THE SPIN GLASS-KONDO COMPETITION IN DISORDERED CERIUM SYSTEMS

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We discuss the competition between the Kondo effect, the spin glass state and a magnetic order observed in disordered Cerium systems. We present firstly the experimental situation of disordered alloys such as $CeNi_{1-x}Cu_x$ and then the different theoretical approaches based on the Kondo lattice model, with different descriptions of the intersite exchange interaction for the spin glass. After the gaussian approach of the Sherrington-Kirkpatrick model, we discuss the Mattis and the van Hemmen models. Then, we present simple cluster calculations in order to describe the percolative evolution of the clusters from the cluster spin glass to the inhomogeneous ferromagnetic order recently observed in $CeNi_{1-x}Cu_x$ disordered alloys and finally we discuss the effect of random and transverse magnetic field.

Keywords: Spin glasses, Kondo Lattice models, disordered Cerium systems.

1. Introduction

The influence of the disorder on the behavior of strongly correlated electron systems has been extensively studied and we will discuss here the strong competition between the Kondo effect, a magnetic order and the spin glass state occurring in disordered Cerium systems. In fact, there are only a few systems which present together the Kondo effect and a spin glass behavior with also a magnetic order. Such a coexistence has been observed in disordered Cerium or Uranium alloys and we will firstly discuss the experimental situation of these systems.

First, the $CeNi_{1-x}Cu_x$ alloys have been extensively studied and their properties vary with the concentration of Nickel. For large x values above 0.7, the alloys are antiferromagnetic at low temperatures, while, for small x values below 0.2, they present a Kondo behavior. But, for $0.3 < x < 0.7$, these alloys change with decreasing temperature from the paramagnetic state to a spin glass one and finally to a ferromagnetic order.^{1,2} There has been recently a considerable progress in the experimental knowledge of the phase diagram and magnetic clusters occur around

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the spin glass-ferromagnetic transition; this new phase diagram will be described in the section 3 introducing some new calculations on magnetic clusters.³⁻⁵

Another example is provided by the $CeRh_xPd_{1-x}$ alloys which are clearly ferromagnetic for x values smaller than 0.65 and present a complicate Kondo behavior for large x values. The Curie temperature disappears for x around 0.8, but there is no quantum critical point. Instead, a peculiar "Kondo-cluster-glass" is found and Non-Fermi-Liquid effects in the specific heat, ac susceptibility and magnetization support a spin glass state described by the Griffiths model.^{6,7}

The alloys $Ce_2Au_{1-x}Co_xSi_3$ present also a peculiar phase diagram: for small x values below 0.4, an antiferromagnetic phase is followed with decreasing temperature by a spin glass phase; then there exists only an antiferromagnetic phase up to roughly $x=0.8$ and finally a Kondo lattice state occurs for very large x values tending to 1.⁸

The compound $CeAgGa$ shows a spin-glass state with a freezing temperature of 5.1 K and a ferromagnetic-like order at the temperature of 3.6 K.⁹ The spin glass state appears to arise from a random distribution of the Ag and Ga atoms around the Ce atoms and this compound looks like the $CeNi_xCu_{1-x}$ alloys for an intermediate concentration. The compound Ce_2NiGe_3 is also a Kondo lattice compound showing a spin glass behavior.¹⁰ In fact, there are often some complicate behaviors involving a Non-Fermi-Liquid effect rather than the classical Kondo effect. For example, in the $CeCoGe_{3-x}Si_x$ alloys (with $0 < x < 3$), the phase diagram shows, with increasing x , an antiferromagnetic order, then a Kondo Fermi Liquid behavior and finally a Non Fermi Liquid (NFL) behavior.¹¹ But, above 6.5 kbar, the compound $CeCoGe_{2.2}Si_{0.8}$ undergoes a change to a spin-glass like behavior.¹²

A similar situation occurs with alloys containing Uranium instead of Cerium. Different behaviors such as antiferromagnetic order, NFL, spin glass and superconductivity have been observed in $U_{1-x}R_xPd_2Al_3$ alloys where R can be La, Th or Y;¹³ in the case of $U_{1-x}La_xPd_2Al_3$ alloys, one observes first an antiferromagnetic order with a decreasing Neel temperature for x increasing from 0 to 0.2, then a spin glass phase up to $x = 0.7$ and finally a Kondo state with a NFL behavior. Similar properties have been shown in $UCu_{5-x}Pd_x$.^{14,15} The phase diagram of $UCu_{5-x}Pd_x$ alloys presents an antiferromagnetic order for an increasing concentration x of order 0.8 and a spin glass behavior for x larger than 1.5. For x between 1 and 1.5, there is occurrence of a NFL behavior certainly connected to the Kondo effect.¹⁵ The NFL properties of these Uranium systems have been also extensively studied.^{16,17} In fact, the competition between the Kondo effect and the spin glass in Uranium systems needs a treatment of the Kondo effect within the Underscreened Kondo Lattice model¹⁸ and we will not discuss here the case of Uranium disordered alloys.

