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2010 J. Phys.: Condens. Matter 22 026002

(http://iopscience.iop.org/0953-8984/22/2/026002)

IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **22** (2010) 026002 (6pp) doi:10.1088/0953-8984/22/2/026002

Kondo screening in high-spin side-coupled two-impurity clusters

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Received 22 October 2009, in final form 11 November 2009
Published 9 December 2009
Online at stacks.iop.org/JPhysCM/22/026002

Abstract
We study the system of two magnetic impurities described by a two-impurity Kondo model where only the first impurity couples directly to the conduction band, while the second impurity interacts with the first through Heisenberg exchange coupling in a 'side-coupled' configuration. We consider various choices of the impurity spins ($S_1 < S_2$, $S_1 = S_2$ and $S_1 > S_2$) and we contrast the regimes where the inter-impurity exchange coupling J is either lower or higher than the Kondo temperature $T_K^{(0)}$ of the first impurity in the absence of the second. This model is a high-spin generalization of the two-impurity model for side-coupled double quantum dots which corresponds to the simplest $S_1 = S_2 = 1/2$ case, where the moments are Kondo-screened successively in two stages for $J < T_K^{(0)}$ (the 'two-stage Kondo effect'). We show that the two-stage Kondo screening occurs generically for $S_2 \geqslant S_1$. For $S_1 \geqslant 1$, the second Kondo temperature $T_K^{(2)}$ is not exponentially reduced, as for $S_1 = 1/2$, but is approximately a power-law function of the coupling J. Furthermore, for $S_1 \geqslant 1$ all three scales ($T_K^{(0)}$, J, $T_K^{(2)}$) explicitly appear in the temperature dependence of the thermodynamic properties. For $S_1 > S_2$, there is no second stage of screening for $J < T_K^{(0)}$. However, in the opposite limit $J > T_K^{(0)}$ the Kondo screening of the effective spin $S_1 - S_2$ is found.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

When magnetic impurities, such as substitutional defects in the bulk or adatoms on the surface, couple with the conduction band electrons through an antiferromagnetic exchange interaction, their spin is screened in the Kondo effect and the level degeneracy is effectively lifted [1–7]. When the separation between two such impurities is small, the impurities interact through the RKKY interaction [8], which may lead to critical behaviour in some parameter regimes [9, 10]. A simplified description of such systems is the two-impurity Kondo model [11–16]: the two magnetic atoms are represented by quantum spin operators which are coupled by some exchange interaction J, and each furthermore interacts with the conduction band electrons through an effective Kondo exchange coupling. With few exceptions [17, 18], most studies of such models focus on spin-1/2 impurities, while real impurities may actually have higher spins [4]. The same may also be the case in artificial atoms, i.e. quantum dots [19], and in molecules with embedded magnetic ions [20]. Due to competing interactions and the vastness of the parameter space, a great variety of different types of magnetic behaviour are expected. In this work we discuss a sub-class of high-spin two-impurity models in which only one of the spins (S_1) couples to the conduction band, while the second spin (S_2) is 'side-coupled' to the first one. Only the $S_1 = S_2 = 1/2$ limit of this family has been studied so far [18] and some results are known for the case of $S_1 = 1/2$ and arbitrary S_2 in the related Anderson–Kondo model [21]. It is shown that the two-stage Kondo screening [18, 22, 23] found in the $S_1 = S_2 = 1/2$ model is a generic feature of all $S_2 \geqslant S_1$ models, although for $S_1 \geqslant 1$ some qualitative differences arise.

2. Model and method

We consider the two-impurity Kondo model:

$$H = \sum_{k\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + J_K \mathbf{s} \cdot \mathbf{S}_1 + J \mathbf{S}_1 \cdot \mathbf{S}_2. \quad (1)$$

