_x + R_y + R_z)/2 \times \int_0^\infty dt \sin(\omega t) J_{R_x} \left(\frac{t}{6}\right) J_{R_y} \left(\frac{t}{6}\right) J_{R_z} \left(\frac{t}{6}\right), \tag{61}
\]

\[
\Lambda_B(\omega; R) = \delta_{R \in B} (-1)(R_x + R_y + R_z + 3)/2 \times \int_0^\infty dt \cos(\omega t) J_{R_x} \left(\frac{t}{6}\right) J_{R_y} \left(\frac{t}{6}\right) J_{R_z} \left(\frac{t}{6}\right). \tag{62}
\]
for $|\omega| \leq 1/2$ and $\mathbf{R} \in B$-lattice. We note that, for $|\mathbf{R}| \gg 1$, the functions $D_{A,B}(\omega; \mathbf{R})$ and $\Lambda_{A,B}(\omega; \mathbf{R})$ oscillate strongly as a function of frequency whereby their amplitude decays approximately proportional to $1/|\mathbf{R}|$.

For $|\omega| > 1/2$, the hybridization functions are purely real, $I_b(\omega; \mathbf{R}) = 0^+$, and, due to particle-hole symmetry, $R_b(-\omega; \mathbf{R}) = -R_b(\omega; \mathbf{R})$ holds. In particular, for $\omega < -1/2$

$$
\Lambda_0(\omega) = -\int_0^\infty d\lambda e^{\lambda \omega} [I_0(\lambda/6)]^3,
$$

$$
\Lambda_B(\omega; \mathbf{R}) = -\int_0^\infty d\lambda e^{\lambda \omega} I_{R_\omega}(\lambda/6)I_{R_\omega}(\lambda/6)I_{R_\omega}(\lambda/6),
$$

(63)

where $I_n(x)$ is the nth-order modified Bessel function.

The continuous impurity contributions to the density of states are given by

$$
D_{b,R}(\omega) = \frac{V^2 I_b(\omega; \mathbf{R})}{N_{b,R}(\omega)},
$$

$$
N_{b,R}(\omega) = [\omega + i\tilde{t}_{12} - V^2 R_b(\omega; \mathbf{R})]^2 + [\pi V^2 I_b(\omega; \mathbf{R})]^2,
$$

(64)

where the upper (lower) sign applies to $b = 1$ ($b = 2$).

In case that the equations

$$
\omega_b \pm i\tilde{t}_{12} - V^2 R_b(\omega; \mathbf{R}) = 0
$$

(65)

have a solution outside the band, i.e., for $\omega_b < -1/2$, then the impurity density of states has a $\delta$-peak contribution because $I_b(\omega_b; \mathbf{R}) = 0^+$. The contribution to the impurity density of states is

$$
D_{b,R}^\delta(\omega) = Z_{R,b} \delta(\omega - \omega_b),
$$

$$
Z_{R,b} = \frac{1}{1 - V^2 R_b(\omega_b; \mathbf{R})}.
$$

(66)

We set $Z_{R,b} \equiv 0$ if eq. (65) has no solution outside the band. Recall that in eqs. (65) and (66) the functions have to be calculated outside the band, i.e., eqs. (63) must be employed to calculate $R_b(\omega; \mathbf{R})$ and its derivative.

Apart from the bare density of states, the host electron contribute

$$
D_{\text{host},b,R}(\omega) = \frac{1}{\pi} \text{Im} \left[ \frac{H_{b,R}(\omega)}{\omega + i\tilde{t}_{12} - H_{b,R}(\omega)} \right],
$$

(67)

where the prime indicates the partial derivative with respect to $\omega$. We thus find

$$
D_{\text{host},b,R}(\omega) = -V^2 I'_b(\omega; \mathbf{R}) \frac{\omega + i\tilde{t}_{12} - V^2 R_b(\omega; \mathbf{R})}{N_{b,R}(\omega)} - \frac{V^2 I_b(\omega; \mathbf{R}) R'_b(\omega; \mathbf{R})}{N_{b,R}(\omega)},
$$

(68)

5.2. Particle density and single-particle energy

The particle density for given $\mathbf{R}$ is obtained from

$$
n_{b,\sigma} = Z_{R,b} + \int_{-1/2}^0 d\omega D_{b,R}(\omega),
$$

(69)

where we suppressed the lattice index in the particle density to shorten the expressions. The two levels do not hybridize explicitly for $\mathbf{R} \in B$-lattice. Therefore, the ground-state energy of the non-interacting two-impurity Anderson model can be cast into the form

$$
E_{sp}(V, \tilde{t}_{12}) = 2(Z_{\mathbf{R},1} \omega_1 + Z_{\mathbf{R},2} \omega_2)
$$

$$
+ \frac{2}{\pi} \sum_b \int_{-1/2}^0 d\omega \cot^{-1} \left[ \eta_{b,\mathbf{R}}(\omega) \right]
$$

(70)

with the phase-shift function

$$
\eta_{b,\mathbf{R}}(\omega) = \frac{\omega + \tilde{t}_{12} - V^2 R_b(\omega; \mathbf{R})}{\pi V^2 I_b(\omega; \mathbf{R})}.
$$

(71)

Note that we introduced the continuous and continuously differentiable function $\cot^{-1}(x) = \cot^{-1}(x) - \pi\Theta(x)$ with the Heaviside step function $\Theta(x)$.

The expression (70) is similar to the ground-state energy of the non-interacting symmetric single-impurity Anderson model [27],

$$
E_{\text{SIAM}}(V) = \frac{2}{\pi} \int_{-1/2}^0 d\omega \cot^{-1} \left( \omega - \frac{V^2 \Lambda_0(\omega)}{\pi V^2 D_0(\omega)} \right).
$$

(72)

Since $\Lambda_0(0) = 0$, we do not have to discriminate between $\cot^{-1}(x)$ and the standard inverse cotangent function $\cot^{-1}(x)$. Moreover, for the SIAM there is no bound state outside the band for $V \ll W$.

The energy functional is given by

$$
\tilde{E}_{\text{var}}(\lambda, \tilde{t}_{12}, n_{1,\uparrow}^0) = E_{sp}(qV, \tilde{t}_{12}) - 2\tilde{t}_{12} (1 - 2n_{1,\uparrow}^0) + E_{\text{int}}(\lambda, n_{1,\uparrow}^0),
$$

(73)

where $q \equiv q(\lambda, n_{1,\uparrow}^0)$ from eq. (69). Moreover, the two constraints (36) and (37) must be obeyed. They worked out in Appendix A as eqs. (A.6) and (A.8). The minimization with respect to $\tilde{t}_{12}$ returns eq. (69). The solution of this implicit equation determines $\tilde{t}_{12}(n_{1,\uparrow}^0)$.

5.3. Limit of small hybridization

For small hybridizations, $V \ll 1$, we may expand $E_{sp}(qV, \tilde{t}_{12})$ in $V^2$. To leading order in $(qV)^2 \ln[(qV)^2]$ and $(qV)^2$,

$$
E_{sp}(qV, \tilde{t}) = 4(qV)^2 d_0 \ln \left[ (qV)^2/C \right]
$$

$$
+ 2(qV)^2 d_0 \ln \left[ (\tilde{t} - \pi \delta_{Rd_R})^2 + (\pi d_0)^2 \right]
$$

$$
- 4(qV)^2 \left( \frac{\tilde{t} - \pi \delta_{Rd_R}}{\pi d_0} \right) \arctan \left( \frac{\tilde{t} - \pi \delta_{Rd_R}}{\pi d_0} \right)
$$

(74)
with \( \bar{t} = \bar{t}_{12}/(qV)^2 \), \( C = 0.7420W \), see Appendix E and \( d_0 = 1.712/W \) is the host electron density of states at the Fermi energy \( E_F = 0 \), see MBG. Moreover, 
\[ \bar{s}_R = (-1)^{(R_x+R_y+R_z+1)/2}, \]
\[ d_R = \int_0^\infty dt R_0(t/6)J_{R_0}(t/6)J_{R_0}(t/6), \]
where \( d_R \) is analyzed in more detail in MBG.

The minimization with respect to \( t \) returns \( \bar{t}_{12} = (qV)^2 \) explicitly, 
\[ \bar{E}_{\text{var}}(\lambda, n_{1,1}) = E_{\text{int}}(\lambda, n_{1,1}) + E_{\text{sp}}(qV, \bar{t}_{12}(n_{1,1}^0)) - 2\bar{t}_{12}(q,n_{1,1}^0)(1-2n_{1,1}^0) \]

\[ = E_{\text{int}}(\lambda, n_{1,1}^0) + 4d_0(qV)^2 \left( \ln (qV^2d_0/C) - \ln [\cos(n_{1,1}^0 - \pi/2)] \right) + 4(qV^2(n_{1,1}^0 - \pi/2)\bar{s}_R d_R \]

as a function of the Gutzwiller variational parameters \( \lambda \) and of the level occupancy \( n_{1,1}^0 \) in the effective non-interacting problem. It can be shown analytically that the choice \( y_m = 0 \) is optimal.

6. Kondo limit

In the Kondo limit of large Hubbard interactions, \( U \gg \Gamma \), the Gutzwiller variational energy functional can be minimized analytically to a far extent.

