Spin Glass and antiferromagnetism in Kondo-lattice disordered systems

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Abstract. The competition between spin glass (SG), antiferromagnetism (AF) and Kondo effect is studied here in a model which consists of two Kondo sublattices with a gaussian random interaction between spins in different sublattices with an antiferromagnetic mean Jo and standard deviation J. In the present approach there is no hopping of the conduction electrons between the sublattices and only spins in different sublattices can interact. The problem is formulated in the path integral formalism where the spin operators are expressed as bilinear combinations of Grassmann fields which can be solved at mean field level within the static approximation and the replica symmetry ansatz. The obtained phase diagram shows the sequence of phases SG, AF and Kondo state for increasing Kondo coupling. This sequence agrees qualitatively with experimental data of the Ce₂Au₁₋ₓCoₓSi₃ compound.

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1 Introduction

It is recognized that there is a strong competition between the Kondo effect and the RKKY interaction in Kondo lattice systems [11]. A transition from a magnetically ordered phase to a heavy fermion one, described by a Fermi-Liquid behaviour, has been observed in many Ce or Yb compounds and extensively studied from a theoretical point of view. There is a quantum critical point (QCP) at the transition and non-Fermi liquid behaviors are also observed near the QCP [2]. The role of disorder has been studied by different approaches including a ”Kondo disorder” model describing a broad distribution of Kondo temperatures [3] or the extensive study of the so-called ”quantum Griffiths” behaviour [4]. On the opposite, the transition from a metallic spin glass phase to a paramagnetic or Kondo phase has been studied recently by using the quantum rotor spin glass model [5] and the existence of an anomalous behavior near \( T = 0 \) has been observed in the transition between a metallic paramagnetic and a metallic spin glass.

Particularly, spin glass (SG) and Kondo state have been found together in some Cerium alloys like CeNi₁₋ₓCuₓ [8], Ce₂Au₁₋ₓCoₓSi₃ [7] and in some disordered Uranium alloys such as UCu₅₋ₓPdx [๔] or U₁₋ₓLaₓPdxAl₃ [9]. In the first case, there is an antiferromagnetic (AF) phase for low contents of Ni. When the Ni doping is increased the phase diagram becomes more complex. For \( x < 0.8 \), the sequence of phases SG-ferromagnetism arises when the temperature is lowered and a Kondo state exists for \( x < 0.2 \). The alloys Ce₂Au₁₋ₓCoₓSi₃ exhibit a phase diagram with a sequence of SG, AF and non magnetic Kondo phases with increasing the cobalt concentration at low temperature; in the high doping situation, the Neel temperature seems to tend to zero. In the two previously mentioned cases of disordered alloys [8,9], the AF, SG and NFL phases have been obtained at low temperatures for different concentrations; in particular, the sequence AF-NFL-SG occurs with increasing \( x \) in UCu₅₋ₓPdx alloys [8] and the opposite sequence AF-SG-NFL with increasing \( x \) in U₁₋ₓLaₓPd₂Al₃ alloys [9].

Thus, the results previously mentioned evidence a quite complicated interplay among the Kondo effect and the RKKY interaction when disorder and frustration are present. Recently, a theoretical effort has been done to understand how a spin glass phase emerges in a Kondo lattice model with an intrasite exchange interaction and an intersite long range random interaction among the localized spins [10]. The mentioned model has been extended to produce also a ferromagnetic order [11]. A particularity of our model [10,11] compared to those discussed in Ref.
is that we do not consider quantum fluctuations by coupling only the $S_z$ components of the localized spins in the spin glass term of the Hamiltonian, thus being unable to discuss a QCP at $T = 0$. One important remaining issue would be to obtain a description of the interplay between antiferromagnetism, Kondo phase and spin glass. The first step in that direction would be to produce a theory at mean field level able to mimic some important aspects, for instance, the sequence of magnetic phases of $Ce_2Au_{1-x}Co_xSi_3$ in terms of a minimum set of energy scales related to the fundamental mechanisms present in the problem. In this work this set is $J_0$ (the random intersite average), $J$ (the random intersite variance) and $J_K$ (the strength of the Kondo coupling). In the following, we take a constant $J_K$ and a Gaussian distribution of the intersite interaction with a random intersite interaction with average $J_0$ and variance $J$. Thus, we will not take values of $J_K$ and $T_K$ given by a random distribution as in the work of Miranda et al [11].

