Quantum phase transitions in models of coupled magnetic impurities

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We discuss models of interacting magnetic impurities coupled to a metallic host, which show one or more boundary quantum phase transitions where the ground-state spin changes as a function of the inter-impurity couplings. The simplest example is realized by two spin-1/2 Kondo impurities coupled to a single orbital of the host. We investigate the phase diagram and crossover behavior of this model and present Numerical Renormalization Group results together with general arguments showing that the singlet-doublet quantum phase transition is either of first order or of the Kosterlitz-Thouless type, depending on the symmetry of the Kondo couplings. Thus we find an exponentially small energy scale within the Kondo regime, and a two-stage Kondo effect, in a single-channel situation. Connections to other models and possible applications are discussed.

The physics of a single magnetic impurity embedded in a metal — known as the Kondo effect — is a well-studied phenomenon in many-body physics. The low-energy physics is completely determined by a single energy scale, the Kondo temperature $T_K$, and the impurity spin is fully quenched in the low-temperature limit, $T \ll T_K$.

Systems with more than one impurity are considerably more complicated because the interaction between impurity spins competes with Kondo screening. This inter-impurity interaction, which can lead to a magnetic ordering transition in lattice models, arises both from direct exchange coupling $I$, which we explicitly include in the following discussion, and from the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction mediated by the conduction electrons.

The simplest systems containing this competition between Kondo effect and inter-impurity interaction are two-impurity models which have been extensively studied in recent years. Generically, two different regimes are possible as function of the inter-impurity exchange $I$: for large antiferromagnetic $I$ the impurities combine to a singlet, and the interaction with the conduction band is weak, whereas for ferromagnetic $I$ the impurity spins add up and are Kondo-screened by conduction electrons in the low-temperature limit.

For two spin-1/2 impurities, each coupled to one channel of conduction electrons (total number of two screening channels $K=2$), there is perfect screening as $T \to 0$ in both regimes and the ground-state spin will be zero. It has been shown that there is no quantum phase transition as $I$ is varied in this two-impurity Kondo model in the generic situation without particle-hole symmetry (whereas one finds an unstable non-Fermi liquid fixed point in the particle-hole symmetric case). However, the finite-$T$ crossover behavior is significantly different in the two regimes: for large antiferromagnetic $I$, the singlet is formed at an energy scale of order $I$. In contrast, for ferromagnetic $I$ where both spins have to be quenched by the Kondo effect, the two screening channels are formed by the even and odd linear combinations of the conduction electrons at both impurity sites. These two channels have generically different densities of states leading to two different Kondo temperatures. Upon lowering the temperature the two spin-1/2 degrees of freedom are “frozen” at these two distinct temperature scales (two-stage Kondo effect).

The purpose of this paper is to discuss models of coupled impurities which can be tuned between regimes with different ground state spin. The spin quantization then requires that these regimes are separated by one or more boundary quantum phase transitions as the inter-impurity couplings are varied. Phases with a non-zero ground state spin are obtained in the situation of underscreening, i.e., if a spin of size $S$ interacts with $K$ conduction electron channels, and $2S > K$, leading to a residual free spin of size $(S-K/2)$ as $T \to 0$. A general two-impurity Kondo model necessarily showing a quantum phase transition then consists of two spins of size $S_1$ and $S_2$, with a direct exchange coupling $I$, and a total of $K$ screening channels, satisfying $2(S_1+S_2) > K$.

Related Kondo- and Anderson models have been discussed recently in the context of multi-level or coupled quantum dots with nearly degenerate singlet and triplet levels. Several authors have considered effective two-impurity models near the singlet-triplet degeneracy with two screening channels which arise in vertical multi-level dots; such a situation corresponds to the physics of the usual two-impurity Kondo model not showing a phase transition in the absence of particle-hole symmetry. The case of a single screening channel and the associated limiting situations of a singlet ground state and an underscreened Kondo model have been mentioned in Refs., but the nature of the transition and the associated quantum-critical behavior have not been examined to date.

We note that a two-impurity model with $2(S_1+S_2) < K$ can give rise to overscreening in a channel-symmetric situation. Overscreening leads to a non-trivial intermediate coupling fixed point already for a single impurity and is a very interesting topic on its own, which shall, however, not be discussed here. Remarkably, the two-impurity two-channel Kondo model ($K=4$), has been
solved exactly by Georges and Sengupta, and shows a continuous line of non-Fermi liquid fixed points.

