

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

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**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  $S_2 \geq S_1$ . For  $S_1 \geq 1$ , 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  $S_1 \geq 1$  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.

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## 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, 2, 3, 4, 5, 6, 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, 12, 13, 14, 15, 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 spin [4]. The same may also be the case in artificial atoms, i.e. quantum dots [19], and in molecules with embedded magnetic ion [20]. Due to competing interactions and the vastness of the parameter space, a great variety of different types of magnetic behavior 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, 21, 22, 23]. It is shown that the two-stage Kondo screening [18] found in this particular model is a generic feature of all  $S_2 \geq S_1$  models, although for  $S_1 \geq 1$  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  $\epsilon_k$ , while  $\mathbf{s} = \{s_x, s_y, s_z\}$  is the spin density of the conduction-band electrons at the position of the first impurity. Operators  $\mathbf{S}_1 = \{S_{1,x}, S_{1,y}, S_{1,z}\}$  and  $\mathbf{S}_2 = \{S_{2,x}, S_{2,y}, S_{2,z}\}$  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, 21, 22], which can also be found in other multi-impurity problems [24, 25]. 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 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 the Kondo screening [21].

We solve the Hamiltonian using the numerical renormalization group (NRG) method [26, 27, 28]. 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 behavior of an impurity model consists in studying the thermodynamic properties of the model as a function of the temperature [26, 27, 29]; the impurity contribution to the entropy,  $S_{\text{imp}}$ , then provides information on the degeneracy of the effective spin multiplets, and the impurity contribution to the magnetic susceptibility,  $\chi_{\text{imp}}$ , defines the effective magnetic moment.

The results reported in this work have been calculated with the discretization parameter  $\Lambda = 2$  using improved discretization schemes [30, 31], without the  $z$ -averaging, and with the NRG truncation cutoff set at  $7\omega_N$ , where  $\omega_N$  is the characteristic energy scale at the  $N$ -th 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  $S_2 \geq S_1$ . The results will be relevant in the discussion of the  $J \rightarrow \infty$  limit.

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

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

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

$$|S, M\rangle = \sum_{m_1, m_2} \langle S_1, m_1, S_2, m_2 | S, M \rangle |S_1, m_1\rangle \otimes |S_2, m_2\rangle. \quad (3)$$

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  $|S, S\rangle$ :

$$p_1 = \langle S, S | S_{z,1} | S, S \rangle \quad (4)$$

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

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

and

$$p_2 = \langle S, S | S_{z,2} | S, S \rangle \quad (7)$$

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

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

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

$$H_C = J_1 \mathbf{S}_1 \cdot \mathbf{s} + J_2 \mathbf{S}_2 \cdot \mathbf{s}, \quad (10)$$

then the coupling of the effective spin takes the following form

$$H_C^{\text{eff}} = J_{\text{eff}} \mathbf{S} \cdot \mathbf{s}, \quad (11)$$

with

$$J_{\text{eff}} = r_1 J_1 + r_2 J_2, \quad (12)$$

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 signs 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  $J \rightarrow \infty$  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 multiple will have spin  $|S_2 - S_1|$ .