Operators $c_{k\sigma}^{\dagger}$ create conduction band electrons with momentum k, spin $\sigma \in \{\uparrow, \downarrow\}$ and energy ϵ_k, while $\boldsymbol{s} = \{s_x, s_y, s_z\}$ is the spin density of the conduction band electrons at the position of the first impurity. Operators $\boldsymbol{S}_1 = \{S_{1,x}, S_{1,y}, S_{1,z}\}$ and $\boldsymbol{S}_2 = \{S_{2,x}, S_{2,y}, S_{2,z}\}$ are the quantum-mechanical impurity spin operators. Furthermore, J_K is the effective Kondo exchange coupling constant and J is the inter-impurity Heisenberg coupling constant. The models considered are the simplest generalization of the spin-1/2 two-impurity models for the side-coupled impurity configuration exhibiting the two-stage Kondo effect [18, 22–24], which can also be found in other multi-impurity problems [25, 26]. Two-stage Kondo screening occurs when the exchange coupling between the impurities is weaker than the energy scale of the Kondo screening of the directly coupled impurity. In such a situation, the moment on the first impurity is Kondo-screened at the Kondo temperature, which corresponds to a decoupled impurity, $T_K^{(0)}$, while the moment on the second is Kondo-screened at some exponentially reduced temperature $T_K^{(2)}$. A simple interpretation is that the second Kondo effect occurs due to exchange coupling between the side-coupled impurity and the Fermi liquid of heavy quasiparticles resulting from the first stage of Kondo screening [22].

We solve the Hamiltonian using the numerical renormalization group (NRG) method [27–29]. In this approach, the continuum of the conduction band electron states is discretized logarithmically with increasingly narrow intervals in the vicinity of the Fermi level. The problem is transformed (tridiagonalized) to the form of a tight-binding Hamiltonian with exponentially decreasing hopping constant which is then diagonalized iteratively. A very effective technique to examine the magnetic behaviour of an impurity model consists in studying the thermodynamic properties of the model as a function of the temperature [27, 28, 30]; the impurity contribution to the entropy, S_{imp}, then provides information on the degeneracy of the effective spin multiplets and the impurity contribution to the magnetic susceptibility, χ_{imp}, defines the effective magnetic moment.

The results reported in this work have been calculated with the discretization parameter $\Lambda = 2$ using improved discretization schemes [31, 32], without the z averaging, and with the NRG truncation cutoff set at $7\omega_N$, where ω_N is the characteristic energy scale at the Nth step of the iteration.

3. Properties of two antiferromagnetically coupled isotropic spins

We first briefly review some properties of a decoupled pair of magnetic impurities described as pure spins with spin quantum numbers S_1 and S_2; without loss of generality, in this section we use the convention that $S_2 \geqslant S_1$. The results will be relevant in the discussion of the $J \to \infty$ limit.

The impurities couple antiferromagnetically into an effective $S = S_2 - S_1$ spin object. The inner product of spin operators is given as

$$\langle \boldsymbol{S}_1 \cdot \boldsymbol{S}_2 \rangle = -S_1(S_2+1). \tag{2}$$

Table 1. Multiplicative factors which determine the effective Kondo exchange coupling of the composite object made of the two spins locked into a $S = S_2 - S_1$ antiferromagnetically aligned state.

S_1	S_2	r_1	r_2
1/2	1	−1/3	4/3
1/2	3/2	−1/4	5/4
1/2	2	−1/5	6/5
1	3/2	−2/3	5/3
1	2	−1/2	3/2
3/2	2	−1	2

The ground state multiplet can be expressed using the Clebsch–Gordan coefficients as

$$|S, M\rangle = \sum_{m_1,m_2} \langle S_1, m_1, S_2, m_2 | S, M\rangle \times |S_1, m_1\rangle \otimes |S_2, m_2\rangle. \tag{3}$$

The projections of the spin-S object on the constituent spin operators can be easily computed as the expectation values of the $S_{z,i}$ operators in the maximum weight states $|S, S\rangle$:

$$p_1 = \langle S, S|S_{z,1}|S, S\rangle \tag{4}$$

$$= \sum_{m_1} |\langle S_1, m_1, S_2, S_2 - S_1 - m_1 | S_2 - S_1, S_2 - S_1\rangle|^2 m_1 \tag{5}$$