**Two spins 1/2, one channel.** The simplest realization of the above proposal is a “double Kondo impurity” (Fig. 1), i.e., a Kondo model of two spins 1/2 coupled to a single orbital of conduction electrons, with the Hamiltonian

\[
H_{\text{int}} = J_1 S_1 \cdot s_0 + J_2 S_2 \cdot s_0 + IS_1 \cdot S_2
\]

(1)

and \(H_{\text{band}} = \sum_{kk} \epsilon_k c^\dagger_k c_k\) in standard notation, \(s_0 = \sum_{\alpha} \epsilon_\alpha \sigma_\alpha c^\dagger_\alpha c_\alpha\) is the conduction band spin operator at the impurity site \(r_0 = 0\), and \(S_{1,2}\) denote the impurity spin operators. The Kondo couplings \(J_1, J_2\) are taken to be antiferromagnetic. For the numerical calculations, we will use a conduction band with a constant density of states, \(\rho(\epsilon) = \rho_0 = 1/(2D)\) for \(|\epsilon| < D = 1\).

The inter-impurity coupling \(I\) obviously drives a quantum phase transition: For large antiferromagnetic \(I\), both impurities form a singlet with zero total spin, whereas large ferromagnetic \(I\) locks the impurities into an \(S = 1\) entity, which is underscreened by the single channel of conduction electrons, leading to a double-gate ground state with residual spin 1/2.

A second observation is that the symmetry w.r.t. to the two Kondo couplings plays a crucial role. Re-writing \(H_{\text{int}}\) in terms of bond boson operators \(s, t_{x,y,z}\) [1], with \(S_{1,2}^\dagger = (\pm t_\alpha t_\gamma - 3s^4) / 4\), one finds

\[
H_{\text{int}} = I (t^a \gamma^a t^\dagger - 3s^4) / 4 - (J_1 + J_2) i e_{\alpha \beta} \gamma^\dagger \gamma t^\dagger \gamma + (J_1 - J_2) s^\dagger (s^\dagger + s^\gamma h.c.).
\]

In the symmetric case, \(J_1 = J_2\), there is no mixing between impurity singlet and triplets, and the state formed by the inter-impurity singlet and a decoupled conduction band is an exact eigenstate of \(H\). The “rest” of the Hilbert space (triplet plus conduction band) forms an underscreened \(S = 1\) Kondo problem. Therefore, for \(J_1 = J_2\) the model shows a simple level crossing at a critical \(T_c\) which marks a first-order transition between the singlet and the underscreened doublet state.

Turning to the asymmetric case, \(J_1 > J_2\), we start with results obtained using the Numerical Renormalization Group (NRG) technique [2]. In Fig. 2 we display the temperature dependence of \(T_{\text{Ximp}}\) for fixed \(J_{1,2}\) and different values of \(I\), where \(\chi_{\text{imp}}\) is the total impurity contribution to the uniform susceptibility. Let us first discuss the case of large asymmetry, Fig. 2a. We can identify \(I_c\), with all curves for \(I > I_c\) leading to \(T_{\text{Ximp}} \to 0\) for \(T \to 0\) (singlet), whereas all curves for \(I \leq I_c\) show \(T_{\text{Ximp}} \to 1/4\), consistent with a residual spin 1/2. The topmost curves \((I < I_c)\) allow an identification of the Kondo temperature for the \(S = 1\) Kondo problem, here we have \(T_K \simeq 10^{-11}\) (small values of \(J_{1,2}\) have been chosen to illustrate the crossover behavior). The curves for \(I < I_c\) approach the fixed point very slowly, indicating an operator which is marginally irrelevant. For \(I\) values near \(I_c\) and \(T \gg T_K\), there is a plateau at \(T_{\text{Ximp}} \simeq 1/2\). For exchange couplings \(I\) larger than but close to \(I_c\), the system first flows to the fixed point with \(T_{\text{Ximp}} = 1/4\), and eventually crosses over to the singlet ground state (for \(T/D < 10^{-12}\) in Fig. 2b). The corresponding crossover scale \(T^*\) is found to depend exponentially on the distance to the critical point, \((I - I_c)\) – there is no power law which would characterize a second-order transition. Even for very small \((I - I_c)\) the NRG does not show any fixed point behavior which could be distinguished from one of the stable fixed points, i.e., there is no sign of a third, infrared unstable, fixed point which could possibly correspond to the critical point at \(I = I_c\).

We have analyzed the structure of the NRG fixed points, and found that for \(I > I_c\) the system consists of a singlet decoupled from the band \((S_1 \cdot S_2) = -3/4\), whereas for all other values including \(I_c\) there is one
Kondo-screened spin 1/2 and one residual decoupled spin 1/2. Close to \( I = I_c \), the NRG flow is identical to the one in the single-impurity Kondo model near the Kosterlitz-Thouless (KT) transition at \( J = 0 \). These results strongly suggest that the phase transition in the double-impurity model is of KT type, and the critical point is characterized by one decoupled spin-1/2 degree of freedom.