In view of that, the aim of this paper is to present a mean field theory to study the interplay among disorder and antiferromagnetic ordering in a Kondo lattice providing the necessary refinements in the previous theory [10] [11]. The model studied is a two Kondo sublattice with an intrasite exchange interaction and an intersite random gaussian interaction only between localized spins in two different sublattices [12]. We consider here that there is no hopping of the conduction electrons between the two sublattices; this assumption, which is used here to really simplify the calculations, does not modify the results since the different AF or SG ordering are in fact essentially due to the localized spins. The possible AF or SG ordering is, therefore, entirely related to the coupling of localized spins between the two sublattices. This fermionic problem is formulated by writing the spins operators as bilinear combinations of Grassmann fields and, through the static approximation and the replica formalism, the partition function is obtained, as already introduced to treat at mean field level the spin glass, Kondo effect and ferromagnetism in Refs. [10] and [11].

The Kondo effect and the RKKY interaction originate from the same intra site exchange interaction, but the necessity of considering an additional intersite exchange term has been already recognized [13] and the full Hamiltonian with both terms has been extensively used to describe the Kondo lattice [14] [15]. Moreover, the presently studied model has explicitly a random coupling term among localized spins in order to describe the spin glass situation [10] [11]. The exchange terms cannot be considered as completely independent from each other and a relationship giving the intersite exchange integral varying as the square of the Kondo local exchange integral $J_K$ has been introduced in order to mimic the $(J_K)^2$ dependence of the RKKY interaction [15]. In fact the relationship corresponds to an approximate representation of the intersite interaction, but we will use it in the last section of our paper in order to have a better agreement with experiment for some disordered Cerium alloys. Recently, the Doniach diagram has been revisited and the suppression of the AF and SG phases investigated, although these orders have been considered as two independent problems [16].

This paper is structured as follows. In Section II the model is introduced and developed in order to get the free energy and the corresponding saddle point equations for the order parameters. In Section III, as mentioned in the previous paragraph, a relationship among $J_0$, $J$ and $J_K$ is introduced allowing to solve the order parameter equations and to build up a temperature versus $J_K$ phase diagram showing the sequence of phases SG-AF-Kondo. The conclusions are presented in the last section.

2 General Formulation

The model considered here consists in two Kondo sublattices $A$ and $B$ with a random coupling only between localized spins in distinct sublattices [12]. Furthermore, there is no hopping of conduction electrons between the sublattices as previously explained. The corresponding Hamiltonian is given by:

$$\mathcal{H} = \mu N = \sum_{p=A,B} \sum_{i,j} \sum_{\sigma=\uparrow,\downarrow} t_{ij} d_{i,p,\sigma}^\dagger d_{j,p,\sigma} + \sum_i \varepsilon_d^{0,p} \hat{d}^i_{0,p}$$

$$+ J_K (\hat{S}^{\uparrow}_{i,p} \hat{S}^{\uparrow}_{i,p} + \hat{S}^{\downarrow}_{i,p} \hat{S}^{\downarrow}_{i,p}) + \sum_{ij} J_{ij} \hat{S}^{\uparrow}_{i,A} \hat{S}^{\uparrow}_{j,B}$$

where $i$ and $j$ sums run over $N$ sites of each sublattice. The intersite interaction $J_{ij}$ is assumed to be a random quantity following a gaussian distribution [12]

$$\mathcal{P}(J_{ij}) = \frac{1}{\sqrt{64 \pi}} \exp \left\{ - \frac{(J_{ij} + 2J_0/N)^2}{64J^2} \right\}.$$  \hspace{1cm} (2)

An anti-ferromagnetic solution for the present choice of $J_{ij}$ can be found for $J_0 > 0$. The case $J_0 < 0$ produces a complex phase diagram with spin glass, ferromagnetic, mixed phase (a spin glass with spontaneous magnetization) [11] and a Kondo state as defined in the Ref. [10].