$$= \frac{S_1(S_2 - S_1)}{S_1 - S_2 - 1}, \tag{6}$$

and

$$p_2 = \langle S, S|S_{z,2}|S, S\rangle \tag{7}$$

$$= \sum_{m_2} |\langle S_1, m_1, S_2, S_2 - S_1 - m_1 | S_2 - S_1, S_2 - S_1\rangle|^2 \times (S_2 - S_1 - m_1) \tag{8}$$

$$= \frac{(S_1 - S_2)(1 + S_2)}{S_1 - S_2 - 1}. \tag{9}$$

If the Hamiltonian describing the coupling of the impurities with the host conduction band is of the form

$$H_C = J_1 \boldsymbol{S}_1 \cdot \boldsymbol{s} + J_2 \boldsymbol{S}_2 \cdot \boldsymbol{s}, \tag{10}$$

then the coupling of the effective spin takes the following form:

$$H_C^{\text{eff}} = J_{\text{eff}} \boldsymbol{S} \cdot \boldsymbol{s}, \tag{11}$$

with

$$J_{\text{eff}} = r_1 J_1 + r_2 J_2, \tag{12}$$

where $r_i = p_i/(S_2 - S_1)$. The ratios r_i are thus the multiplicative factors which determine the effective Kondo exchange coupling of the composite object; they are tabulated in table 1. Note that the sign of r_1 is always negative, while the sign of r_2 is always positive. This implies that, in the side-coupled configuration discussed in this work, the Kondo screening of the effective spin in the $J \to \infty$ limit occurs only if the impurity which couples to the conduction band is the one with larger spin; in this case the impurity ground state multiplet will have spin $|S_2 - S_1| - 1/2$. In the opposite case, the exchange coupling to the conduction band is ferromagnetic and the ground state multiplet will have spin $|S_2 - S_1|$.

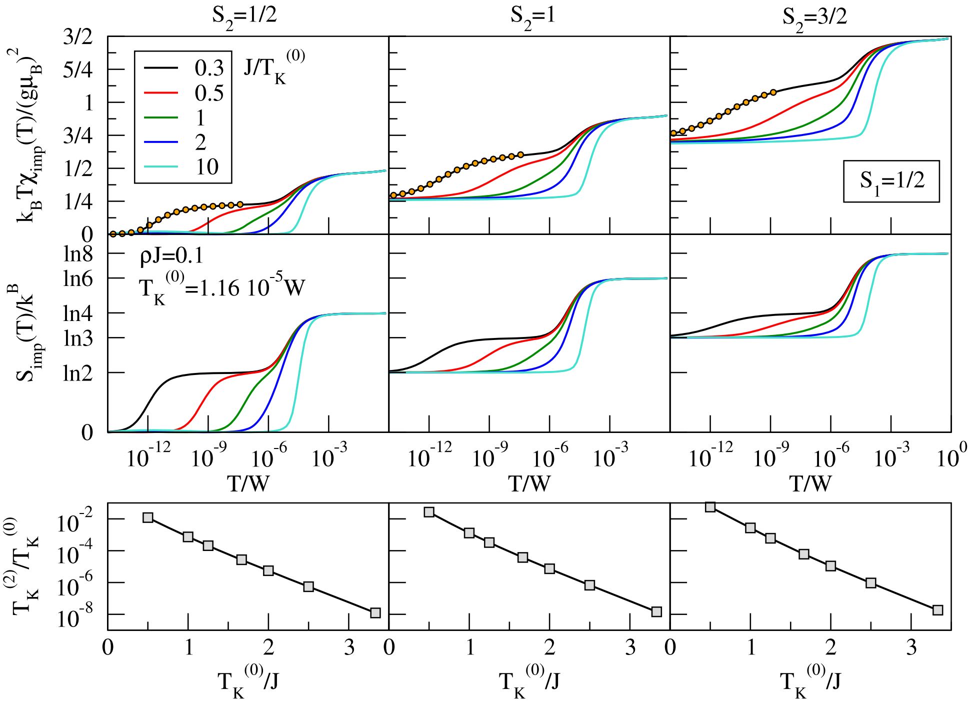

Figure 1. Thermodynamic properties of the isotropic two-impurity clusters with $S_1 = 1/2$. We plot the impurity contribution to the magnetic susceptibility, χ_{imp}, and the impurity contribution to the entropy, S_{imp}. The symbols indicate fits to the universal spin-S_2 Kondo magnetic susceptibility curves, which were used to extract the J dependence of the second Kondo temperature, displayed in the bottom line of panels.

