Theoretical models for a complex magnetic system: The case of CeNi\textsubscript{1-x}Cu\textsubscript{x}

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Abstract. Special features in the phase diagram and in the low temperature hysteresis cycles have been experimentally observed in the complex CeNi\textsubscript{1-x}Cu\textsubscript{x} system. We present also here theoretical approaches, based firstly on a Kondo lattice model which describes the coexistence between a spin glass or a cluster glass state, the Kondo regime and the ferromagnetic ordering. The second model is a Monte Carlo simulation on a 3D lattice with clusters and random anisotropy and reproduces the existence of steps in the magnetizations cycles at very low temperatures. The theoretical results are compared with the experimental data of the very complex magnetic behaviour of CeNi\textsubscript{1-x}Cu\textsubscript{x} alloys. In particular, we can account for the existence of a cluster spin glass state which changes continuously into an inhomogeneous ferromagnetic phase at very low temperatures.

1. Introduction
There has been an extensive study of the competition between different interactions, such as the indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction, the large Kondo 4f-conduction band hybridization or the strong crystalline field effect in strongly correlated f-electron systems and many very interesting physical phenomena, like heavy fermions [1,2], non-Fermi Liquid behavior [3] or quantum criticalities [4], have been studied in detail from both an experimental and a theoretical approach. Moreover, some alloys show an additional disorder and the simultaneous existence of competing magnetic interactions and disorder can give rise to a rich variety of magnetic behaviors related to spin-glass phases, clustered magnetic states or super-paramagnetism and also to many theoretical approaches, like the study of the Griffiths phases or the Non Fermi Liquid State [5, 6, 7]. One of the more representative examples of such a situation is the well studied alloys CeNi\textsubscript{1-x}Cu\textsubscript{x}, where the change of the matrix concentration acts as a chemical pressure and gives different magnetic behaviors. In this paper, we will review briefly the different experimental facts which establish now...
the existence of a percolative transition from a cluster glass state to an inhomogeneous ferromagnetic state and then we will summarize recent theoretical approaches to account for the magnetic phase diagram and the peculiar magnetization cycles observed at very low temperatures.

2. CeNi$_{1-x}$Cu$_x$: Experimental phase diagram.
The main objective of this study was the investigation of the very peculiar behavior of the CeNi$_{1-x}$Cu$_x$ alloys close to the Quantum Critical Point expected around $x = 0.2$. First experimental studies down to 2K lead to propose a ground-state phase diagram represented in Figure 1 [8]. The magnetic interactions are modified by substitution of Cu by Ni, then decreasing the number of conduction electrons and giving a change from antiferromagnetism (AFM) (CeCu) to ferromagnetism (FM) (well characterized by neutron diffraction in CeNi$_{0.4}$Cu$_{0.6}$ [9]), as observed in other similar RNi$_{1-x}$Cu$_x$ series, which is evolving towards the disappearance of the long-range magnetic order around CeNi$_{0.8}$Cu$_{0.2}$.

Also, a “spin-glass-like” phase was found at temperatures above the ferromagnetic order state. This experimental phase diagram was firstly compared to the classical spin-glass model considering a weakly correlated disorder with randomness in the interactions and frustration [10] (see inset of Figure 1) or accounted for by the Kondo-spin glass model presented later on [11]. That picture proposed the existence of a mixed phase (coexisting spin-glass and FM) that would evolve into spin-glass for increasing temperatures, before becoming paramagnetic. It should be pointed out that such models have provided only a qualitative explanation of the experimental phase diagram. However, theoretical phase diagrams for metallic spin glasses in strongly correlated systems deal mainly with the behavior near the zero quantum critical point, which required the study of the very low-temperature behavior, where quantum effects should predominate.

Further experimental study down to the very low temperature range for the compositional region near the magnetic-non magnetic crossover close to the CeNi compound (around $x = 0.2$) has shed light on the magnetic ground state evolution along the series: (i) measurements of the ac susceptibility, dc magnetization and specific heat down to very low temperature (~ 100 mK) show that a cluster-glass state is formed at a freezing temperature $T_f$ well above the ferromagnetic state characterized by neutron diffraction [12,13]. However, no indication of Curie temperature, $T_c$, associated to the onset of long-range ferromagnetic order was observed by any of these techniques. (ii) Muon spin relaxation ($\mu$SR) indicate the formation of an intermediate inhomogeneous magnetic state developing above $T_f$ and consisting of dynamic spin clusters, which become static at $T_f$. These new results allowed us to propose a percolative process in order to describe the emergence of the long-range ferromagnetic state from the cluster-glass one below $T_f$. According to such a mechanism, the size of the magnetic clusters would increase below $T_f$, leading to a domain-like ferromagnetic state at very low temperature [14]. This behavior has been confirmed by two kinds of experiments: Small Angle Neutron Scattering (SANS) measurements, which demonstrate the existence of magnetic clusters of around 20 Å below $T_f$ and, very low temperature (of order 100 mK) hysteresis loops obtained in the ferromagnetic state, presenting steps of the magnetization. These steps have been attributed to the avalanches of domain flips, as a mesoscopic analogue of the Barkhausen noise [15].