6.1. Simplification of the variational energy functional

In the Kondo limit \( U \gg \Gamma \), we can safely set \( \lambda_1 = \lambda_9 = 0 \). Moreover, to obtain explicit expressions, we set \( \lambda_{11} = x_m = 0 \). This is an excellent approximation for all \( n \equiv n_{1,1}^0 \) but finite \( \lambda_{11}, x_m \) slightly improve the variational energy in the limits \( n \to 0 \) and \( n \to 1 \).

The two constraints \( (A.6) \) and \( (A.8) \) become
\[ 1 = 3\lambda_6 n^2 \bar{n} + 4(\lambda_2^2 n^2 \bar{n} + \lambda_5^2 n \bar{n}^2) + \frac{\lambda_{10}^2}{2} (n^4 + \bar{n}^4), \]
\[ 2n = 3\lambda_6^2 n^2 \bar{n} + 6\lambda_2^2 n^2 \bar{n} + 2\lambda_5^2 n \bar{n}^2 + \lambda_{10}^2 n^4. \]

These two equations can be solved analytically for \( \lambda_6 \) and \( \lambda_{10} \) as a function of \( \lambda_2, \lambda_5 \).

The interaction energy reads
\[ -\frac{2E_{\text{int}}}{U} = 3\lambda_6^2 n^2 \bar{n} + \frac{\lambda_{10}^2}{2} (n^4 + \bar{n}^4). \]

Using eq. \( (75) \), it only depends on \( \lambda_2, \lambda_5 \),
\[ \frac{2}{U} (E_{\text{int}} + \frac{U}{2}) = 4\lambda_2^2 n^3 + 4\lambda_5^2 n \bar{n}^3. \]

Furthermore, to leading order in \( 1/U \) we find for the hybridization renormalization factor
\[ q = \frac{\lambda_2 \lambda_6}{2} n^3 + \frac{3\lambda_2 \lambda_5}{2} n^2 \bar{n} + \frac{3\lambda_5 \lambda_{10}}{2} n \bar{n}^2 + \frac{\lambda_{10}^2}{2} n^3 \equiv \gamma_2(n)\lambda_2 + \gamma_4(n)\lambda_4. \]

With the atomic values for \( \lambda_6^* \) and \( \lambda_{10}^* \) from eq. \( (77) \) we have explicitly
\[ \gamma_2 = \gamma_2(n) = \frac{3}{2} n^2 \lambda_6 + \frac{1}{2} n \lambda_{10} \]
\[ = \frac{3}{2} n^2 \bar{n} \mathcal{E} \left( \frac{1}{n^2 \bar{n}^2} \left( 1 - (n - \bar{n})(n^4 + \bar{n}^4) \right) \frac{1}{n^2 \bar{n}^2} \right) \]
\[ + \frac{1}{2} n^3 \mathcal{E} \frac{2(n - \bar{n})}{n^4 - \bar{n}^4}, \]

\[ \gamma_4 = \gamma_4(n) = \frac{3}{2} n^2 \lambda_6 + \frac{1}{2} n \lambda_{10} \]
\[ = \frac{3}{2} n^2 \bar{n} \mathcal{E} \left( \frac{1}{n^2 \bar{n}^2} \left( 1 - (n - \bar{n})(n^4 + \bar{n}^4) \right) \frac{1}{n^2 \bar{n}^2} \right) \]
\[ + \frac{1}{2} n^3 \mathcal{E} \frac{2(n - \bar{n})}{n^4 - \bar{n}^4}. \]

Eq. \( (81) \) reveals that \( \lambda_2, \lambda_4 \) are of the order of \( q \). We set \( \lambda_2 = q\mu_2 \) and \( \lambda_4 = q\mu_4 \) so that the condition \( (80) \) is fulfilled if the variational parameters \( \mu_2 \) and \( \mu_4 \) obey \( 1 = \mu_2 \gamma_2 + \mu_4 \gamma_4 \) or \( \mu_4 = (1 - \mu_2 / \gamma_2) / \gamma_4 \). Thus, the optimization of \( (80) \) with respect to \( \mu_2 \) leads to
\[ \lambda_2 = q\frac{n^2 \gamma_2}{\gamma_2 n^2 + \gamma_4 n^2}, \quad \lambda_4 = q\frac{n^2 \gamma_4}{\gamma_2 n^2 + \gamma_4 n^2}, \]
and the remaining variational parameters are \( q \) and \( n \equiv n_{1,1}^0 \).
6.2. Optimization of the density parameter

The optimal value for the level occupancy remains to be determined. We insert the optimal value for \( q(n) \) from eq. (39) in eq. (57) to find \((\ln(\epsilon)) = 1\)

\[
\tilde{E}_{\text{var}}^K(n) = \tilde{E}_{\text{var}}^K(q(n), n) = -B[q(n)]^2 = -\frac{B}{e^{A(n)}}
\]

\[
= -\frac{4C}{\pi e} \cos(\pi n - \pi/2) \times \exp\left(\frac{-\pi p(n)U}{16\Gamma} - \frac{(n\pi - \pi/2)}{d_0}\right) \tag{90}
\]

with \( \Gamma = \pi d_0 V^2 \). The result suggests a Kondo-type variational energy as in the single-impurity case, see eq. (30) and Appendix D, eq. (D.21). This, however, is actually not the case in the present Kondo limit, as we show now.

The density-dependent factor \( p(n) \) monotonically increases (decreases) for \( n < 1/2 \) (\( n > 1/2 \)) and reaches its minimum at \( n = 0 \) (\( n = 1 \)), \( p(0) = p(1) = 0 \). Therefore, we expand \( A(n) \) around \( n = 0 \) (\( n = 1 \)). For small Kondo couplings, we expand \( \tilde{E}_{\text{var}}^K(n) \) to linear order in \( n \) for \( \tilde{R}_dR_\| > 0 \) and to linear order in \( (1 - n) \) for \( \tilde{R}_dR_\| < 0 \), respectively. With \( \tilde{n} \equiv n \) for \( n \to 0 \) and \( \tilde{n} = 1 - n \) for \( n \to 1 \) we find

\[
\tilde{E}_{\text{var}}^K(n \to 0) \approx -\frac{4C}{e} \exp\left[\frac{\pi d_R}{2d_0}\right] \times \tilde{n} \exp\left[-\frac{\pi U}{\Gamma} + \frac{\pi d_R}{d_0}\right] \tag{91}
\]

The minimum is found at

\[
\tilde{n}_{\text{opt}} = \left(\frac{\pi U}{\Gamma} + \frac{\pi d_R}{d_0}\right)^{-1} \approx \frac{\Gamma}{\pi U} \tag{92}
\]

with

\[
\tilde{E}_{\text{var}}^K = -\frac{4CT}{\pi U} \exp\left[\frac{\pi d_R}{d_0} - 2\right] + \mathcal{O}(1/U^2) \tag{93}
\]

Eq. (93) shows the absence of a Kondo screening. The two magnetic impurities sense each other via the host-electron bath. They form a spin singlet with a Heisenberg-type energy gain proportional to \( \Gamma/U \). It is spatially modulated by a distance-dependent factor of order unity, \( \exp(\pi d_R/(2d_0)) = \mathcal{O}(1) \).

Note that \( n \equiv n_{1,\uparrow}^1 \) which is close to zero or unity, has no physical significance. In fact, the physical level occupancy \( \langle \hat{n}_{1,\uparrow} \rangle_G = \langle \hat{h}_{1,\uparrow}^\dagger \hat{h}_{1,\uparrow} \rangle_G \) in eq. (15) is close to half filling. For strong coupling where \( \lambda_1 = \lambda_2 = 0 \), \( \lambda_3 \), \( \lambda_4 \) are given by eq. (84), we find for \( n_{\text{opt}} \to 1 \)

\[
\langle \hat{n}_{1,\uparrow} \rangle_G = \langle \hat{h}_{1,\uparrow}^\dagger \hat{h}_{1,\uparrow} \rangle_G = \frac{1}{2} + 2(q^{\text{opt}})^2 \tilde{n}_{\text{opt}} \tag{94}
\]

with the optimal value for the hybridization reduction factor

\[
(q^{\text{opt}})^2 = \frac{C}{\mathcal{U}} \exp\left[\frac{\pi d_R}{2d_0} - 2\right] + \mathcal{O}(1/U^2) \tag{95}
\]

Note that we derived these results under the condition \( q \ll 1 \) which required \( U \gg \Gamma \), i.e., we had to address the Kondo limit. In eq. (95) only the ratio \( W/U \) enters and \( q \ll 1 \) also requires \( U \gg W \), i.e., we should not be far from the spin-model limit. Under these conditions, the impurity levels are almost exactly half filled, with corrections of the order \( 1/U^2 \).

The energy gain (93) can be interpreted in terms of an effective direct electron transfer between the impurities, see our discussion in Sect. 5.3 and eq. (23).

\[
|t_{12}^\text{direct}(R)| = \sqrt{\frac{C}{\pi}} \exp\left[\frac{\pi d_R}{4d_0} - 1\right] \sim \mathcal{V} \tag{96}
\]

Recall that, for \( V \ll W \), \( E_{\text{var}}^K \) is an exact variational bound to the ground-state energy in the Kondo limit. The result (96) suggests a direct electron transfer proportional to \( V \) also in the exact solution. This is indeed the case for a few-orbital toy model, as we show in Appendix F and has also been found in the original antiferromagnetic Hartree-Fock study [13].