4. Results

We fix $\rho J_{\mathrm{K}} = 0.1$ throughout this work. The Kondo temperature of a decoupled impurity is thus $T_{\mathrm{K}}^{(0)} = 1.16 \times 10^{-5} W$ (Wilson's definition) and it is the same for any value of spin S_1 [33–35].

The thermodynamic properties of the system for the case when the first spin is $S_1 = 1/2$ are shown in figure 1. For $S_2 = 1/2$ we recover exactly the prototypical two-stage Kondo screening where the second Kondo temperature is given by [18, 22]

$$T_{\mathrm{K}}^{(2)} = c_1 T_{\mathrm{K}}^{(1)} \mathrm{e}^{-c_2 \frac{T_{\mathrm{K}}^{(1)}}{J}}, \tag{13}$$

where c_1 and c_2 are some numerical constants; the scaling of $T_{\mathrm{K}}^{(2)}$ with $T_{\mathrm{K}}^{(0)}/J$ is shown in the subfigure in the bottom panel.

For $S_2 \geqslant 1$, we observe very similar behaviour: for $J < T_{\mathrm{K}}^{(0)}$, after the initial screening of the first impurity, the second impurity undergoes spin-S_2 Kondo screening which reduces its spin by one half-unit, as if it were coupled directly to the conduction band. The only effect of the first impurity is thus to induce a much lower effective bandwidth $D_{\mathrm{eff}} \propto T_{\mathrm{K}}^{(0)}$ and increased density of states $1/\rho_{\mathrm{eff}} \propto T_{\mathrm{K}}^{(0)}$. This picture is confirmed by the scaling of $T_{\mathrm{K}}^{(2)}$ with $T_{\mathrm{K}}^{(0)}/J$, which is very similar for all three values of S_2 in figure 1.

We now consider the case when the first spin is $S_1 = 1$. We remind the reader that, when a single impurity with spin $S \geqslant 1$ couples to the conduction band, one half-unit of the spin is screened in a spin-S Kondo effect, giving rise to a spin $S - 1/2$ composite object which couples with the conduction band electrons with a ferromagnetic effective exchange coupling. Thus it remains unscreened in the ground state [36–40, 34, 41]. The situation becomes more involved in the presence of an additional impurity, see figure 2.

For $S_1 = 1$ and $S_2 = 1/2$, in the limit $J \gg T_{\mathrm{K}}^{(0)}$, the two spins couple into a spin-1/2 object which couples to the conduction band with an antiferromagnetic effective Kondo exchange coupling $J_{\mathrm{eff}} = r_1 J$ with $r_1 = 4/3$ (see table 1) and undergoes the usual spin-1/2 Kondo effect, which results in fully compensated impurity spins and a non-degenerate singlet ground state. The expression for J_{eff} holds strictly only when J is much larger than any other scale in the problem (in particular J_{K} and the bandwidth W); for $J \approx 60 J_{\mathrm{K}}$ we indeed find that the Kondo temperature is $T_{\mathrm{K}} = 1.7 \times 10^{-4} W$, which agrees with the expected scale of $T_{\mathrm{K}} \approx W\sqrt{\rho J(4/3)} \exp\{-1/[\rho J(4/3)]\} \approx 2 \times 10^{-4} W$. For lower J, the effective bandwidth is of the order of J rather than W and thus the Kondo temperature is reduced accordingly. An example of such behaviour is shown in figure 2 for $J/T_{\mathrm{K}}^{(0)} = 100$. The initial free-spin $\ln 6$ entropy is reduced to $\ln 2$ at $T \sim J$ upon formation of the effective composite spin. This is followed by the conventional spin-1/2 Kondo screening (see the fit of the magnetic susceptibility with the universal Kondo curves).