The spin variables present in the Eq. (1) are defined closely to Ref. [10] modified for the case of two interacting distinct sub-lattices given as: $\hat{s}^+_{i,p} = d^\dagger_{i,p,\uparrow} d^\dagger_{i,p,\downarrow} = (\hat{s}^\dagger_{i,p})$, $\hat{s}^+_{i,p} = \hat{f}^\dagger_{i,p,\uparrow} \hat{f}_{i,p,\downarrow} = (\hat{s}^\dagger_{i,p})$ and $\hat{s}^+_{i,p} = \frac{1}{2} \left[ \hat{f}_{i,p,\uparrow} \hat{f}_{i,p,\downarrow} - \hat{f}_{i,p,\downarrow} \hat{f}_{i,p,\uparrow} \right]$ where $d^\dagger_{i,p,\sigma}$, $\hat{d}^\dagger_{i,p,\sigma}$ ($\hat{f}_{i,p,\sigma}$, $\hat{f}_{i,p,\sigma}$) are the creation and destruction operators for conduction (localized) fermions.

The partition function can be given by using Grassman variables $\psi_{i,p,\sigma}(\tau)$ for the localized fermion and $\varphi_{i,p,\sigma}(\tau)$ for the conducting ones:

$$Z = \int \prod_{p=A,B} \mathcal{D}(\psi_{i,p,\sigma}) \prod_{p=A,B} \mathcal{D}(\varphi_{i,p,\sigma}) e^{-A}$$

where the action is $A = A_0 + A_K + A_{SG}$ with

$$A_0 = \sum_{p=A,B} \sum_{i,j} \sum_{\omega} \left( g_{ij,p}(\omega) \right)^{-1} \left[ \varphi^+_{i,p,\uparrow}(\omega) \varphi_{j,p,\uparrow}(\omega) + \varphi^+_{i,p,\downarrow}(\omega) \varphi_{j,p,\downarrow}(\omega) \right]$$
characterized by the complex order parameter $\lambda$ 

\[ \text{Eq. (8)} \]

to solve this problem at mean field level. In that 

identity

\( \exp(\lambda_\sigma) \)

\( \langle \cdots \rangle \)

\( \text{reference the chemical potentials of the conduction and} \)

\( \text{localized bands, respectively.} \)

The actions $A_{SG}$ and $A_K$ are given by

\[ A_{SG} = \beta \sum_{i,j} \sum_{\nu} J_{ij} S_{i,A}(\nu) S_{j,B}^*(\nu) \]  

and

\[ A_K = \sum_{\sigma} \beta J_K \left[ \sum_{i} \lambda_{\sigma} \varphi^*_{i,A,\sigma}(\omega) \varphi_{i,A,\sigma}(\omega) \right] \]

with the free energy is given by the replica method

\[ \frac{Z^{(\text{stat})}}{Z_0} = \int \prod_{p=A,B} D(\psi^*_p \psi_p) e^{A_{SG} + A_{\text{eff}}} \]  

where

\[ A_{\text{eff}} = \sum_{i,j,\sigma} \varphi^*_{i,j}(\omega) g_{ij}(\omega) \varphi_{i,j}(\omega) \]  

with

\[ g_{ij}(\omega) = \left[ \begin{array}{cc} 0 & -F_A(\omega) \\ 0 & \sum_{\omega} \right] \]  

\[ \Psi_i^* (\omega) = \left[ \begin{array}{c} \varphi^*_{i,A,\sigma}(\omega) \\ \varphi^*_{i,B,\sigma}(\omega) \end{array} \right] \]  

\[ F_p(\omega) = \beta^2 J_k^3 |\lambda_p|^2 \sum_k e^{i(k\mathbf{R}_i-R_j)} \gamma_k(\omega) \]

where $g_{ij}(\omega)$ is given in Eq. (10) and

\[ \gamma_k(\omega) = \frac{1}{\sum_{\omega} \} - \beta \epsilon_k \)  