Considering the whole analysis of the results, we present a revised magnetic phase diagram for the CeNi$_{1-x}$Cu$_x$ (Figure 2). The new information provided by the latter diagram can be summarized as follows: (i) The presence of an intermediate magnetic state as temperature decreases from the paramagnetic state, where dynamic clusters develop below $T^*$. The volume fraction of these clusters increases as temperature is lowered and they become frozen at $T_f$. (ii) Since no sharp transition is detected at a Curie temperature $T_c$, the border defining the ferromagnetic state cannot be precisely determined. The gradually increase in the FM correlation length $\zeta$ when lowering the temperature below $T_f$ is illustrated in the phase diagram by a gradation in colour. (iii) The complete evolution of $T_f$ has been determined along the series, even close to the CeNi side.
3. Theoretical models for CeNi$_{1-x}$Cu$_x$ alloys.

The very interesting case of CeNi$_{1-x}$Cu$_x$ alloys has been extensively studied, because, in addition to the competition between the Kondo effect and the magnetic order in the lattice, disorder and anisotropy effects are very important and give rise to a spin glass or a cluster glass state. From a theoretical point of view, several theoretical models have been successively built in order to improve the description of more precise experimental results. The first model is a Kondo-Ising Lattice (KIL) model with an additional Ising intersite interaction between localized spins [11] and with the disorder treated in the Sherrington-Kirkpatrick (SK) model [16]. The first approach has been then extended to include a ferromagnetic (FM) ordering and a global phase diagram Temperature versus the strength of Kondo coupling $J_K$ has been obtained displaying a SG phase, a Kondo one and an additional FM ordering [17]. A review of these models is available in ref. [2].

Recently, a new approach has been proposed by using a random site model to describe the intersite interaction, which gives a more local description of the disorder, as introduced by Amit [18,19]. This model improves the description of the experimental phase diagram of the CeNi$_{1-x}$Cu$_x$ system, because it yields an inhomogeneous disordered ferromagnetic ordering below the spin glass phase [19]. However, further theoretical improvements to describe the magnetic clusters with both the Kondo effect and the disorder are in progress in CeNi$_{1-x}$Cu$_x$ alloys and preliminary calculations have been performed to study the competition between the Kondo effect and a cluster glass phase within the Kondo lattice model with both an inter-cluster random Gaussian interaction and an exact diagonalization inside each cluster. The calculations, which have been done for clusters with small and constant sizes, show that the spin glass order parameter and the Kondo correlation function have completely opposite variation versus $J_K$ and can, therefore, give a good description of the cluster glass-ferromagnetic coexistence [20].

Another model has been recently developed to reproduce the multistep pattern observed in the hysteresis loops of CeNi$_{1-x}$Cu$_x$ alloys [15,21]. This model describes a simple cubic lattice of magnetically coupled Ising spins divided in clusters of random sizes. Within this model, the temperature effects are not restricted to the thermal activation of the spins, but a phenomenological description of the percolative process is also considered. The model considers a three dimensional Ising system on a simple cubic lattice. The lattice is separated in clusters of random mesoscopic size
and each cluster is characterized by a random anisotropy direction. The interaction between the spins is mainly ferromagnetic but one allows a given concentration of antiferromagnetic interactions to represent the disorder and the fact that the magnetic order is different in the limit of high Ni (small x) or high Cu concentrations. The model describes a lattice of Ising spins, which represent a system of spins $\frac{1}{2}$. This situation corresponds to a Ce$^{3+}$ ion in an orthorhombic environment, as its $J = 5/2$ ground state is split into three doublets and therefore only one doublet is relevant at very low temperatures. The spins interact among them through the magnetic interactions which can be ferromagnetic or antiferromagnetic. The lattice is divided in clusters of random sizes, and each cluster, n, is characterized by an anisotropy field $A_n$, the absolute value of which is assumed to be constant, while the orientation with respect to the z-axis is random. This model is able to well describe the jumps in the magnetization at very low temperature described in ref. [15], but also it accounts for the smooth-off of these jumps at higher temperatures as a combined effect of the thermal activation of the spins together with the percolation of the clusters for decreasing temperature [21].

4. Conclusions.

Thus, the CeNi$_{1-x}$Cu$_x$ alloys represent a very interesting and original case where there is a strong competition between the Kondo effect, the ferromagnetic ordering and the disorder. A highly developed experimental study has evidenced for a percolative transition from a spin cluster glass to an inhomogeneous ferromagnetic ordering with decreasing temperature for concentrations close to the Quantum Critical Point around $x = 0.2$. Several theoretical works have been developed in order to account for the global phase diagram of this system and to describe the percolation of clusters at very low temperatures.

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