In the opposite limit $J \ll T_{\mathrm{K}}^{(0)}$ we observe the initial spin-1 Kondo screening of the first spin: the magnetic susceptibility goes toward 1/2 and the entropy toward $2 \ln 2$. The screening is, however, abruptly interrupted at $T \sim J$. This can be interpreted as the formation of a spin-singlet object composed from the residual spin-1/2 resulting from the Kondo screening and the side-coupled spin-1/2 impurity. The end result is the

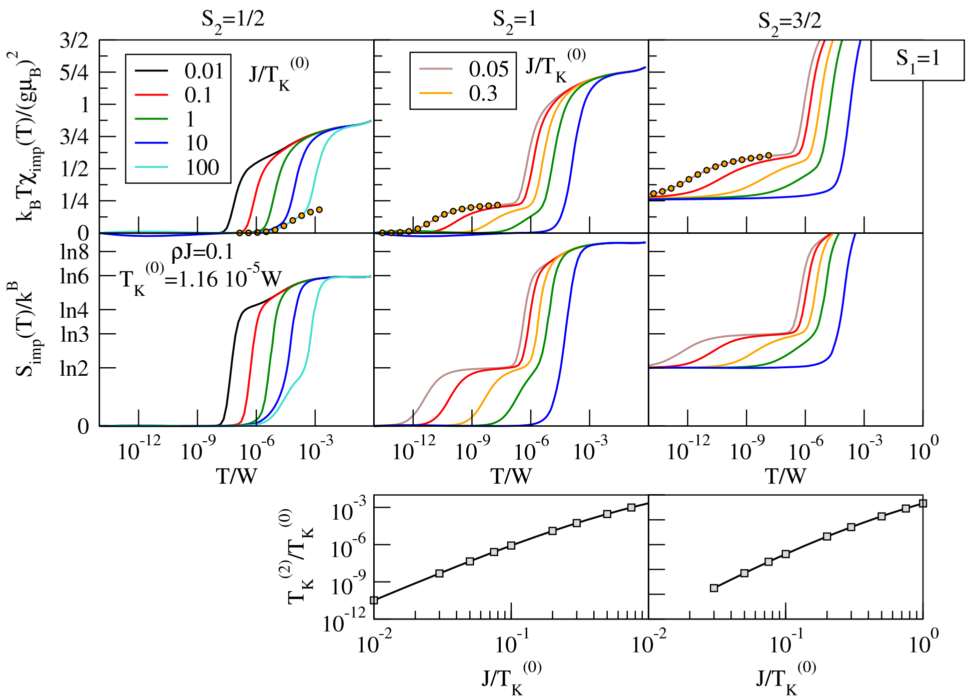

Figure 2. Thermodynamic properties of the two-impurity clusters with $S_1 = 1$. For $S_2 = 1$ and $S_2 = 3/2$, the scaling of the second Kondo temperature with J is shown in the bottom panels; it should be noted that the horizontal axis here corresponds to $J/T_K^{(0)}$ and that the scale is logarithmic, while in figure 1 the axis corresponded to the inverse $T_K^{(0)}/J$, while the scale was linear. The symbols for $S_2 = 1/2$ and 1 correspond to fits using the universal spin-1/2 Kondo magnetic susceptibility, while the symbols for $S_2 = 3/2$ correspond to a fit using the universal spin-1 Kondo magnetic susceptibility.

same in both large-J and small-J limits; the crossover between the two is smooth as a function of J.