Moreover, the induced direct coupling does not vanish for large impurity separations, \( |R| \to \infty \). The origin of this somewhat surprising result lies in the fact that, for \( U \approx W \gg \Gamma \), the impurity electrons couple to all electrons in the system so that the physical distance of the impurity levels is of minor importance. This is also reflected in the energetic position of the impurity levels of the effective non-interacting two-impurity Hamiltonian. They are located deep in the bare band at \( |n_{1,\uparrow}^{\text{opt}}(\Gamma \gg 1)| = (C/e^2)W = \mathcal{O}(W/2) \) so that not only energy levels close to the Fermi energy are involved in the exchange interaction.

In summary, the results in this section show that, in the Kondo limit \( U \gg \Gamma \) and for \( U \approx W \), there is only a phase with singlet pairs and no transition to a phase with weakly coupled impurities can be realized. Therefore, the transition is only conceivable for \( U/\Gamma \) values that require a numerical minimization of the variational energy functional. This will be discussed in Sect. 7. Nevertheless, as we discuss next, signatures of the transition are already discernible in the Kondo-limit energy functional (80).

6.3. Quantum phase transition from the Kondo energy functional for small hybridizations

When the hybridization is (unrealistically) strong, we can obtain a nontrivial Kondo-type solution of the variational energy functional (90) that permits an analytic discussion of the Varma-Jones scenario on the basis of our Gutzwiller variational approach.

To obtain an exponential energy dependence, the minimum in eq. (90) has to show up for values of \( n \) that are not close to zero or unity. This is possible if

\[
\frac{U}{\Gamma} < 8\pi[1 - (d_R/d_0)^2] \tag{97}
\]
The RKKY energy enhancement is given by the variational Kondo energy, see Appendix D, eq. (D.21). As in eq. (30) with the single-impurity Gutzwiller variational approach to the two-impurity Anderson model at particle-hole symmetry.

We minimize numerically the full energy functional into Heisenberg singlets. Beyond a critical separation, we find a Heisenberg-type singlet, we find \( E(R) \) \( \approx \frac{\pi}{4} \Gamma \approx \frac{1}{2} \frac{U}{\Gamma} \) \( \approx \frac{\pi}{4} \Gamma, \) the impurity spins (discontinuously) bind into a Heisenberg-type singlet at particle-hole symmetry.

Equation (99) permits a simple interpretation of the RKKY energy (77). For the case of a general \( V \), we evaluate and store \( 10^6 \) values in the interval \([-1/2, 0]\) for the densities \( D_{A,B}(\omega_j; R) \) and their Hilbert transforms \( \Lambda_{A,B}(\omega_j; R) \) for each \( R \). These discrete values provide sampling points for the frequency integrations. The relative accuracy of all data is better than \( 10^{-6} \). We do not encounter bound states, \( Z_{R,b} = 0 \) in eqs. (69), (70).

### 7.1. Ground-state energy and phase transition

In Fig. 1 we show the variational ground-state energy as a function of the density \( n = n^0, \) to illustrate the variational transition at \( R = 0, 0 > \) for \( V = 0.2 \) and \( W = 1 \). Below the transition, \( U \lesssim U_c(R) = 12.172 \Gamma \), the optimal variational wave function describes weakly RKKY-interacting Kondo spins, compare eq. (97), with \( n^0 \approx 0.517 \), see eq. (98). Above the transition, \( U \gtrsim U_c(R) \), the system prefers to form a Heisenberg-type singlet at \( n^0 \approx 0.966 \).

As seen from the figure, the energy functional resembles that of a fourth-order Landau functional for phase transitions \([31]\) with even and odd powers, where \( n \) is the order parameter, \( \Gamma/U \) acts as temperature and \( d_R \) plays the role of an external field. Therefore, we find a discontinuity in \( n \) and a tricritical point \([32]\).

In general, at the transition the value of \( n \) jumps from \( n_K \) for \( U < U_c \) to \( n_H \) for \( U > U_c \). As we discuss in more detail in Sect. 7.3, this jump can also be seen in the multiplet occupations and in the expectation value for the inter-impurity electron transfer,

\[
\tau = -\frac{\langle \hat{T}_{d} \rangle}{2|t_{12}|} = \langle \hat{n}_{1,1} \rangle G - \frac{1}{2}.
\]

![Variational ground-state energy](image.png)

**Figure 1.** Variational ground-state energy \( E_{\text{var}}(n) \) as a function of the density \( n = n^0, \) in the effective non-interacting two-impurity model \([33]\) for \( V = 0.2, W = 1 \) and three values of \( U \) at and in the vicinity of the critical value \( U_c(R) = 12.172 \Gamma \). The energies are shifted by their value at the minimum, \( E_{\text{var}}(n^0)/\Gamma = -6.175732, -6.250614, -6.347167 \) for \( U/\Gamma = 11.972, 12.172, 12.372 \), respectively. For \( U < U_c(R) \), we observe weakly RKKY-interacting Kondo-screened spins, \( n^0 \approx 0.517 \), for \( U > U_c(R) \) the impurity spins are bound into Heisenberg-type singlets, \( n^0 \approx 0.966 \).

For a fixed impurity separation \( R \), the scenario of Varma and Jones can be realized as a function of \( U/\Gamma \). For a small enough \( U/\Gamma \), we have \( n \approx 1/2 \) in the effective single-particle Hamiltonian, and the impurities represent weakly interacting Kondo-screened spins, as expressed by eq. (99). Upon increasing \( U/\Gamma \), the impurity spins (discontinuously) bind into a Heisenberg-type singlet, we find \( |n - 1/2| \approx 0.4 \), and the Kondo screening is absent. The same scenario can be obtained for a suitable fixed \( U/\Gamma \) as a function of \( R \). For short distances, the impurity spins are bound into Heisenberg singlets. Beyond a critical separation, \( |R| > R_c \), weakly interacting Kondo-screened spins appear.

### 7. Numerical minimization

We minimize numerically the full energy functional in eq. (95) using a conjugate gradient method in combination with the augmented penalty method \([28, 29, 30]\). On a modern CPU, the optimization for fixed model parameters is a matter of seconds if we use the small-\( V \) approximation for the single-particle energy (77).

For the case of a general \( V \), we evaluate and store \( 10^6 \) values in the interval \([-1/2, 0]\) for the densities \( D_{A,B}(\omega_j; R) \) and their Hilbert transforms \( \Lambda_{A,B}(\omega_j; R) \) for each \( R \). These discrete values provide sampling points for the frequency integrations. The relative accuracy of all data is better than \( 10^{-6} \). We do not encounter bound states, \( Z_{R,b} = 0 \) in eqs. (69), (70).
7.2. Phase diagram

In Fig. 2 we show the ground-state phase diagram for $V = 0.2$ and $V = 0.01$ ($W = 1$), respectively. Below the transition line $U_c(R)$, weakly interacting impurities are observed. The critical line marks discontinuous changes in the variational parameter $n = n_{\uparrow,\uparrow}$ that show up, e.g., in the inter-impurity transfer matrix element $\tau$, eq. (101). As seen in the inset, the jump $\Delta \tau$ goes to zero at the critical endpoint $(R_c, U_c)$. The mean-field exponent of one half is seen from the linear behavior of $(\Delta \tau)^2$.

The transition is continuous below $R_c$ so that for $R =< 1, 0, 0>$ the singlet state continuously forms from weakly-coupled impurities. Therefore, in numerical simulations of the two-impurity Anderson model, the impurity distance must not be chosen too small to observe a conceivable transition, as suggested by our variational approach.

In Fig. 3 we show the critical interaction strength $U_c/\Gamma$ as a function of $V$ for various values of $R =< R, 0, 0>$ ($R = 3, 5, \infty$). In the numerically accessible region, $2 \cdot 10^{-3} \leq V \leq 2 \cdot 10^{-1}$ the critical parameter lies in the range $U_c/\Gamma = 12 \ldots 16$. This demonstrates that $\Gamma$ is indeed the relevant energy scale with which $U$ must be compared. Furthermore, this confirms out previous claim in Sect. 7.2 that the transition never occurs in the Kondo or spin-model limits, $U \gg \Gamma$ or $U \gg W$, respectively. Therefore, the transition is difficult to access using effective spin models that approximate the two-impurity Anderson model, or by any perturbative method [33, 34, 35].

[Figure 2 and Figure 3 are shown here]

7.3. Multiplet occupations and effective inter-impurity transfer matrix element

To gain further insight into the properties of the two different Gutzwiller variational states, we discuss the probability to find the two impurities in a spin triplet and a spin singlet configuration. From spin symmetry we find that $\langle \hat{m}_S \rangle_G = \langle \hat{m}_T \rangle_G = \langle \hat{m}_S \rangle_G$ so that $p_t = 3\langle \hat{m}_S \rangle_G = 3\lambda_0^2 n^2 \bar{n}^2$ (102)
is the probability to find a triplet state. The probability for a spin-singlet configuration is given by

\[ p_s = \langle \hat{m}_{10} \rangle_G = \frac{\lambda_2^2 + \frac{x_m^2}{2}(n^4 + \bar{n}^4) + \lambda_{10} x_m (n^4 - \bar{n}^4)}{2}. \]  

(103)

In Fig. 4, we show \( p_t \) and \( p_s \) for \( R = <5,0,0> \) as a function of \( U/\Gamma \) across the transition. For \( V = 0.2 \), \( p_t + p_s \approx 1 \) for all \( U \gtrsim 12 \Gamma \) so that we are close to the Kondo limit where the impurities are singly occupied. For \( U > U_c(R) \), the probability \( p_t \) to find one of the three triplet states is small compared to \( p_s \) so that we can safely argue that the two impurity spins form a Heisenberg-type singlet state. In contrast, for \( U < U_c(R) \), the probability \( p_s \) for a singlet configuration is only marginally enhanced over the probability \( p_t/3 \) for one of the triplet states. This shows that we have two almost independent (Kondo-screened) spins that display only a small RKKY-interaction induced tendency towards forming a singlet.