**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

## 4. Results

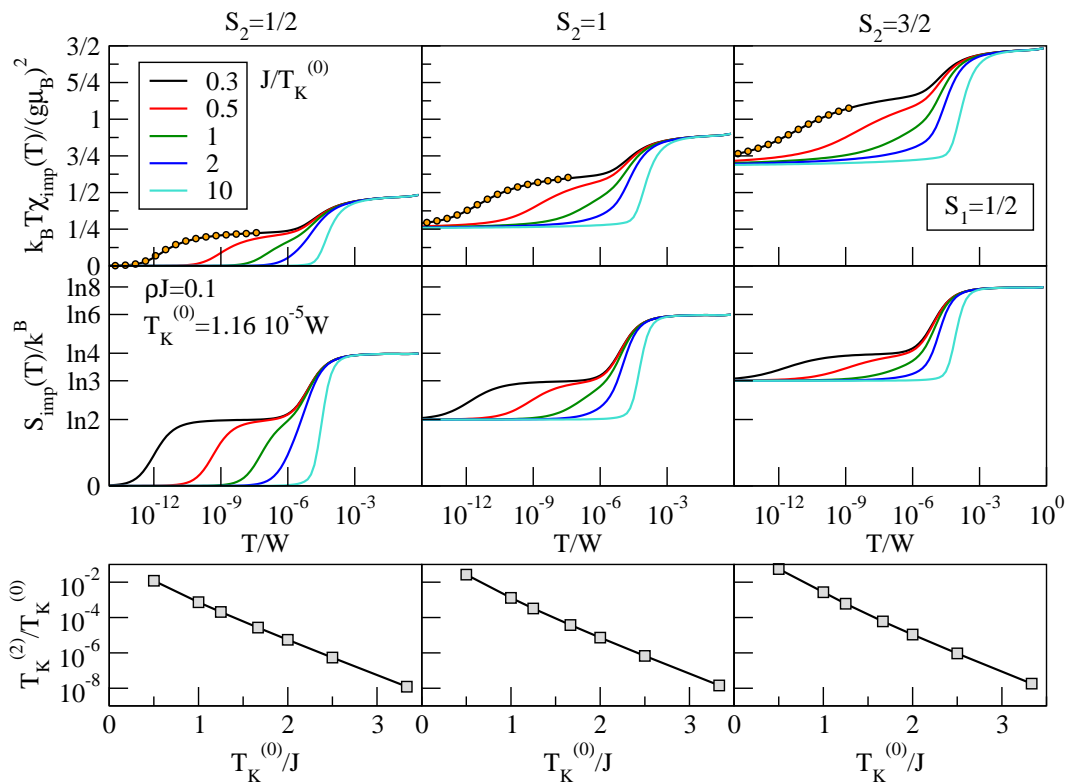
We fix  $\rho J_K = 0.1$  throughout this work. The Kondo temperature of a decoupled impurity is thus  $T_K^{(0)} = 1.16 \times 10^{-5} W$  (Wilson's definition) and it is the same for any value of spin  $S_1$  [32, 33, 34].

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, 21]

$$T_K^{(2)} = c_1 T_K^{(1)} e^{-c_2 \frac{T_K^{(1)}}{J}}, \quad (13)$$

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

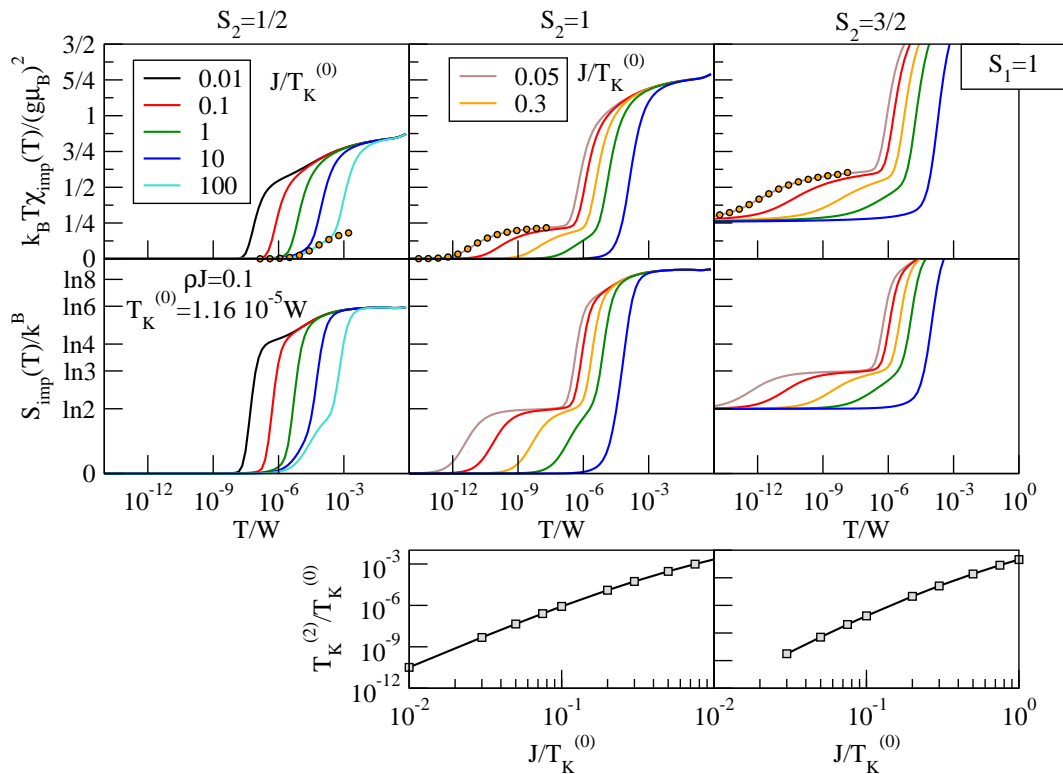
For  $S_2 \geq 1$ , we observe very similar behaviour: for  $J < T_K^{(0)}$ , after the initial screening of the first impurity, the second impurity undergoes spin- $S_2$  Kondo screening which reduces its spin by one half-unit, as if it were coupled directly to the conduction band. The only effect of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

We now consider the case when the first spin is  $S_1 = 1$ . We remind the reader that when a single impurity with spin  $S \geq 1$  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 [35, 36, 37, 38, 39, 33, 40]. The situation becomes more involved in the presence of an additional impurity, see Figure 2.