For $S_1 = 1$ and $S_2 = 1$, the behaviour for $J \gg T_K^{(0)}$ is particularly simple, since the two spins bind at the temperature $T \sim J$ into a singlet and they no longer play any role. For $J \ll T_K^{(0)}$ the Kondo screening of the $S_1 = 1$ spin into a residual spin-1/2 is interrupted at the temperature $T \sim J$. The residual spin-1/2 then binds with the side-coupled $S_2 = 1$ into a new spin-1/2 composite object. Unlike the residual spin-1/2 resulting from the incomplete screening of a spin-1 Kondo impurity, which remains uncompensated since it couples to the conduction band ferromagnetically, the spin-1/2 composite object that emerges in this case couples with the conduction band antiferromagnetically. Thus at some lower temperature which we again denote $T_K^{(2)}$ it is compensated in a spin-1/2 Kondo effect. This thus constitutes a non-trivial generalization of the two-stage Kondo screening phenomenology encountered in the $S_1 = 1/2$ cases. The differences, however, are notable: (1) there are not two, but three, energy scales: $T_K^{(0)}$, where the spin-1 Kondo screening takes place, J, where this screening is abruptly interrupted, and $T_K^{(2)}$, where the second Kondo screening occurs; and (2) the scaling of the second Kondo temperature $T_K^{(2)}$ is not exponential with $1/J$. In the conventional two-stage Kondo effect with $S_1 = 1/2$, the only role of the coupling J is to set the lower Kondo temperature; no feature is observed there in the thermodynamic properties of the system at $T \sim J$. Here, the coupling J is essential to produce a composite spin object which then couples antiferromagnetically with the rest of the system. Thus this scale is directly observable as a sharp change in the effective impurity degrees of freedom at $T \sim J$. The second Kondo temperature is defined by a power law with the exponent near 3, with some corrections (see the lower panels in figure 2).

For $S_1 = 1$ and $S_2 = 3/2$ the results for $J \gg T_K^{(0)}$ are trivial: at the temperature $T \sim J$, the spins lock into a spin-1/2 object which couples ferromagnetically with the conduction band; thus the composite spin remains unscreened. This is in accord with the expected behaviour in this limit (see section 3). For $J \ll T_K^{(0)}$ the results are, however, very intriguing: the Kondo screening of the $S_1 = 1$ spin is interrupted at the temperature $T \sim J$. At this point, the residual spin-1/2 couples antiferromagnetically with the $S_2 = 3/2$ spin into a spin-1 composite object. This composite object, interestingly, couples antiferromagnetically with the conduction band electrons, which leads to Kondo screening of one half-unit of spin at some lower temperature which we denote, yet again, as $T_K^{(2)}$. The final residual spin-1/2 is not compensated, since it couples ferromagnetically with the conduction band. Thus we again observe a two-stage Kondo effect of the same universality class as in the $S_1 = S_2 = 1$ case. This result may, in fact, be generalized: for any $S_2 \geqslant S_1$, the impurity spins will be compensated for in two screening stages (the compensation is only partial for $S_2 \neq S_1$).

To further substantiate the claim that the results are generic, we show in figure 3 the results for the $S_1 = 3/2$ case. For $S_2 = 1/2$ and 1, i.e. for $S_2 < S_1$, we again

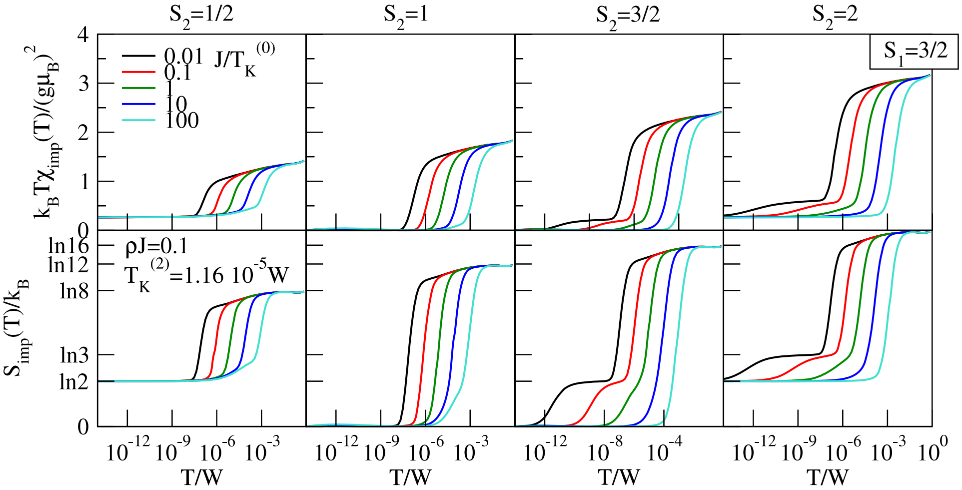

Figure 3. Thermodynamic properties of the two-impurity clusters with $S_1 = 3/2$. For $S_2 = 1/2$, the Kondo screening in the large-J limit is of the spin-1 type, while for $S_2 = 1$ the Kondo screening in the same limit is of the spin-1/2 type. For $S_2 = 3/2$, the second-stage Kondo screening in the small-J limit is of the spin-1/2 type, while for $S_2 = 2$ it is of the spin-1 type.