For \( V = 0.01 \), the transition occurs far from the Kondo limit. As seen from Appendix D, the single-impurity Anderson model for \( V = 0.01 \) quantitatively enters the Kondo regime for \( U/T \gtrsim 50 \). Therefore, in contrast to the case \( V = 0.2 \), the impurity electrons remain fairly itinerant across the transition. This can be seen in Fig. 4 as the sum of the probabilities for spin singlet and triplet configurations is noticeably below unity.

\[ V = 0.2, W = 1, R = <5,0,0> \]

\[ V = 0.01, W = 1, R = <5,0,0> \]

Figure 4. Probabilities \( p_t \) and \( p_s \) to find the impurity spins in the triplet and the singlet configuration at \( R = <5,0,0> \) as a function of the interaction strength \( U/\Gamma \) for \( V = 0.2 \), upper part (\( V = 0.01 \), lower part) at \( R = <5,7>,0,0 > \) and \( W = 1 \). At the critical value \( U_c(R) \) the system changes from weakly coupled impurities to the spin-pair phase with sizable inter-impurity electron transfer.

\[ V = 0.2, W = 1, R = <5,0,0> \]

\[ V = 0.01, W = 1, R = <5,0,0> \]

Figure 5. Inter-impurity transfer matrix element \( \tau = \langle \hat{b}_1 \hat{c}_1 \rangle_G - 1/2 \), eq. (101), as a function of the interaction strength \( U/\Gamma \) for \( V = 0.2 \), upper part (\( V = 0.01 \), lower part) at \( R = <5,7>,0,0 > \) and \( W = 1 \). At the critical value \( U_c(R) \) the system changes from weakly coupled impurities to the spin-singlet phase.

In Fig. 5 we show the inter-impurity transfer matrix element \( \tau \) at \( R = <5,7>,0,0 > \) as a function
of the interaction strength \(U/\Gamma\) for \(V = 0.2\) (\(V = 0.01\)), \(W = 1\). The transfer matrix element \(\tau\) differs slightly from zero for all \(U/\Gamma\) because the \(h\)-orbital symmetry is broken already at \(U = 0\), see MBG.

At the transition to the spin-pair phase, the absolute value of the inter-impurity transfer matrix element \(\tau\) increases discontinuously, in general. In the Kondo limit, \(U \gg \Gamma\), the effective transfer matrix element decays proportional to \(1/U^2\), see eq. (94).

When we compare the energies of two variational states that describe different physical situations, we observe a level-crossing as a function of a control parameter, typically some interaction strength. The variational energy remains continuous but, in general, its derivatives are discontinuous. In our case, we see that the probabilities for spin singlet and spin triplet display jump discontinuities at some critical Hubbard interaction. Given the conceptual problems of variational approaches, variational predictions for discontinuous quantum phase transitions should not be overrated.

8. Conclusions

In this work, we analyzed Gutzwiller-correlated variational wave functions as possible ground states for the particle-hole and spin symmetric two-impurity Anderson model. The single-particle product state permits orbital-symmetry breaking in the two-level description that corresponds to a finite single-electron transfer matrix element between the two impurities. As known from the two-site Hubbard model, the two impurities thus have a strong tendency to build a singlet state. As a consequence, we find quantum phase transitions between a regime with weakly coupled, partly Kondo-screened impurities to a spin-pair regime where the impurities form a spin singlet.

It is an advantage of our variational method that it covers effortlessly the whole parameter regime, i.e., we can readily optimize the variational energy function for all \((V, U, W)\) and all impurity separations \(R\). For host electrons that move between nearest neighbors on a simple cubic lattice, we generically find a discontinuous quantum phase transition in the range \(U_c/\Gamma = 12 \ldots 16\) for \(2 \cdot 10^{-3} W \leq V \leq 2 \cdot 10^{-1} W\) where \(\Gamma = \pi d_0 V^2\) and \(d_0 \sim 1/W\) is the density of states at the Fermi energy. Since \(U > 3\Gamma\), the transition cannot be reached using weak-coupling perturbation theory. For small \(V \ll W\), the transition is also far from the Kondo and spin limits where the impurities are singly occupied. Therefore, the transition in the two-impurity Anderson model cannot be described in terms of the two-impurity Kondo limit, in general. It is only in the (unrealistic) case of fairly large hybridizations, \(V = 0.2W\), that we approach the Kondo limit where the impurities are (almost) only singly occupied.

The main difference between our present study and previous approaches to the two-impurity Anderson model lies in the fact that our variational state permits an effective electron transfer between the impurity sites (\(h\)-orbital symmetry breaking). Note that the \(h\)-orbital symmetry is broken already for \(U = 0\) when the impurities are on different sublattices. Therefore, this
feature is generic for the two-impurity Anderson model. For large interactions, $U \gg \Gamma$, the ground-state energy is bound from above by the gain proportional to $V^2/U$. Since this is an exact bound we argue that a direct coupling of the impurities via an electron transfer proportional to $V$ also is a feature of the exact ground state, up to possible non-analytic corrections. This picture was seen in Alexander and Anderson’s antiferromagnetic Hartree-Fock analysis [18] and is supported by our few-orbital toy-model study.

For a qualitative understanding of our results we refer to our central finding in MBG, namely, that even at $U = 0$ we must consider two hybridized impurities. The RKKY approximation starts from bare impurities and thus does not give the correct size and distance-dependence of the interaction between the impurities. These effects are generally not included in analytic approaches, e.g., the real part of the hybridization functions is often ignored. For this reason, the $h$-orbital symmetry breaking is frequently excluded from the beginning.

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Appendix A. Explicit form of the constraints

Appendix A.1. Square of the Gutzwiller correlator

In general, the product $\hat{P}_G^+ \hat{P}_G$ has the form

$$\hat{P}_G^+ \hat{P}_G = \sum_r \lambda_r^2 \hat{m}_r + |\lambda_m|^2 (\hat{m}_{10} + \hat{m}_{11})$$

$$+ [\lambda_m (\lambda_{10} + \lambda_{11})] 10 \{11\} + \text{h.c.}$$

$$= 1 + \hat{P}_G^+ \hat{P}_G |_s + \hat{P}_G^+ \hat{P}_G |_d$$

(A.1)

because $\hat{m}_r$ are projection operators, and we separated the spin-flip and orbital-flip terms from the density terms. We have

$$\hat{P}_G^+ \hat{P}_G |_s = \left[ (\lambda_r^2 - \lambda_m^2) \hat{h}_{1,\uparrow} \hat{h}_{1,\downarrow} \hat{h}_{2,\uparrow} \hat{h}_{2,\downarrow} \right.$$  

$$+ \frac{1}{2} \hat{h}_{1,\uparrow} \hat{h}_{1,\downarrow} \hat{h}_{2,\downarrow} \hat{h}_{2,\uparrow} \times \left[ \lambda_{10}^2 - \lambda_m^2 + (\lambda_m - \lambda_{10}^2 + \lambda_{11}^2) \right] + \text{h.c.} \right.$$  

(A.2)

and

$$\hat{P}_G^+ \hat{P}_G |_d = - 1 + \lambda_r^2 \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \lambda_{10}^2 \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \lambda_{11}^2 \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \lambda_{10}^2 \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \left( \lambda_{10}^2 + \lambda_{11}^2 \right) \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow}$$

where we used the abbreviation $\hat{n}_{b,\sigma} = 1 - \hat{n}_{b,\sigma}$. Due to the constraints [30], [37], and [38], we can cast $\hat{P}_G^+ \hat{P}_G |_d$ into a form where local Hartree bubbles are absent,

$$\hat{P}_G^+ \hat{P}_G |_d = \sum_{(b,\sigma)\leq(b',\sigma')} X_{b,\sigma;\psi',\sigma'} \delta \hat{n}_{b,\sigma} \delta \hat{n}_{\psi',\sigma'}$$

$$+ \sum_{(b,\sigma)\leq(b',\sigma')} Y_{b,\sigma;\psi',\sigma'} \delta \hat{n}_{b,\sigma} \delta \hat{n}_{\psi',\sigma'}$$

$$+ Z \hat{n}_{1,\uparrow} \delta \hat{n}_{1,\downarrow} \delta \hat{n}_{2,\uparrow} \delta \hat{n}_{2,\downarrow},$$

(A.4)

where we introduced the abbreviation $\delta \hat{n}_{b,\sigma} = \hat{n}_{b,\sigma} - n_{b,\sigma}^0$, $n_{b,\sigma}^0 = \langle \varphi | \hat{n}_{b,\sigma} | \varphi \rangle$, and the orbital level order $(1, \uparrow) < (1, \downarrow) < (2, \uparrow) < (2, \downarrow)$. The spin/orbital-flip contribution $\hat{P}_G^+ \hat{P}_G |_d$ is free of Hartree bubbles due to the constraint [39]. For the calculation of the variational ground-state energy, we do not have to know the coefficients $X$, $Y$, and $Z$ explicitly.