In the following, we give heuristic scaling arguments supporting this picture. Close to the critical point, we can assume that the effective inter-impurity coupling, \( I_{\text{eff}} = I + I_{\text{RKKY}} \), is the smallest energy scale of the problem. In the spirit of poor man’s scaling, both Kondo couplings will grow upon reducing the band cut-off, and the initial asymmetry between \( J_1 \) and \( J_2 \) increases under this process. The larger coupling, \( J_1 \), will become of order unity at an energy scale \( T_K \sim D \exp[-D/(2J_1)] \), indicating that the spin \( S_1 \) becomes screened. This screening freezes the spin \( S_1 \) and the conduction electron spin \( s_0 \) into a singlet, thus terminating the flow of \( J_2 \) and effectively decoupling the spin \( S_2 \) from the band.

If we now switch on a small \( I_{\text{eff}} \ll T_K \), it will couple the remaining spin \( S_1 \) via the screened singlet to the conduction band. This effective Kondo coupling can be estimated by second-order perturbation theory to be of order \( I_{\text{eff}} D/T_K \). Therefore, ferromagnetic \( I_{\text{eff}} \) flows to zero as known from the ferromagnetic Kondo effect, and one ends up in a weak-coupling regime for the second spin, with a doublet ground state. On the other hand, antiferromagnetic \( I_{\text{eff}} \) grows logarithmically and leads to screening of the second spin 1/2. But, in contrast to a single-spin Kondo effect, this flow is cut-off when the effective coupling of the second spin becomes comparable to the singlet binding energy of the first spin \( S_1, T_K \), due to the indirect nature of this exchange. It is immediately clear what happens at this scale: The two spins \( S_1 \) and \( S_2 \) acquire a large direct antiferromagnetic coupling, \( J \), i.e., they form a singlet among themselves and effectively decouple from the band. Therefore, this regime of weak antiferromagnetic \( I_{\text{eff}} \) is continuously connected to the one of strong antiferromagnetic \( I \) where the two spins form an inter-impurity singlet from the outset. We can identify \( I_{\text{eff}} \) with \( (I - I_c) \), and the crossover scale \( T^* \) near the critical point is given by \( T^* \sim D \exp(-T_K/[I - I_c]) \).

In the case of maximal asymmetry, \( J_2 = 0 \), the picture developed above is most transparent. Here, no RKKY interaction is generated, and the critical coupling value is obviously \( I_c = 0 \). For \( J_2 > 0 \), we find \( I_c \sim J_1 J_2/D > 0 \) since the RKKY coupling is generically ferromagnetic because the two antiferromagnetic Kondo couplings prefer a parallel alignment of \( S_1 \) and \( S_2 \). Similarly, \( J_2 < 0 \) (but \( J_1 > 0 \)) gives a KT transition at \( I_c < 0 \).

Summarizing, the critical point in the double-impurity problem is defined by the coupling value where one spin completely decouples from the band, and the other spin is Kondo-screened. The boundary quantum phase transition is of the same type as in the single-impurity Kondo model when the bare coupling is tuned through zero, \( I = 0 \).

![FIG. 3. Schematic phase diagrams of the double-impurity Kondo model (Fig. 3a). The \( S_{\text{eff}} \) values in the different regimes are defined through the Curie term in the total impurity susceptibility, \( T_{\chi_{\text{imp}}} = S_{\text{eff}}(S_{\text{eff}} + 1)/3 \). a) For \( T = 0 \) and fixed \( J_1 \). The solid line is a line of KT transitions, the open dot is the first-order transition for \( J_1 = J_2 \). b) For \( T \geq 0 \) and fixed \( J_{1,2} \) with not too small asymmetry \((J_1 - J_2)\). The solid dot is the KT transition, the dashed lines denote crossovers.](image)

of the KT universality class.\(^3\)\(^2\) Close to the critical point for \( I > I_c \), we find a new exponentially small energy scale \( T^* \) within the Kondo regime – leading to two exponentially small scales, with \( T^* \ll T_K \) – and the crossovers can be understood as a two-stage Kondo effect. Such a two-stage behavior was so far only known in two-channel models\(^2\) remarkably, here it is realized with a single conduction electron channel.

We emphasize, however, that the stable fixed point for \( I > I_c \) is not given by two Kondo-screened impurities, but by an inter-impurity singlet. This has immediate consequences for the conduction electron phase shift. For ferromagnetic \( I_{\text{eff}} \) \((I < I_c)\), there is one screened spin 1/2 leading to a phase shift of \( \pi/2 \), whereas for \( I > I_c \) the low-T limit of the phase shift is zero.