We have done a brief experimental review, but indeed details and especially the different phase diagrams can be found in the papers cited in reference. Now, after this brief review, we will present in the next section the basic models which have accounted for the experimental results, then in the following section the derivation of some simple calculations which can describe the clusters occurring in both the

spin glass and the ferromagnetic phases and finally in the last section the effect of random and transverse magnetic field.

2. Models for the Kondo-Spin Glass-Magnetic Order Competition

There are different works on the competition between the two well known Kondo and spin glass effects,^{19–21} but here we will present the models that we have recently developed to account for the phase diagrams of disordered Cerium alloys showing the competition between a spin glass phase, a Kondo state and a magnetic order, as observed in $CeNi_{1-x}Cu_x$ alloys. The previous disordered alloy is a very good experimental example to be accounted by our models: the compound CeNi has a Kondo behavior and the compound CeCu has a simple magnetic behavior; when the concentration x decreases from 1 to 0, the importance of the Kondo effect increases and consequently the intrasite Kondo coupling J_K increases. Thus, we can compare the different theoretical models with increasing J_K (as shown in Figure 1) to the experimental phase diagram observed in $CeNi_{1-x}Cu_x$ alloys with decreasing the concentration x .

To describe this behavior, we have taken first a conduction band and localized spins on each Cerium site; then we consider a Kondo interaction between the localized spin $\mathbf{S}_i$ and the spin $\mathbf{s}_i$ of a conduction electron at the same site. Then we add an intersite interaction between nearest-neighboring localized spins in order to describe the spin glass effect. In fact, it is well known that the intersite interaction depends on the model chosen to describe the spin glass physics and we will review very briefly the first models used and we will present different comparisons with experiment. Thus, we have performed different calculations for the spin glass-Kondo interaction and the main difference between them lies in the approach used to describe the spin glass.

We start from the following general Hamiltonian :

$$H = \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}} n_{\mathbf{k}\sigma}^c + \sum_{i\sigma} E_0 n_{i\sigma}^f + J_K \sum_i \mathbf{s}_i \cdot \mathbf{S}_i + \sum_{i,j} J_{ij} S_{fi}^z S_{fj}^z \quad (1)$$

where $\varepsilon_{\mathbf{k}}$ is the energy of the conduction band, $J_K > 0$ is the intrasite Kondo coupling and J_{ij} is the intersite interaction used to describe the spin glass effect.

In fact, the disorder is described by the interaction between the spins $\mathbf{S}_i$ of different magnetic Cerium atoms, as it is the case in spin-glass systems and the randomness cannot come from the intrasite Kondo effect, but only from intersite interactions. Also, we have taken here the Ising model which was already used in many spin glass calculations. In fact, the ground state is often a doublet in Cerium systems, which gives a good justification for the use of the simpler Ising model.

To describe the nature of the spin glass, we start firstly with the Sherrington-Kirkpatrick model²² in which the couplings J_{ij} are taken here as independent random variables described by a gaussian distribution :

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

In the preceding expression, the average value J_0 has been taken firstly equal to zero²³ and this calculation describes the SG-Kondo transition. In order to try to obtain a comparison with experiment, we have considered the case $J_0 < 0$, which produces a complex phase diagram with spin glass, ferromagnetic, mixed (a spin glass with spontaneous magnetization) and Kondo phases²⁴ and the case $J_0 > 0$, which corresponds to an antiferromagnetic phase in competition with Spin Glass and Kondo phases.²⁵ Another calculation has been done in order to include the effect of a transverse magnetic field.²⁶