Appendix A.2. Constraints

The representation [A.4] requires the constraints [30] and [37] to be fulfilled. Using eq. [A.3] we find

$$1 = \lambda_r^2 (\hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow})$$

$$+ \lambda_{10}^2 (\hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow})$$

$$+ \lambda_{11}^2 (\hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow})$$

$$+ \lambda_{10}^2 (\hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow})$$

$$+ \lambda_{11}^2 (\hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow})$$

$$+ \lambda_{10}^2 (\hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow})$$

$$+ \lambda_{11}^2 (\hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow})$$

$$+ \left( \lambda_r^2 + \lambda_{10}^2 \right) (\hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow} + \hat{n}_{1,\uparrow} \hat{n}_{1,\downarrow} \hat{n}_{2,\uparrow} \hat{n}_{2,\downarrow})$$
Appendix B. Calculation of matrix elements

In this appendix we calculate the matrix elements for the orbital occupancies, the hybridization, and the interaction energy.

Appendix B.1. Orbital occupancies

For the evaluation of the matrix element \( \langle \hat{n}_{1,\uparrow} \hat{n}_{2,\uparrow} \hat{n}_{3,\downarrow} \hat{n}_{4,\downarrow} \rangle \) we first calculate

\[
\langle \hat{n}_{1,\uparrow} \hat{n}_{2,\uparrow} \rangle = \frac{\lambda_2 n_{1,\uparrow} n_{2,\uparrow} + \lambda_6 (n_{1,\uparrow} + n_{2,\downarrow})}{2}
\]

Then, we use \( \hat{P}_G^+ \hat{P}_G = \langle \hat{n}_{1,\uparrow} \hat{n}_{2,\uparrow} \rangle \) to find

\[
\langle \hat{n}_{1,\uparrow} \hat{n}_{2,\uparrow} \rangle = \frac{\lambda_2 n_{1,\uparrow} n_{2,\uparrow} + \lambda_6 (n_{1,\uparrow} + n_{2,\downarrow})}{2}
\]

As a function of the Gutzwiller variational parameters \( \lambda \) and of \( n_{\uparrow} \), we find

\[
n_{1,\uparrow} = \lambda_2 n_{1,\uparrow} n_{2,\uparrow} + \lambda_6 n_{1,\uparrow} + n_{2,\downarrow}
\]

Eq. (A.7) with \( b = 2, \sigma = \uparrow \) is fulfilled due to particle-hole symmetry that leads to \( n_{1,\uparrow} = n_{2,\downarrow} \). The two equations (A.5) and (A.7) fix \( \lambda_6 \) and \( \lambda_10 \) as a function of the remaining variational parameters \( \lambda_1, \lambda_2, \lambda_4, \lambda_9, \lambda_11 \) and \( \lambda_m \). The equations for \( (b,\downarrow) \) do not provide new information because we impose spin-flip symmetry.

Appendix B.2. Correlated impurity occupancy

For the correlated impurity occupancy we thus find

\[
\langle \hat{n}_{1,\uparrow} \rangle = \frac{\lambda_2 n_{1,\uparrow} n_{2,\uparrow} + \lambda_6 n_{1,\uparrow} + n_{2,\downarrow}}{2}
\]

We refer to (B.2) for details.
Appendix B.2. Interaction

Using the atomic spectrum $E_r$, we find from eq. (50)
\[
2 E_{\text{int}} \over U = \lambda_1^2 \langle \varphi_0 | \hat{m}_1 + \hat{m}_{16} | \varphi_0 \rangle + \lambda_2^2 \langle \varphi_0 | \hat{m}_9 + \hat{m}_{12} | \varphi_0 \rangle \\
- \lambda_3^2 \langle \varphi_0 | \hat{m}_6 + \hat{m}_{7} + \hat{m}_{13} | \varphi_0 \rangle \\
- (\lambda_4 - \lambda_m^2) \langle \varphi_0 | \hat{m}_{10} | \varphi_0 \rangle \\
+ (\lambda_5 - \lambda_m^2) \langle \varphi_0 | \hat{m}_{11} | \varphi_0 \rangle \\
+ \left[ \lambda_m (\lambda_{11} - \lambda_{10}) \langle \varphi_0 | (|11\rangle \langle 10|) | \varphi_0 \rangle + \text{c.c.} \right].
\]

(B.4)

After evaluation of the expectation values we find
\[
2 E_{\text{int}} \over U = \lambda_1^2 (n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 + n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 n_{1,1}^0) \\
- \lambda_2^2 (n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 + n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 n_{1,1}^0) \\
+ \frac{\lambda_3^2 - \lambda_\text{m}^2}{2} \times (n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 + n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 n_{1,1}^0) \\
+ (\lambda_4^* + \lambda_\text{m}) (\lambda_{11} - \lambda_{10}) \times (n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 - n_{1,1}^0 n_{1,1}^0 n_{1,1}^0 n_{1,1}^0). 
\]

(B.5)

Appendix B.3. Hybridization

For the evaluation of the matrix element (47) we must express $|\Gamma \rangle \langle \Gamma | \hat{h}_{1,1} \Gamma' \rangle \langle \Gamma'|$ in second quantization. Due to the action of the annihilation operator, the number of impurity electrons in $|\Gamma \rangle$ (and $|\Gamma' \rangle$) is $n_r = 0, 1, 2, 3$ ($n_r = n_r + 1$). The non-vanishing matrix elements are
\[
n = 0: \langle 1 | \langle \hat{h}_{1,1}^{\dagger} | 2 \rangle = \hat{h}_{1,1} n_{1,1} n_{1,1} n_{1,1} n_{1,1}, \tag{B.6}
\]
\[
n = 1: \langle 3 | \langle \hat{h}_{1,1}^{\dagger} | 10 \rangle \langle 10 | = \frac{1}{2} \left[ \hat{h}_{1,1} n_{1,1} n_{1,1} n_{1,1} n_{1,1} - \hat{h}_{1,1} n_{1,1} n_{1,1} n_{1,1} n_{1,1} \right], \tag{B.7}
\]
\[
\langle 3 | \langle \hat{h}_{1,1}^{\dagger} | 11 \rangle = \frac{1}{2} \left[ \hat{h}_{1,1} n_{1,1} n_{1,1} n_{1,1} n_{1,1} + \hat{h}_{1,1} n_{1,1} n_{1,1} n_{1,1} n_{1,1} \right], \tag{B.8}
\]
\[
\langle 3 | \langle \hat{h}_{1,1}^{\dagger} | 10 \rangle = \frac{1}{2} \left[ \hat{h}_{1,1} n_{1,1} n_{1,1} n_{1,1} n_{1,1} \right], \tag{B.9}
\]
\[
\langle 4 | \langle \hat{h}_{1,1}^{\dagger} | 6 \rangle = \hat{h}_{1,1} n_{1,1} n_{1,1} n_{1,1} n_{1,1}, \tag{B.10}
\]
\[
\langle 5 | \langle \hat{h}_{1,1}^{\dagger} | 7 \rangle = \frac{1}{2} \left[ \hat{h}_{1,1} n_{1,1} n_{1,1} n_{1,1} n_{1,1} + \hat{h}_{1,1} n_{1,1} n_{1,1} n_{1,1} n_{1,1} \right], \tag{B.11}
\]
\[
\langle 5 | \langle \hat{h}_{1,1}^{\dagger} | 9 \rangle = \frac{1}{2} \left[ \hat{h}_{1,1} n_{1,1} n_{1,1} n_{1,1} n_{1,1} - \hat{h}_{1,1} n_{1,1} n_{1,1} n_{1,1} n_{1,1} \right]. \tag{B.12}
\]

Appendix C. Results for $R \in A$-lattice

In this appendix we collect some results for the case that the two impurities lie on the same sublattice. We restrict ourselves to the case of small hybridizations, $V \ll W$, and use approximate analytic formulae for the single-particle energy contributions.

Appendix C.1. Single-particle quantities

To leading order in $(qV)^2 \ln [(qV)^2]$ and $(qV)^2$ we have from MBG
\[
E_{\text{sp}}(q, x) = 4(qV)^2 d_0 \left[ \ln \left( (qV)^2 2\pi d_0 / (C x) \right) + \frac{x^2 - 4}{4x} G(x) \right]. \tag{C.1}
\]
where \(G(0 < x < 2) \equiv G_1(x)\) and \(G(x \geq 2) \equiv G_2(x)\) with

\[
G_1(x) = \frac{\pi}{\sqrt{4 - x^2}} - \frac{2}{\sqrt{4 - x^2}} \arctan \left( \frac{x^2 - 2}{x\sqrt{4 - x^2}} \right),
\]

\[
G_2(x) = -\frac{1}{\sqrt{x^2 - 4}} \ln \left( \frac{x^2 - 2 - x\sqrt{x^2 - 4}}{x^2 - 2 + x\sqrt{x^2 - 4}} \right),
\]
and

\[
x \equiv x_R(\bar{t}) = \frac{2\pi d_0}{\sqrt{t^2 + \pi^2(d_0^2 - d_R^2)}},
\]
or

\[
\bar{t}(x) = \pm \frac{\pi}{\sqrt{4 - x^2}} \sqrt{d_0^2 - d_R^2} x.
\]

For a real \(\bar{t}\) we must restrict \(x\) to the region \(0 \leq x \leq 2d_0/\sqrt{d_0^2 - d_R^2}\).