The free energy is given by the replica method

\[ \beta F = \beta J_K \left| |\lambda_A|^2 + |\lambda_B|^2 \right| \]

\[ \lim_{n \to 0} \frac{1}{2N} \left[ \left\langle \left. \left( Z^{(\text{stat})}(J_{ij}) \right) \right| \right. \right]_{CA} - 1 \]

where the configurational average $\langle \langle \cdots \rangle \rangle_{CA}$ is performed with the gaussian distribution given by Eq. (2) applied in Eq. (2) which gives

\[ \left\langle \left. \left( Z^{(\text{stat})}(J_{ij}) \right) \right| \right. \right]_{CA} = \int D(\psi^* \psi) e^{A_{\text{eff}}} (Z_{\text{stat}}) \]
where

\[ \langle \langle Z_{sc} \rangle \rangle_{CA} = \prod_{i,j} \left\langle \exp \left( \beta J_{ij} \sum_{\alpha} S_{i,A,\alpha}^z S_{j,B,\alpha}^z \right) \right\rangle_{CA} \] (21)

Therefore, the resulting averaged partition function can be linearized by using the usual Hubbard-Stratonovich transformation and introducing auxiliary fields of the spin glass part [12]. The details of this procedure are given in the Appendix.

The free energy can be found using the averaged partition function (see Appendix) in Eq. (19). Thus, one gets,

\[
\beta F = \beta J_K (|\lambda_A|^2 + |\lambda_B|^2) - \beta^2 \frac{J^2}{2} q_A q_B \\
+ \beta^2 \frac{J^2}{2} (\bar{q}_A \bar{q}_B) - \beta J_0 m_A m_B \\
- \lim_{n \to 0} \frac{1}{2Nn} \int_{-\infty}^{\infty} \prod_{i=1}^N D\xi_i,A D\xi_i,B \prod_{\alpha} \int_{-\infty}^{\infty} D\zeta_{\alpha,i,A} D\zeta_{\alpha,i,B} \\
\times \exp \left[ \sum_{\omega,\sigma} \ln \det \left[ G_{ij}^\alpha (\omega) h_{i,p}^\alpha \right] \right] (22)
\]

where the \( G_{ij}^\alpha (\omega) h_{i,p}^\alpha \) is given by

\[
G_{ij}^\alpha (\omega) h_{i,p}^\alpha = \begin{bmatrix}
(i\omega - \beta \epsilon_0^f - \sigma h_{i,A}^\alpha)\delta_{ij} - F_A(\omega) & 0 \\
0 & (i\omega - \beta \epsilon_0^f - \sigma h_{i,B}^\alpha)\delta_{ij} - F_B(\omega)
\end{bmatrix}
\] (23)

with \( F_p \) defined in Eq. (17) and \( h_{i,A}^\alpha \) and \( h_{i,B}^\alpha \), being random gaussian fields (see Eq. (24) from Apendix) applied to the sites of one sublattice (A or B) which depends on the parameters of the other sub-lattice. At this point, the static susceptibility \( \chi_p \) can be introduced related with replic symmetry diagonal order parameter \( \bar{q}_p = \bar{\chi}_p + q_p \) where \( \chi_p = \chi_p / \beta \).

The central issue here is to adopt the proper decoupling approximation which allows one to calculate the matrix \( G_{ij}^\alpha (\omega) h_{i,p}^\alpha \). The elements given in Eq. (22) are referred to the original sublattice A (B) where to each site \( i \) there is a random gaussian field \( h_{i,A}^\alpha \) (\( h_{i,B}^\alpha \)) applied. The decoupling procedure is to consider these random fields \( h_{i,A}^\alpha \) (\( h_{i,B}^\alpha \)) as applied in two fictitious Kondo sub-lattices. Therefore, in each site \( \mu \) of the new sublattice, the applied field \( h_{i,A}^\alpha \) (\( h_{i,B}^\alpha \)) is constant. That is equivalent to replace the Green’s function \( G_{ij}^\alpha (\omega) h_{i,p}^\alpha \) by \( G_{ij}^\alpha (\omega) h_{i,p}^\alpha \) in Eq. (23). It is, therefore, possible to go to the reciprocal space, where it is assumed a constant band \( \rho(\varepsilon) = \frac{1}{2\pi} \) for \(-D < \varepsilon < D\) for the conduction electrons in each sub-lattice. The sum over the Matzubara frequencies in Eq. (22) can be done following a standard procedure. Thus, the free energy is found to be