find the emergence of the Kondo screening of the rigidly antiferromagnetically bound $S_2 - S_1$ spin in the large-J limit and the formation of a $S = (S_2 - 1/2) - S_1$ bound state at $T \sim J$ in the small-J limit. Furthermore, in the case of $S_2 \geqslant S_1$, the two-stage Kondo screening is again observed in the small-J limit, again with the power-law dependence of $T_K^{(2)}$ on J.

5. Discussion and conclusion

We have shown that, when a second impurity is side-coupled to a Kondo impurity with sufficiently small Heisenberg coupling J, the spin will be screened in two stages for all systems, where the spin of the side-coupled impurity S_2 is equal to or greater than the spin of the directly coupled one, S_1. When $S_1 = 1/2$, the second Kondo temperature is exponentially reduced, while for $S_1 \geqslant 1$, it is a power-law function of the coupling J. The difference stems from the fact that, for $S_1 = 1/2$, the second stage of the Kondo screening occurs with a *local* spin S_2 which interacts with a Fermi liquid of heavy electrons resulting from the first screening stage, while for $S_1 \geqslant 1$ the first screening stage leaves behind a residual uncompensated spin $S_1 - 1/2$, which is an *extended* object. This residual spin then rigidly binds with the spin of the side-coupled impurity at the temperature scale of $T \approx J$ to produce a new *extended* spin object which then undergoes Kondo screening. Similar behaviour is found in the anisotropic single-impurity Kondo model, where an easy-plane anisotropy leads to a formation of an extended effective spin-1/2 degree of freedom which is Kondo-screened [35]; in this problem, the second Kondo temperature is a power-law function of the longitudinal magnetic anisotropy constant D. No theory has been devised yet to map this class of problems with effective extended spin degrees of freedom onto the conventional Kondo model with a localized spin operator. Thus there is at present no analytical account of these power-law dependences. Nevertheless, it is clear that a power-law dependence of the second Kondo temperature, i.e. $T_K^{(2)} \propto \exp(-1/\rho_{\mathrm{eff}} J_{\mathrm{eff}}) \propto J^{\alpha}$, implies an inverse logarithmic dependence of the effective impurity parameters, i.e. $\rho_{\mathrm{eff}} J_{\mathrm{eff}} \propto -1/\ln J$. This form is suggestive of the energy dependence of the renormalized ferromagnetic exchange coupling of the residual spin in the underscreened Kondo model, $\tilde{J}(\omega) = 1/\ln(\omega/T_0)$, where T_0 is some low energy scale [39]. This indicates that the ferromagnetic residual coupling might play a decisive role in determining the total effective antiferromagnetic exchange coupling of the composite spin object. In this scenario, the bare parameter J leads to the emergence of the composite spin object by antiferromagnetic binding of the residual spin with the side-coupled impurity spin, which occurs on the energy scale of $\omega = J$, while the coupling of this object with the surrounding electron liquid is controlled solely by $\tilde{J}(\omega = J) = 1/\ln(J/T_0)$. This may be explained by the fact that the side-coupled impurity interacts with the electron liquid only indirectly through the first impurity and that the sign of the relevant multiplicative factor r_1 is always negative (see table 1). Thus the sign of the effective exchange interaction is flipped.

In conclusion it may also be remarked that a common feature of all models considered in this work is that the ground state in no way depends on the $J/T_K^{(0)}$ ratio; for any non-zero Heisenberg coupling between the impurities we always end up in the same fixed point, only the temperature dependence of the spin compensation differs greatly. This no longer holds for problems with additional magnetic anisotropy terms (i.e. two-impurity extensions of models studied in [35, 42]), where level crossings may also occur as a function of J. This behaviour will be addressed in future works.

Acknowledgment

The author acknowledges the support of the Slovenian Research Agency (ARRS) under grant no. Z1-2058.

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