The minimization of \(\bar{E}_{\text{var}}(\lambda, \bar{t}, n_{1,\uparrow})\) with respect to \(\bar{t}\) links \(n_{0,\uparrow}^0\) to \(x\),

\[
n_{0,\uparrow}^0(x) = \frac{1}{2} \pm \frac{\sqrt{(4 - x^2)d_0 + x^2d_R^2} G(x)}{4\pi d_0},
\]
see MBG. We may use \(x\) instead of \(n_{0,\uparrow}^0\) as our variational parameter. Eq. (C.5) shows that for every solution of the minimization equations with \(0 \leq x < 2d_0/\sqrt{d_0^2 - d_R^2}\) we obtain two equivalent solutions for the density, \(n_{0,\uparrow,a}^0 > 1/2\) and \(n_{0,\uparrow,a}^0 = 1 - n_{0,\uparrow,a}^0 < 1/2\). In the following we shall investigate solutions with \(n_{0,\uparrow,a}^0 \geq 1/2\).

The variational energy functional to be minimized is given by

\[
\bar{E}_{\text{var}}(\lambda, x) = E_{\text{int}}(\lambda, n_{0,\uparrow}^0(x)) + E_{\text{xy}}(q, x)
\]

\[
- 2(qV)^2 \bar{t}(x)[1 - 2n_{0,\uparrow}^0(x)]
\]

\[
= E_{\text{int}}(\lambda, n_{0,\uparrow}^0(x))
\]

\[
+ 4(qV)^2 d_0 \left[ \ln \frac{2\pi d_0 (qV)^2}{C_{\lambda}} \right]
\]

\[
+ \frac{x G(x)}{4} \left( \frac{d_R}{d_0} \right)^2 \right]
\]

(C.6)

as a function of the Gutzwiller variational parameters \(\lambda\) and of the parameter \(x\). As in the main text, we choose \(y_m = 0\) in the following.

Appendix C.2. Analytical expressions in the Kondo limit for small hybridizations

When we repeat the steps in Sect. 6.1 we find

\[
\bar{E}_{\text{var}}^K(x) = -\frac{2\pi e}{\pi c} \exp \left( -\frac{\pi e}{\pi c} \frac{n(x)U}{16\Gamma} - \frac{x G(x)}{4} \left( \frac{d_R}{d_0} \right)^2 \right)
\]

(C.7)

for the variational energy in the Kondo limit with \(\Gamma = \pi d_0 V^2\). When \(x = 2d_0/\sqrt{d_0^2 - d_R^2}\) \(n = 1/2\) is the minimum of \(\bar{E}_{\text{var}}^K(x)\), the variational state describes two weakly interacting Kondo-screened impurity spins,

\[
E_{\text{var}}^K = 2E_{\text{opt}}^{\text{SLAM}} \left( 1 - \varepsilon_{\text{RKKY}}^1(\mathbf{R}) \right),
\]

(C.8)

compare eq. (59), with the RKKY energy reduction

\[
\varepsilon_{\text{RKKY}}^1(\mathbf{R}) = \frac{1}{2} \frac{d_R^2}{d_0^2} \ll 1,
\]

(C.9)

compare eq. (100), where the upper index ‘A’ indicates that the impurities are on the same sublattice.

For large \(U\), the two impurity spins are coupled into a singlet. With \(n(x) \approx 1 - x/(2\pi)\) and \(p[n(x)] \approx 8x/\pi\) and neglecting high-order corrections in \(1/U\) we find \(n_{\text{opt}} = 1 - \Gamma/(\pi U)\) as in eq. (92) and

\[
\bar{E}_{\text{var}}^{K,\text{opt}} = -\frac{4C_{\lambda} \Gamma}{\pi^2} \frac{\Gamma}{\pi U} + \mathcal{O}(1/U^2),
\]

(C.10)

as in eq. (93). Since there is no level splitting for \(V = 0\), \(R\)-dependent corrections to first order in \(1/U\) are absent for \(R \in A\)-lattice.

Appendix C.3. Ground-state phase diagram

Fig. C1 shows the ground-state phase diagram. The critical line does not terminate at a tricritical point because a level splitting is absent at the RKKY level. For \(R \in A\)-lattice, the quantum phase transition is discontinuous for all \(R\) because the \(h\)-orbital symmetry is not broken for \(0 \leq U \leq U_c(\mathbf{R})\), and there is no direct electron transfer between the impurities, \(\tau(U < U_c(\mathbf{R})) = 0\). This is seen in the inset of Fig. C1 where we show the variational energy close to the transition for \(R = (4, 0, 0)\).

Figure C1. Ground-state phase diagram for \(R = (4, 0, 0)\) and even \(R\) for \(V = 0.01\). Weakly interacting impurities are found below the critical curve \(U_c(\mathbf{R})\), the phase with spin pairs is found above. The line gives the values for continuous \(R\) in the evaluation of \(d_0\) using the small-\(V\) expression (22). The phase transition is discontinuous for all even \(R\). Inset: Variational ground-state energy \(E_{\text{var}}(n)\) as a function of the density \(n = n_{0,\uparrow}^0\) in the effective non-interacting two-impurity model (53) at \(R = (4, 0, 0)\) for \(V = 0.01, W = 1\) and three values of \(U\) at and in the vicinity of the critical value \(U_c(\mathbf{R}) \approx 14.336\Gamma\) using the small-\(V\) expression (77).
Since the RKKY interaction does not split the $h$-orbital energies, we always find an extremum at $n = 1/2$. Correspondingly, we have $p_s = p_t/3$ for the singlet and triplet occupation probabilities below the transition. Above the transition, $n \neq 1/2$ holds for the optimal variational energy and $\tau$ jumps to a finite value. Likewise, $p_s$ and $p_t$ are discontinuous. Above the transition, $\tau(U > U_c(R))$ decays proportional to $1/U^2$ for large $U/T$.

Apart from the behavior below the transition and apart from the case of neighboring impurities, the differences between odd an even impurity separations are small.

Appendix D. Gutzwiller approach to the single-impurity Anderson model (SIAM)

For comparison and future reference, in this appendix we collect the results for the symmetric SIAM \(^{(24)}\). The results were derived earlier from Gutzwiller variational wave functions, see, e.g., Ref. \(^{(39)}\), and using Kotliar-Ruckenstein slave bosons, see, e.g., Ref. \(^{(40)}\).

Appendix D.1. Gutzwiller variational ground state

As our variational ground state we use the Gutzwiller Ansatz

$$|\Psi_G\rangle = \hat{P}_G|\varphi_0\rangle, \quad \hat{P}_G = \sum_I \lambda_I \hat{m}_I. \tag{D.1}$$

Here,

$$|\varphi_0\rangle = \prod_{k,\sigma} \hat{a}_{k,\sigma}^\dagger |\text{vac}\rangle \tag{D.2}$$

is the ground state of some effective single-particle Hamiltonian,

$$\hat{H}_0^{\text{eff}}|\varphi_0\rangle = E_{\text{wp}}|\varphi_0\rangle. \tag{D.3}$$

The effective single-particle Hamiltonian can be cast into the form

$$\hat{H}_0^{\text{eff}} = \hat{T} + q\hat{V}. \tag{D.4}$$

In equation \(^{(D.1)}\) we employ the projection operators onto the four possible impurity configurations, $I \in \{0, \uparrow, \downarrow, \hat{d}\}$,

$$\hat{m}_0 = \left(1 - \hat{d}_\uparrow^\dagger \hat{d}_\uparrow\right) \left(1 - \hat{d}_\downarrow^\dagger \hat{d}_\downarrow\right), \quad \hat{m}_\uparrow = \hat{d}_\uparrow^\dagger \hat{d}_\uparrow \left(1 - \hat{d}_\downarrow^\dagger \hat{d}_\downarrow\right),$$

$$\hat{m}_\downarrow = \hat{d}_\downarrow^\dagger \hat{d}_\downarrow \left(1 - \hat{d}_\uparrow^\dagger \hat{d}_\uparrow\right),$$

$$\hat{m}_\hat{d} = \hat{d}_\uparrow^\dagger \hat{d}_\uparrow \hat{d}_\downarrow^\dagger \hat{d}_\downarrow, \tag{D.5}$$

and $\lambda_I$ are real-valued variational parameters. We demand that

$$\hat{P}_G^2 = 1 + x \left(\hat{d}_\uparrow^\dagger \hat{d}_\uparrow - 1/2\right) \left(\hat{d}_\downarrow^\dagger \hat{d}_\downarrow - 1/2\right) \tag{D.6}$$

for the paramagnetic half-filled system. This leads to the conditions

$$\lambda_0 = \lambda_d \quad \text{and} \quad \lambda_\sigma = \sqrt{2 - \lambda_d^2}, \quad x = 4(\lambda_d^2 - 1), \tag{D.7}$$

so that $\lambda_d$ is the only remaining variational parameter.