\[
\beta F = \beta J_K (|\lambda_A|^2 + |\lambda_B|^2) + \beta^2 \frac{J^2}{2} \bar{\chi}_A \bar{\chi}_B + \frac{\beta^2 J^2}{2} (\bar{\chi}_A q_B + \bar{\chi}_B q_A) - \beta J_0 m_A m_B \\
- \frac{1}{2} \int_{-\infty}^{\infty} D\xi_i,A \int_{-\infty}^{\infty} D\xi_i,B \\
\ln \left. \prod_{p=A,B} \int_{-\infty}^{\infty} Dz_p e^{E(h_p)} \right|_{p=A,B} (24)
\]

with

\[
E(h_p) = \frac{1}{\beta} \int_{-\beta D}^{\beta D} dx \left[ \cosh \left( \frac{x + h_p}{2} \right) + \cosh \left( \frac{x - h_p}{2} \right) + \beta^2 J_0^2 |\lambda_p|^2 \right].
\] (25)

The saddle point equations for the order parameters \( q_p, m_p, \bar{\chi}_p \) and \( |\lambda_p| \) follow from Eqs. (24) and (25).

3 Results

The numerical solutions of equations for the order parameters \( q_p, \bar{\chi}_p, m_p \) (\( p = A,B \)) and \( |\lambda_p| \) allow one to build up a phase diagram of temperature \( T \) versus the set of relevant energy scales \( J_0, J \) and \( J_K \) defined in Section II. The thermodynamic phases are identified as: (a) an antiferromagnetic (AF) phase corresponding to \( m_A = -m_B \) or \( m_3 = 0 \) (see Eq. (29)-(32)); (b) a spin glass phase for \( q_A \neq 0 \) and \( q_B \neq 0 \) (\( q_A \neq 0 \)); (c) a Kondo state given by \( |\lambda_A| \neq 0 \) and \( |\lambda_B| \neq 0 \). The phase diagrams obtained by the present calculations are derived for several sets of parameters \( J_K/J \) and \( J_0/J \) considered firstly as independent from each other. But, as it was discussed in the introduction, the intrasite and intersite exchange terms cannot be considered as completely independent from each other and a \( J_K^2 \)-dependence of \( J_0 \) was introduced in order to mimic the RKKY interaction [15]. Thus, a phase diagram is shown in Fig. (1) by keeping \( J \) constant, taking then:

\[
\frac{J_0}{J} = \alpha \left( \frac{J_k}{J} \right)^2.
\] (26)

and by choosing finally \( \alpha = 0.0051 \). The factor \( \alpha \) was chosen here to have the AF phase starting at \( J_k/J \approx 12 \). The numerical solutions of the order parameters yield the sequence of phases SG-AF-Kondo shown in Fig. (1). The Kondo temperature \( T_K \) is decreasing for decreasing values of \( J_k/J \) down to \( T_K/J \approx 15 \). From this point onwards there is a phase transition leading to antiferromagnetic order with the Neel temperature \( T_N \) monotonically decreasing with \( J_k/J \). For still smaller values of \( J_k/J \) (and therefore smaller values of \( J_0/J \)) the spin glass behaviour becomes dominant.