It is also interesting to discuss the limit of small asymmetry of the Kondo couplings. As shown above, the singlet-doublet transition is of first order in the symmetric case. Therefore, very small asymmetry introduces another low-energy scale, \( T_{\text{as}} \propto |J_1 - J_2| \), and the KT behavior will be visible only for \( T \ll T_{\text{as}} \). We have verified this behavior numerically, and sample results are shown in Fig. 3a. Here, \( T_{\text{as}} \approx 10^{-10}D \), \( T_K \approx 10^{-3}D \), and a new regime \((\text{compared to Fig. }2a)\) occurs for \( T_{\text{as}} < T < T_K \) where effectively level crossing between the singlet and the underscreened doublet is observed. Our numerical results are consistent with \( T_{\text{as}} \sim T_K D |J_1 - J_2|/(J_1 J_2) \sim |T_{K1} - T_{K2}| \), with \( T_{K1,2} \sim D \exp[-D/(2J_{1,2})] \).

Schematic phase diagrams inferred from both the numerical results and the above arguments are shown in Fig. 3. At \( T = 0 \), Fig. 3a, A, we have a line of KT transitions terminating at a first-order point for \( J_1 = J_2 \). The finite-\( T \) crossovers are especially interesting, Fig. 3b: Above \( T_K \), the physics is dominated by the crossing of the \( S = 0 \) and \( S = 1 \) impurity levels – in particular, near \( I = I_c \) the two spins 1/2 fluctuate independently, leading to the plateau at \( T_{\chi_{\text{imp}}} \approx 2 \times S(S+1)/3 \) with \( S = 1/2 \). In contrast, below \( T_K \) the behavior is determined by the KT quantum phase transition at \( I_c \).
Two spins 1, two channels. We briefly want to comment on a different model, first considered in Ref. 21, which consists of two spins 1 coupled to a total of two screening channels \((K = 2)\), i.e., two underscreened \(S = 1\) Kondo impurities, Fig. 1b. The ground state spin can be tuned between zero and one, and therefore there must be a quantum phase transition as the coupling \(I\) is varied. Ref. 22 did not consider such a transition, but focussed exclusively on a possible phase transition associated with a critical non-Fermi liquid fixed point and a jump in the phase shift. It was found that such a transition does not exist for spins 1.

In analogy to the discussion above, we expect that at sufficiently low temperatures and \(I_{\text{eff}} \to 0\), each impurity realizes an underscreened \(S = 1\) Kondo effect, i.e., a spin 1/2 remains unscreened at each of the impurity sites. Turning on a ferromagnetic \(I_{\text{eff}}\) the two residual spins couple to an unscreened spin 1. In contrast, \(I_{\text{eff}} > 0\) promotes an effective antiferromagnetic coupling, both among the two residual spins as well as to the conduction electrons. However, the energy gain due to singlet formation with the conduction band is exponentially small in \(I_{\text{eff}}\), whereas the energy of the inter-impurity singlet is linear in \(I_{\text{eff}}\). Therefore, singlet formation between the two residual spins 1/2 is favorable, and this singlet will be effectively decoupled from the band.

It follows that the boundary quantum phase transition in this model associated with the change in the ground state spin is always of first order, although it will show non-trivial finite-\(T\) crossover behavior due to residual ferromagnetic Kondo couplings. Interestingly, this transition does not affect the conduction electron phase shift, as the phase shift is determined by the two Kondo-screened spin-1/2 degrees of freedom only, regardless of whether the two others form a decoupled singlet or triplet.

Applications of the double-impurity model. Two impurities coupled to a single orbital may be hard to realize directly in metals, however, the double-impurity model is obtained from the usual two-impurity model in the limit of small inter-impurity distance \(R\). In this limit, the screening channel associated with the odd linear combination of conduction electrons is weak, and the physics of the double-impurity model can be observed over a wide temperature range. Experimentally, such a situation can be achieved with magnetic ad-atoms on a metal surface manipulated by scanning tunneling microscope (STM) techniques.

Other applications can be constructed using coupled or multi-level quantum dots. In the former case, each dot represents a single “impurity” spin, and the inter-impurity interaction \(I\) can be varied either by tuning the inter-dot tunneling rate or the direct exchange interaction. For multi-level dots, we point out that the single-channel situation considered here appears to be the generic case for lateral devices – in contrast to a two-channel model appropriate for vertical dots. In fact, the results of Ref. 23 indicate that the KT transition discussed for the double-impurity model can indeed be realized in a multilevel dot.

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