Our choice for the variational parameters $\lambda_0$ and $\lambda_\sigma$ guarantees that the Gutzwiller variational state is normalized,

$$\langle \Psi_G | \Psi_G \rangle = \langle \varphi_0 | \hat{P}_G^2 | \varphi_0 \rangle = 1 \tag{D.8}$$

because

$$\langle \varphi_0 | \hat{d}_\sigma^\dagger \hat{d}_\sigma | \varphi_0 \rangle = 1/2 \tag{D.9}$$

at particle-hole symmetry. Likewise,

$$\langle \Psi_G | \hat{d}_\sigma^\dagger \hat{d}_\sigma | \Psi_G \rangle = \langle \varphi_0 | \hat{d}_\sigma^\dagger \hat{d}_\sigma | \varphi_0 \rangle = 1/2 \tag{D.10}$$

so that the Gutzwiller variational ground state \(^{(D.1)}\) respects particle-hole symmetry.

Appendix D.2. Calculation of the variational energy

For the operator of the kinetic energy we find

$$\langle \Psi_G | \hat{T} | \Psi_G \rangle = \sum_{k,\sigma} \epsilon(k) \langle \varphi_0 | \hat{c}_{k,\sigma}^\dagger \hat{c}_{k,\sigma} | \varphi_0 \rangle$$

$$= \sum_{k,\sigma} \epsilon(k) \langle \varphi_0 | \hat{c}_{k,\sigma}^\dagger \hat{c}_{k,\sigma} | \varphi_0 \rangle, \tag{D.11}$$

where we used eqs. \(^{(D.6)}\) and \(^{(D.9)}\).

For the hybridization operator we find

$$\langle \Psi_G | \hat{V} | \Psi_G \rangle = \frac{1}{\sqrt{L}} \sum_{k,\sigma} V_k \langle \varphi_0 | \hat{P}_G \hat{d}_{\sigma}^\dagger \hat{c}_{k,\sigma} | \varphi_0 \rangle + \text{c.c.}$$

$$= q \frac{1}{\sqrt{L}} \sum_{k,\sigma} \left(V_k \langle \varphi_0 | \hat{d}_{\sigma}^\dagger \hat{c}_{k,\sigma} | \varphi_0 \rangle + \text{c.c.} \right), \tag{D.12}$$

where we used eqs. \(^{(D.1)}\), \(^{(D.5)}\), and \(^{(D.7)}\) to arrive at

$$\hat{P}_G \hat{d}_{\sigma}^\dagger \hat{P}_G = \left[\lambda_\sigma (1 - \hat{d}_\sigma^\dagger \hat{d}_\sigma) + \lambda_d \hat{d}_\sigma^\dagger \hat{d}_\sigma \right] \hat{d}_{\sigma}^\dagger$$

$$\times \left[\lambda_\sigma (1 - \hat{d}_\sigma^\dagger \hat{d}_\sigma) + \lambda_d \hat{d}_\sigma^\dagger \hat{d}_\sigma \right]$$

$$= \lambda_\sigma \lambda_d \hat{d}_\sigma^\dagger. \tag{D.13}$$

Moreover, we introduced the abbreviation

$$q^2 = 2 - \lambda_d^2 \quad \text{and} \quad \lambda_d^2 = 1 - \sqrt{1 - q^2}. \tag{D.14}$$

Equation \(^{(D.12)}\) shows that the Gutzwiller correlator rescales the hybridization by the factor $q$.

For the operator of the interaction energy we find from eqs. \(^{(5)}\) and \(^{(D.10)}\)

$$\langle \Psi_G | \hat{H}_{\text{int}} | \Psi_G \rangle = - \frac{U}{4} + U \langle \varphi_0 | \hat{m}_d \hat{P}_G^2 | \varphi_0 \rangle$$

$$= - \frac{U}{4} + U \lambda_d^2, \tag{D.15}$$

where we also used eq. \(^{(D.1)}\).

Appendix D.3. Minimization of the variational energy

Using the results of the previous subsection, we can express the variational energy as a function of the single variational parameter $q$. 

Appendix D.3.1. Optimization of the single-particle state

The variational energy can be cast into the form

\[ E_{\text{var}}(q, |\varphi_0\rangle) = \frac{\langle \varphi_0 | \hat{T} + q \hat{V} | \varphi_0 \rangle}{\langle \varphi_0 | \hat{V} | \varphi_0 \rangle} = \frac{U(1 - \sqrt{1 - q^2})}{4}, \tag{D.16} \]

where we dropped the constant \(-U/4\).

The optimization of the variational energy with respect to the single-particle state \(|\varphi_0\rangle\) returns the single-particle Schrödinger equation (D.3) with \(H_0\) from eq. (D.4).

\[ q \]

\[ q_0 \]

Figure D1. Hybridization reduction factor \(q\) for the symmetric single-impurity Anderson model as a function of \(U/\Gamma\) (\(\Gamma = \pi d_0 V^2\), \(V = 0.01\), \(d_0 = 1.712\)), in comparison with the analytical strong-coupling expansion, eq. (D.20). The inset shows the probability for a single occupancy as a function of \(U/\Gamma\).

Appendix D.3.2. Gutzwiller variational ground-state energy in the strong-coupling limit

For the optimal single-particle state \(|\varphi_0\rangle\), the variational ground-state energy in presence of the impurity can be written as

\[ E_{\text{var}}(q) = E_0(qV) + \frac{U(1 - \sqrt{1 - q^2})}{4}, \tag{D.17} \]

where \(E_0(V)\) is the energy for the non-interacting SIAM. For a small, constant hybridization \(V_K \equiv V \ll 1\) we have

\[ E_{\text{var}}(q) = 2d_0(qV)^2 \ln \left( \frac{\pi d_0(qV)^2}{C} \right) + \frac{U(1 - \sqrt{1 - q^2})}{4}, \tag{D.18} \]

where \(d_0\) is the density of states per spin direction at the Fermi energy and \(C\) is a constant that depends on the form of the density of states. The minimization of \(E_{\text{var}}(q)\) with respect to \(q\) leads to an implicit equation for \(q(U)\). Its solution for \(V = 0.01\) is shown in Fig. (D.21).

For large interactions, \(U \gg \Gamma = \pi d_0 V^2\), we have \(q \to 0\) and the minimization equation with respect to \(q^2\) becomes

\[ 0 = \ln(q^2) + 1 + \ln(\Gamma/C) + \frac{\pi U}{16d_0}, \tag{D.19} \]

with the solution \((\ln(e) = 1)\)

\[ q_0^2 = \frac{C}{\pi \Gamma} \exp \left( -\frac{\pi U}{16d_0} \right). \tag{D.20} \]

With \(J_K = 4V^2/U\) the optimized variational ground-state energy in the Kondo limit reads

\[ E_{\text{opt}}(J_K \to 0) = -\frac{2C}{\pi \Gamma} \exp \left( -\frac{1}{4d_0 J_K} \right). \tag{D.21} \]

The Gutzwiller variational energy reproduces the exponentially small binding energy but lacks a factor of two in the exponent, i.e., the exact Kondo temperature obeys \(T_K \sim \exp[-1/(2J_Kd_0)]\) [1].

Appendix D.3.3. Kondo limit for the two-impurity Anderson model for a half-filled effective single-particle Hamiltonian

When we restrict ourselves to the case \(n_{b,\sigma} = n = \bar{n} = 1/2\) for the half-filled effective single-particle Hamiltonian, the analysis in Sect. 6.1 carries over to the Kondo limit because \(p(1/2) = 1\), and all density-dependent asymmetric terms vanish. Therefore, in the Kondo limit this variational state describes two isolated impurities with energy

\[ E_{\text{opt}}^\text{SIAM}(J_K \to 0) = 2E_{\text{opt}}^\text{SIAM}(J_K \to 0) = -\frac{4C}{\pi \Gamma} \exp \left( -\frac{1}{4d_0 J_K} \right). \tag{D.22} \]

Appendix E. Cut-off energies for small hybridizations

For completeness, we derive an expression for the constant \(C\) in eq. (D.18) for all density of states.

Appendix E.1. Single-impurity Anderson model

For a general density of states and all \(V \ll 1\) in the non-interacting single-impurity Anderson model, the ground-state energy correction due to the hybridization of the impurity with the host electrons is given by [27]

\[ E_{\text{SIAM}} = \frac{2}{\pi} \int_{-1/2}^0 d\epsilon \cot^{-1} \left( \frac{\epsilon - v \Lambda_0(\epsilon)}{\pi \nu D_0(\epsilon)} \right) = 2 \nu d_0 \ln \left( \frac{\pi \nu d_0}{C} \right) + \mathcal{O}(v^2 \ln \nu), \tag{E.1} \]

where \(v = V^2\), \(D_0(\epsilon)\) is the density of states, and \(\Lambda_0(\epsilon)\) is its Hilbert transform. In the second step, we used the approximation for small \(V\) employed in eq. (D.18).