In these calculations, we have taken the Sherrington-Kirkpatrick model with a gaussian distribution of the intersite exchange integrals.²² The first studied case takes $J_0=0$ ²³ and we discuss the results as a function of only the 2 parameters J and J_K characteristic of the spin glass and of the Kondo states. The functional integration techniques and the mean field approximation are used here ; to describe the Kondo effect, we use the correlation $\lambda_\sigma = \langle c_{i\sigma}^\dagger f_{i\sigma} \rangle$ between conduction and f electrons. The resulting phase diagram in the $T - J_K$ plane for a fixed J value gives a spin glass phase for small J_K values and a Kondo phase for large J_K values. But in fact, the experiments on $CeNi_{1-x}Cu_x$ alloys show clearly a additional competition with a ferromagnetic order. Then, in order to obtain a more complex phase diagram with a ferromagnetic²⁴ or an antiferromagnetic²⁵ phase occurring at low temperatures for small J_K values, we have taken the same model with a non zero value J_0 of the center of the gaussian distribution. Thus, it results that the Kondo phase obtained for large J_K values is still there, but that there is a competition between the spin glass phase and the magnetic phase for smaller J_K values. However, in the ferromagnetic case, when J_K is typically of order J_0 , we have obtained, with decreasing temperature, successively a ferromagnetic phase, then a mixed ferromagnetic-spin glass phase and finally a spin glass phase. The occurrence of a spin glass, Kondo and ferromagnetic phases in the same phase diagram is a good result, but the evidence of a spin glass phase at very low temperatures below the ferromagnetic one is in disagreement with experimental results previously obtained for $CeNi_{1-x}Cu_x$ alloys.

In order to improve the agreement with experiment, we have more recently developed a generalization of the Mattis model²⁷ and of the van Hemmen model²⁸ which both describe the bonds J_{ij} joining the localized spins as depending on a random choice on each atom. We have developed firstly a generalization of the Mattis model,²⁷ which represents an interpolation between ferromagnetism and a highly disordered spin glass.²⁹

In the Mattis model,²⁷ which has been proposed as a solvable model to the spin glass problem, the bonds joining the localized spins have been defined as separable random variables ξ_i . In this model the couplings between spins are given by :

$$J_{ij} = \frac{1}{N} \sum_{\mu\nu} J_{\mu\nu} \xi_i^\mu \xi_j^\nu, \quad (3)$$

where $\xi_i^\mu = \pm 1$ ($\mu = 1, 2, \dots, p$; $i = 1, 2, \dots, N$) are independent random distributed variables. For the classical Ising model, if $\mu = \nu = 1$, the original Mattis model²⁷ is recovered. However, if $J_{\mu\nu} = J\delta_{\mu\nu}$ and $p = N$ with the N^2 random variables ξ_i^μ having mean zero and variance one, in the limit of N large J_{ij} tends to a Gaussian variable with mean zero and variance $N^{-1/2}J$ as in the SK model.²² Therefore, we can consider this model as an interpolation between ferromagnetism and highly disordered spin glass.³⁰

For a reasonable value of the relative importance of the ferromagnetic and spin glass phases, i.e. for a relatively small J_K/J ratio, we have obtained, with decreasing temperature, a spin glass phase, then a mixed ferromagnetic-spin glass phase and finally a ferromagnetic phase. The Kondo phase is always present for large J_K/J . This theoretical result accounts better for the experimental phase diagram of $CeNi_{1-x}Cu_x$ alloys., with in particular at low temperatures the ferromagnetic phase below the spin glass phase.

Finally, we have used recently the van Hemmen model²⁸ to describe the interplay of Kondo effect, Spin Glass and Ferromagnetism in disordered Ce alloys. This model gives a good description of this interplay and appears to be simpler than the previous models, in particular for the treatment of clusters, as we will see later on.³¹

Thus, in order to improve again the agreement with the experimental case of $CeNi_{1-x}Cu_x$ alloys, we introduce here a new kind of disordered coupling J_{ij} given by van Hemmen (vH)²⁸ as:

$$J_{ij} = \frac{J}{N}(\xi_i\eta_j + \eta_i\xi_j) + \frac{I_0}{N} \quad (4)$$

where ξ_i and η_j are equal to ± 1 and are random variables which follow a bimodal distribution.³¹ We take here both a random SG contribution and a ferromagnetic one respectively proportional to the parameters J and I_0 . Our calculations using the vH approach have given different phase diagrams which depend on the ratio I_0/J . Several cases are described in Ref.³¹ and we have obtained the typical case of a transition with decreasing temperature from spin glass (SG) to ferromagnetism (FE) with an intermediate "mixed" phase SG+FE for small J_K values and still the Kondo phase for large J_K values.³¹ The use of the van Hemmen model offers some advantages: first the calculation does not need the use of the replica method and is simpler; second there exists a real mixed phase between Spin Glass and Ferromagnetism, which is in good agreement with the experimental phase diagram of $CeNi_{1-x}Cu_x$ alloys,³¹ as shown on Figure 1.