**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  $S_2 = 1$  corresponds 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.

For  $S_1 = 1$  and  $S_2 = 1/2$ , in the limit  $J \gg T_K^{(0)}$  the two spins couple into a spin-1/2 object which couples to the conduction band with an antiferromagnetic effective Kondo exchange coupling  $J_{\text{eff}} = r_1 J$  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  $J_{\text{eff}}$  holds strictly only when  $J$  is much larger than any other scale in the problem (in particular  $J_K$  and the band-width  $W$ ); for  $J \approx 60J_K$  we indeed find that the Kondo temperature is  $T_K = 1.7 \times 10^{-4}W$  which agrees with the expected scale of  $T_K \approx W \sqrt{\rho J(4/3)} \exp\{-1/[\rho J(4/3)]\} \approx 2 \times 10^{-4}W$ . For lower  $J$ , the effective band-width is of order  $J$  rather than  $W$ , thus the Kondo temperature is reduced accordingly. An example of such behaviour is shown in Figure 2

for  $J/T_K^{(0)} = 100$ . 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).

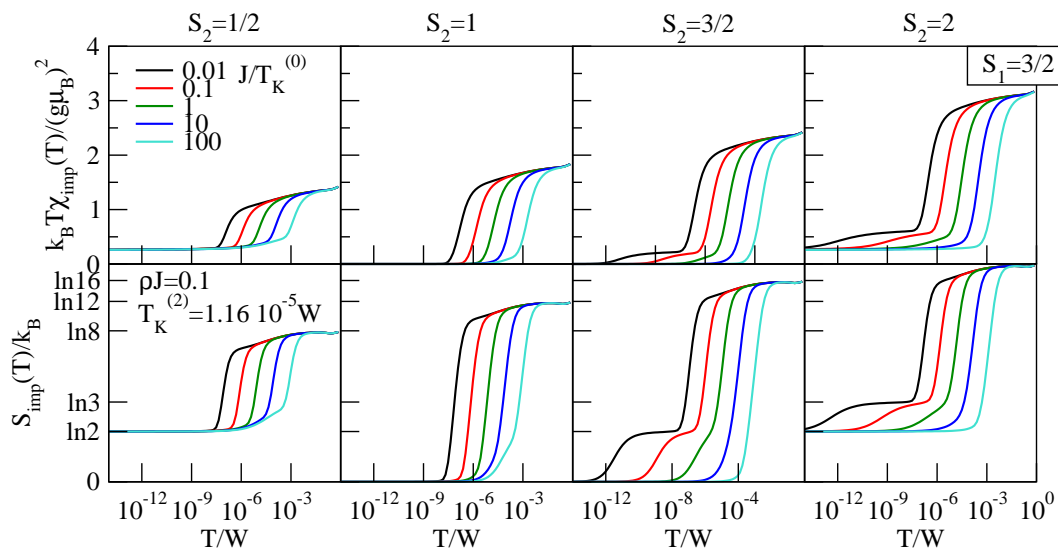
In the opposite limit  $J \ll T_K^{(0)}$  we observe the initial spin-1 Kondo screening of the first spin: the magnetic susceptibility goes toward 1/2 and the entropy toward  $2 \ln 2$ . The screening is, however, abruptly interrupted at  $T \sim J$ . This can be interpreted as the formation of a spin-singlet 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 same in both large- $J$  and small- $J$  limits; the cross-over 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; 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 behavior 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  $S_2 \geq S_1$ , the impurity spins will be compensated 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  $S_2 = 1$ , i.e. for  $S_2 < S_1$  we again 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  $S_2 \geq S_1$ , the two-stage Kondo screening is again observed in the small- $J$  limit, again with power-law dependence of  $T_K^{(2)}$  on  $J$ .



**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.

## 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 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  $S_1 \geq 1$ , 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  $S_1 \geq 1$  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 [34]; 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 presently no analytical account of these power-law dependences.

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 Refs. [34, 41]), where level crossings may also occur as a function of  $J$ . This behavior will be addressed in future works.

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