Therefore, for \(v \to 0\) we find by differentiating both...
sides of eq. (E.1) with respect to $v$ that $L(v) = R(v)$ with

$$L(v) = \int_{-1/2}^{0} de \frac{D_0(\epsilon)}{(e - v\Lambda_0(\epsilon))^2 + (\pi vD_0(\epsilon))^2},$$

$$R(v) = d_0 \left( \ln(v) + 1 + \ln(\pi d_0/C) \right)$$

(E.2)

to order $\ln(v)$ and order unity. We add and subtract an integral that can be evaluated analytically and write

$$L(v) = L(v) - \int_{-1/2}^{0} de \frac{d_0 e}{e^2 + (\pi v d_0)^2} + d_0 \left( \ln(v) + \ln(2) + \ln(\pi d_0) \right).$$

(E.3)

We thus find from $L(v) = R(v)$ in the limit $v \rightarrow 0$ that

$$\ln[\epsilon/(2C)] = \int_{-1/2}^{0} de \left[ \frac{D_0(\epsilon)e/d_0}{(e - v\Lambda_0(\epsilon))^2 + (\pi vD_0(\epsilon))^2} - \frac{e}{e^2 + (\pi v d_0)^2} \right].$$

(E.4)

Letting $v \rightarrow 0$ in this expression gives

$$C = \frac{e}{2} \exp \left[ - \int_{-1/2}^{0} de \frac{D_0(\epsilon)e/d_0}{d_0 e} \right].$$

(E.5)

For a constant density of states with $D_{\text{cons}}(\epsilon) = 1$ for $|\epsilon| \leq 1/2$ we thus obtain $C_{\text{cons}} = e/2 \approx 1.36$, and for a semi-elliptic density of states with $D_{\text{se}}(\epsilon) = (4/\pi)\sqrt{1 - 4\epsilon^2}$ ($|\epsilon| \leq 1/2$), we get $C_{\text{se}} = 1$ [27]. For the simple-cubic lattice with electron dispersion [58] we may rewrite the integral in eq. (E.5) to find

$$\ln C_{\text{se}} = 1 - \ln(2) - \frac{6}{\pi d_0^6} \int_{0}^{\infty} dx [J_0(x)]^3 [\gamma + \ln(3x) - \text{Ci}(3x)],$$

where $J_0(x)$ is the Bessel function to order $n = 0$, $\gamma$ is Euler’s constant, $\text{Ci}(x)$ is the cosine integral, and $d_0 = 1.712$. The integral is readily evaluated numerically [11] to $C_{\text{se}} \approx 0.7420$.

**Appendix E.2. Two-impurity Anderson model**

The ground-state energy can be calculated using the density of states,

$$E_{\text{TIAM}}(V) = E_{\text{TIAM}}^d(V) + E_{\text{host}}^d(V),$$

$$E_{\text{TIAM}}^d(V) = 2 \sum_b \int_{-1/2}^{0} d\omega D_0(\omega),$$

(E.7)

$$E_{\text{host}}^d(V) = 2 \int_{-1/2}^{0} \sum_b d\omega D_{\text{host},b}(\omega),$$

(E.8)

where we suppress the $\mathbf{R}$-dependence for convenience.

**Appendix E.2.1. Impurity contribution** Using $d_0 = D_0(0)$ and the abbreviations $\alpha = V^2(\tilde{t} - \pi \tilde{s}_R \tilde{d}_R)$, $\beta = \pi V^2 d_0$, and $\tilde{t}_{12} = V^2 \tilde{t}$ we eliminate the logarithmically divergent terms in the integrand,

$$\frac{E_{\text{TIAM}}^d(V)}{2V^2 d_0} = \int_{-1/2}^{0} d\omega \omega \left( \frac{D_1(\omega) + D_2(\omega)}{V^2 d_0} - \frac{1}{(\omega + \alpha)^2 + \beta^2} - \frac{1}{(\omega - \alpha)^2 + \beta^2} \right) + \ln(\alpha^2 + \beta^2) - \frac{1}{2} \ln[(\alpha + 1/2)^2 + \beta^2] - 2\frac{\alpha}{\beta} \arctan \left( \frac{\alpha}{\beta} \right) + \frac{\alpha}{\beta} \arctan \left( \frac{\alpha + 1/2}{\beta} \right).$$

(E.9)

Now, we are in the position to let $V \rightarrow 0$ both in the integrand as well as in all other terms ($\alpha \rightarrow 0$, $\beta \rightarrow 0$, and $\alpha/\beta$ remains finite),

$$\frac{\Delta E_{\text{TIAM}}^d(V)}{2V^2 d_0} \approx 2 \int_{-1/2}^{0} d\omega \frac{D_0(\omega) - d_0}{d\omega} + 2 \ln(V^2) + \ln(4) + \ln \left[ (\tilde{t} - \pi \tilde{s}_R \tilde{d}_R)^2 + (\pi d_0)^2 \right] - 2 \frac{(\tilde{t} - \pi \tilde{s}_R \tilde{d}_R)}{\pi d_0} \tan^{-1} \left( \frac{\tilde{t} - \pi \tilde{s}_R \tilde{d}_R}{\pi d_0} \right).$$

(E.10)

**Appendix E.2.2. Host contribution** For small $V$ we have

$$D_{\text{host}}(\omega) \approx \frac{V^2}{\pi} \text{Im} \left[ \frac{R_1(\omega; \mathbf{R}) - \pi \tilde{s}_R'(\omega; \mathbf{R})}{\omega - \alpha + i3} \right] + \frac{V^2}{\pi} \text{Im} \left[ \frac{R_2(\omega; \mathbf{R}) - \pi \tilde{s}_R'(\omega; \mathbf{R})}{\omega + \alpha + i3} \right].$$

(E.11)

In the energy integral (E.8) we may safely let $V \rightarrow 0$ to obtain the second-order term,

$$E_{\text{TIAM}}^d(V) \approx -4V^2 \int_{-1/2}^{0} d\omega \omega \frac{D_0(\omega)}{\omega} = -4V^2 d_0,$$

(E.12)

where we used $D_0(0) = d_0$ and $D_0(-1/2) = 0$.

**Appendix E.2.3. Calculation of $C$** Altogether, we find up to and including all terms to order $V^2$

$$\frac{E_{\text{TIAM}}(V)}{4V^2 d_0} \approx \ln \left( \frac{V^2}{C} \right) + \frac{1}{2} \ln \left[ (\tilde{t} - \pi \tilde{s}_R \tilde{d}_R)^2 + (\pi d_0)^2 \right] - \frac{(\tilde{t} - \pi \tilde{s}_R \tilde{d}_R)}{\pi d_0} \tan^{-1} \left( \frac{\tilde{t} - \pi \tilde{s}_R \tilde{d}_R}{\pi d_0} \right)$$

(E.13)

with

$$\ln \left( \frac{1}{C} \right) = \ln(2) - 1 + \int_{-1/2}^{0} d\omega \frac{D_0(\omega) - d_0}{d\omega}$$

(E.14)
case of half band-filling with Hubbard interaction given by eq. (8). We study the electrons in the impurity orbitals interact via the amplitude between the host-electron orbitals is impurity orbital on each site. The electron transfer only two sites with one host-electron orbital and one half band-filling, we address an exactly solvable four-orbital model at the Anderson model, eq. (E.5).

Appendix F. Four-orbital toy model

For a simple illustration of a central result of this work, we address an exactly solvable four-orbital model at particle-hole symmetry in agreement with the result for the single-impurity ground-state energy $\Delta$.

Figure F1. Ground-state energy $\Delta E(U, V) = E_0(U, V) + W + U/2$ scaled by its limiting large-$U$ behavior as a function of $U$ for $V = 0.6, 0.4, 0.2, 0.02$. Apparently, $\Delta E(U, V) \sim -4V^2/U$.

In Fig. F1 we show the ground-state energy as a function of $U$ for $V = 0.6, 0.4, 0.2, 0.02$. The $V$-dependent energy correction is given by $\Delta E(U, V) = E_0(U, V) + W + U/2$, where $E_0(U, V)$ is the ground-state energy. For large values $U/V$ we see that $\Delta E(U, V) \sim -4V^2/U$, as in our variational description, eq. (F.3). This indicates that there is an effective direct electron transfer between the impurity orbitals of the order $V$ in the spin limit, see eq. (E.15).

To elucidate the properties of the ground state further, we consider the probability to find a spin singlet formed between the impurities,

$$P_s^{\text{imp}}(U, V) = \langle \Psi_0 | \frac{1}{4} - \mathbf{S}_1 \cdot \mathbf{S}_2 | \Psi_0 \rangle,$$  \hspace{1cm} (F.3)

and the probability to find a spin singlet formed between an impurity state and its local host electron state (‘Kondo singlet’),

$$P_s^{K}(U, V) = \langle \Psi_0 | \frac{1}{4} - \mathbf{s}_1 \cdot \mathbf{s}_l | \Psi_0 \rangle,$$  \hspace{1cm} (F.4)

which are equal for site $l = 1, 2$. In Fig. F2 we show both quantities as a function of $U$ for $V = 0.4$.

For small interactions, there is a tendency to form a Kondo spin singlet because $P_s^{K}$ initially increases as a function of $U$. However, $P_s^{K}$ starts to decrease for $U \gtrsim W$. Moreover, the probability to find a singlet formed by the two impurity spins dominates for $U \gtrsim W$, $P_s^{\text{imp}} > P_s^{K}$. Eventually, the impurity spins form a Heisenberg-type singlet pair.

Figure F2. Probabilities for a spin singlet between the impurity electrons, $P_s^{\text{imp}}(U, V)$, and between impurity and host electrons, $P_s^{K}(U, V)$, as a function of $U$ for $V = 0.4$.

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