Our recent work, with the Kondo effect, ferromagnetism and the van Hemmen model used for the spin glass, gives therefore a good description of the phase diagram of $CeNi_{1-x}Cu_x$ alloys,³¹ but recent experimental progress has shown the occurrence of clusters and we will describe in the next section recent experimental and theoretical progress. However, in conclusion of this section, we can say that the more "local" description^{29,31} given by the Mattis or the van Hemmen model seems

Fig. 1. Phase diagram $T/J - J_K/J$ for $J_0/J=1.4$.³¹

to be better here than the "average" description using the Sherrington-Kirkpatrick approach^{23,24} to describe the phase diagram of $CeNi_{1-x}Cu_x$ alloys.

3. The Kondo Cluster — Glass Model

Up to now, we have discussed different approaches to describe the Kondo-spin glass-ferromagnetism competition and we have applied them successfully to the first description of the phase diagram of Cerium disordered alloys like $CeNi_{1-x}Cu_x$. But, more sophisticated experiments performed at very low temperatures have shown the presence of magnetic clusters in these alloys. More precisely, measurements of the ac susceptibility, dc magnetization and specific heat down to 0.1 K show that a cluster-glass state is formed at a freezing temperature T_f well above the ferromagnetic state. Recent Muon spin relaxation and Small Angle Neutron scattering measurements confirm the existence of magnetic clusters and of a percolative transition with decreasing temperature.³ Thus, it is now established that, in $CeNi_{1-x}Cu_x$ alloys for x between 0.3 and 0.6, an inhomogeneous "cluster spin glass" (CSG) occurs below the paramagnetic regime and is followed, with decreasing temperature, by an inhomogeneous ferromagnetic (IFM) order, with a percolative evolution of the clusters from the CSG state to the IFM state.^{4,5} Figure 2 summarizes the evolution of the cluster glass which does not show a phase transition, but a percolative evolution. A similar cluster spin glass has been observed in $CePd_{1-x}Rh_x$.^{6,7}

We have developed several works to account for the cluster glass and the starting point is to consider small clusters with a constant size and with an intra-site Kondo interaction, an intra-cluster ferromagnetic interaction and an inter-cluster random magnetic interaction. The first work uses the classical Sherrington-Kirkpatrick model to treat the spin glass interaction,³² but more recently we have developed the more convincing van Hemmen model.³³

Fig. 2. Experimental phase diagram for $CeNi_{1-x}Cu_x$ alloys.⁵

To study the competition between the Kondo effect and the cluster spin glass, we use the following Hamiltonian:

$$H = \sum_{a=1}^{N_{cl}} \sum_{i=1}^{n_s} \epsilon_0 \sum_{\sigma=\uparrow\downarrow} \hat{n}_{i\sigma a}^f - J_0 \sum_{a=1}^{N_{cl}} \sum_{i<j}^{n_s} \hat{S}_{ia}^z \hat{S}_{ja}^z + \sum_a \sum_{i,j}^{N_{cl}} t_{ij}^{aa} d_{i\sigma a}^\dagger d_{j\sigma a} \quad (5)$$

$$+ J_K \sum_{a=1}^{N_{cl}} \sum_{i=1}^{n_s} \left(\hat{S}_{ia}^+ s_{ia}^- + \hat{S}_{ia}^- s_{ia}^+ \right) - \sum_{a<b}^{N_{cl}} J_{ab} \hat{S}_a^z \hat{S}_b^z$$

We have here $N = N_{cl} n_s$, where N_{cl} and n_s are the number of clusters and the number of spins in each cluster, respectively. The hopping t_{ij}^{aa} is only inside the cluster. The indices (a, b) refer to clusters while (i, j) indicate spins inside a cluster.

So, we can write $S_{ia}^+ = f_{ia\uparrow}^\dagger f_{ia\downarrow}$, $s_{ia}^- = d_{ia\downarrow}^\dagger d_{ia\uparrow}$ and

$$\hat{S}_a^z = \sum_{i=1}^{n_s} \hat{S}_{ia}^z = \sum_{i=1}^{n_s} \sum_{\sigma=\uparrow\downarrow} \sigma \hat{f}_{i\sigma a}^\dagger \hat{f}_{i\sigma a} \quad (6)$$

The intercluster coupling J_{ab} is a random variable given in the van Hemmen model by an equation similar to equation (4). Details on our first treatment using the Sherrington-Kirkpatrick model can be found in Ref. 32 and we discuss here only the van Hemmen (vH) model.³³