Thus, in this mean field theory it is possible to identify three sorts of distinct regimes depending on the strength
of the Kondo coupling $J_k$. The first one (for high $J_k$) is the complete screening of the localized magnetic moments due to the Kondo effect, which is characterized by the order parameter $|\lambda_p|$ related to the formation of d-f singlet throughout the whole two sublattices. In the second one (for decreasing $J_k$), the magnetic moments of one particular sublattice, for instance sublattice $A$, survive to the screening process (the complete unscreening means $|\lambda_A| = 0$ and $|\lambda_B| = 0$) and start to be in an antiparallel alignment with an internal field $h_A$, which depends on the magnetization and susceptibility of the sublattice $B$, until this internal field is spread out to the whole sublattice producing the AF order. In the last regime (for $J_k/J < 12$ or $J_0 < 0.734 J$), the effects of randomness begin to be dominant. In the effective field $h_p' (p = A, B)$, the replica spin glass order parameter $q_p'$ (p = B, A) component starts to be non-null indicating the non-trivial ergodicity breaking leading to a spin glass phase at the transition temperature $T_f$. Finally, let us remark that we never obtain theoretically mixed phases where there is the coexistence of the Kondo phase with the other phases. This is due to the mean field approximation used here, as already observed [10,11].

The experimental phase diagram of the alloy Ce$_2$Au$_{1-x}$Co$_x$Si$_3$ [14] can be addressed if the $J_k$ coupling is associated with the content of Co. The obtained result shown in Fig. 1 displays the same sequence of phases at low temperature as the experimental one. In brief, the increasing $J_k$ favours the transition from a SG phase to a AF ordering and then to the screening of the localized moments. Thus, this mean field description is able to account for experimental aspects of Ce$_2$Au$_{1-x}$Co$_x$Si$_3$ at low temperatures.

There is some disagreement related to the location of the AF line transition. The experimental behavior [15] shows the Neel temperature $T_N$ decreasing apparently towards a quantum critical point (QCP) with increasing Co doping. However, recently the spin flipping has been simulated with the presence of a transversal field in the x-direction to study a QCP in fermionic spin glass [17]. The same approach has been extended to investigate the spin glass freezing in a Kondo lattice [18]. Hence, this method could also be used in the present model in order to clarify the role of the QCP in the interplay between spin glass and antiferromagnetism in a Kondo lattice. That will be subject for future investigations.

In conclusion, we have studied in detail the competition between SG, AF and Kondo phases in a mean field approach of Kondo-lattice disordered alloys. Using the peculiar relationship [20] between the different parameters of the model, we have obtained a phase diagram showing the sequence SG-AF-Kondo phases in good agreement with the experimental phase diagram of the disordered alloys Ce$_2$Au$_{1-x}$Co$_x$Si$_3$, if we assume that $J_k$ increases with increasing Cobalt concentration. Other phase sequences, involving also SG, AF and Kondo phases, obtained in disordered Uranium alloys, are not accounted for, at present, by our model. But the choice of parameters and their relationship with the varying concentration in these alloys are really delicate questions which are not answered at present. Moreover, in such Uranium systems, the Kondo phases have clearly a non-Fermi liquid behavior which is not described here. Finally, the present model can account for the presence of SG, AF and Kondo phases, but further work is necessary in order to obtain a better agreement with experimental data for Cerium and Uranium disordered alloys.

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Appendix

The averaged partition function can be given by [12]:

$$\langle Z_{SG} \rangle = \prod_{p=A,B} \sum_{\alpha} \frac{1}{N} \int \frac{\sqrt{N\beta J_0}}{2\pi} e^{-\frac{N\beta J_0}{2\pi} \sum_{\alpha} M_p^\alpha}$$

$$\times \prod_{p=A,B} \int \frac{\sqrt{N\beta^2 J_2}}{2\pi} e^{-\frac{N\beta^2 J_2}{2\pi} \sum_{\alpha} \sum_{\beta} Q_p^{\alpha\beta}}$$

$$\times \prod_{\alpha\beta} \int \frac{\sqrt{N\beta J_0}}{2\pi} e^{-\frac{N\beta J_0}{2\pi} \sum_{\alpha} M_3^\alpha}$$

$$- \frac{\beta^2 J^2}{2} \sum_{\alpha\beta} \left( \frac{Q_3^{\alpha\beta}}{2} \right)^2 + \frac{1}{N} \ln A(M_3^\alpha, Q_3^{\alpha\beta})$$
\[ + \frac{\beta J_0}{2} \sum_\alpha \left[ (M_3^\alpha)^2 + \sum_{p=A,B} (M_p^\alpha)^2 \right] \} \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) \right) 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