This model starts from Kondo and ferromagnetic intra-cluster interactions and an inter-cluster vH interaction given by equation (4). The inter-cluster interactions are treated in a mean field level without the use of replica method, in which the magnetization and spin glass order parameters are introduced in the partition function. These order parameters are chosen to minimize an effective one-cluster free energy and the resulting intra-cluster interactions are then obtained by exact diagonalization. For this purpose, it is assumed that the localized fermions can act on

Fig. 3. (a) Order parameters q , m and λ as a function of T/J for $J_K/J = 0$ and 2. (b) q , m and λ versus J_K/J for $T/J = 0.2$. These results are for $n_s = 5$ and $I_0/J = 0.1$.³³

a basis of states where each site is exactly occupied by only one fermion, while the conduction fermions can act on states with four possible occupancies at each site (site unoccupied, single occupied and double occupied).

The number of atoms n_s inside each cluster is presently limited to 4 or 5.³³ The parameters λ , q and m design respectively the Kondo order parameter, the spin glass order parameter and the magnetization. However, for relatively large values of J_K and I_0 , we have obtained successively, with decreasing temperature, a Kondo phase followed by a Kondo cluster spin glass where both the Kondo and the cluster spin glass (CSG) order parameters are non zero; finally a mixed CSG and ferromagnetic phase is obtained. The results are shown in Figure 3 corresponding to clusters with a number $n_s = 5$ of atoms. The Kondo correlation is increasing with J_K as usual, and the figure shows a competition between the Kondo effect and the magnetic orders, the cluster spin glass (CSG) and the mixed phase (CSG+FE). These magnetic orders are strongly dependent on the ferromagnetic inter-cluster interaction I_0 and J_K .³³ It is important to remark that the results depend on the size of the cluster if we compare the results obtained for $n_s = 5$ to those obtained for $n_s = 4$.³³ We have performed here calculations with a fixed number of atoms per cluster and unfortunately we cannot at present do a calculation with an increasing cluster size with decreasing temperature, as observed experimentally. However, such sequence of phases can present a first account for the experimental percolative CSG-IFM transition observed in disordered Cerium alloys.^{5,33}

4. The Effect of a Random and Transverse Magnetic Field

The last calculation that we have recently performed concerns the different effects of random fields on the global phase diagram already obtained in Ref. 23. The main motivation of this new type of disorder is that for intermediate doping in the $CeNi_{1-x}Cu_x$ alloys, the hysteresis curve exhibits cycles with sharp macroscopic jumps in the magnetization at low temperatures.³⁵ Recently, these jumps have been attributed to the presence of random anisotropies.³⁵ When these anisotropies are

very strong, they can be related with the presence of random fields.³⁴ Therefore, it is added to the Hamiltonian given in Eq.1 a random field h_i following a gaussian distribution as shown below:

$$P(h_i) = \left[\frac{1}{2\pi\Delta^2} \right]^{1/2} \exp \left[-\frac{h_i^2}{2\Delta^2} \right]. \quad (7)$$

The results found for the phase diagram³⁷ indicate that the random field can disrupt all magnetic long range orderings including the SG phase. Moreover, at lower temperature, it is also found that it is necessary to take larger values of the Kondo interaction in order to find a Kondo solution. In that sense, we can say that the presence of a random field also suppresses the Kondo singlet formation. Another interesting result related to the presence of a random field is the coexistence of the Kondo state and the so called independent spin phase. This phase represents a magnetic state in which the localized spins, even remaining uncorrelated, are oriented randomly since they couple locally with the random field.³⁸

5. Conclusion

We have discussed in this work the competition between Kondo effect, magnetic order and spin glass which occurs in some Cerium disordered alloys. The experimental results have shown clearly that this competition can produce several non-trivial results as deviations of the Fermi liquid behaviour or cluster glass formation. From the theoretical point of view, the effects of disorder have been studied using the Kondo lattice model with an Ising intersite term by choosing several types of random distribution for the intersite spin coupling J_{ij} . This model has been also extended to include clusters of spins and finally, also random fields to provide a better description of the experimental scenario found in, for instance, $CeNi_{1-x}Cu_x$ alloys.

Although there was a considerable amount of theoretical results found already for the competition between Kondo effect, magnetic order and spin glass, some issues still remain unsolved. For example, a formulation in terms of spins clusters instead of canonical ones able to explain the effects of random fields in the mentioned competition is not done. Another very interesting open question is a proper treatment of the Kondo effect within the Underscreened Kondo Lattice to study the interplay between Kondo effect and spin glass as observed in some Uranium disordered alloys.

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