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Spin-glass, antiferromagnetism and Kondo behavior in $Ce_2Au_{1-x}Co_xSi_3$ alloys

SUBHAM MAJUMDAR[1], E V SAMPATHKUMARAN[1,*], ST BERGER[2], M DELLA MEA[2], H MICHOR[2], E BAUER[2], M BRANDO[3], J HEMBERGER[3] and A LOIDL[3]

[1]Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai 400 005, India

[2]Institute for Experimental Physics, Vienna University of Technology, A-1040 Wien, Austria

[3]Experimentalphysik V, Electronische Korrelationen und Magnetismus, Institut für Physik, Universität Augsburg, D-86135 Augsburg, Germany

*Email: sampath@tifr.res.in

Abstract. Recently, the solid solution $Ce_2Au_{1-x}Co_xSi_3$ has been shown to exhibit many magnetic anomalies associated with the competition between magnetic ordering and the Kondo effect. Here we report high pressure electrical resistivity of Ce_2AuSi_3, ac susceptibility (χ) and magnetoresistance of various alloys of this solid solution in order to gain better knowledge of the magnetism of these alloys. High pressure resistivity behavior is consistent with the proposal that Ce_2AuSi_3 lies at the left-hand side of the maximum in Doniach's magnetic phase diagram. The ac χ data reveal that there are in fact two magnetic transitions, one at 2 K and the other at 3 K for this compound, both of which are spin-glass-like. However, as the Co concentration is increased, antiferromagnetism is stabilized for intermediate compositions before attaining non-magnetism for the Co end member.

Keywords. $Ce_2Au_{1-x}Co_xSi_3$ alloy; spin-glass; antiferromagnetism; Kondo behaviour; magnetoresistance.

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1. Introduction

The study of the consequences of the competition between the Kondo effect and magnetic ordering in Ce-based systems continue to be an active topic of research. In this regard, there is a recent theoretical prediction [1] that disordered Kondo lattice alloys may be spin-glasses, particularly near the quantum critical point (QCP) in addition to non-Fermi liquid characteristics. The predictions of this theory were also substantiated [1] by experimental evidences based on a few other solid solutions, viz., $CeNi_{1-x}Cu_x$ and $CeCoGe_{3-x}Cu_x$. It is therefore of interest to carefully investigate more such solid solutions in order to arrive at a global understanding of such predictions. In this respect, we considered it worthwhile to probe the recently discovered [2,3] solid solution, $Ce_2Au_{1-x}Co_xSi_3$, in more detail. With this primary motivation, we have carried out ac susceptibility (χ) and magnetoresistance (MR) measurements on selected compositions of this alloy series at low temperatures.

In addition, we have carried out high pressure electrical resistivity (ρ) measurements on Ce_2AuSi_3 to throw light on some of the earlier conclusions [3].

These alloys, crystallizing in a AlB_2-derived hexagonal structure, have been found to exhibit many interesting magnetic characteristics [3]. While the parent Au compound magnetically orders below 4 K, Ce_2CoSi_3 is a non-magnetic Kondo lattice. As Au is progressively replaced by Co, the magnetic ordering temperature (T_0) goes through a maximum (around 7 K) as a function of x thereby suggesting that this alloy series is one of the few solid solutions spanning a wide range of Doniach's magnetic phase diagram [4]. For some intermediate compositions, two magnetic transitions were observed with the data revealing interesting changes in magnetic structure both as a function of x and T. Thus, this alloy series offers several situations to test the theory of [1].

2. Experimental

The samples employed were synthesized as discussed in [3] and characterized by X-ray diffraction. For Ce_2AuSi_3, high pressure ρ measurements in the temperature range 1.5–30 K were carried out in a hydrostatic pressure medium up to 17 kbar. For many alloys ($x =$ 0.0, 0.2, 0.3, 0.4 and 0.6), ac χ behavior (1.7–15 K) was probed employing a commercial magnetometer at various frequencies (1, 10, 100, 1000 Hz); the ρ behavior as a function of externally applied magnetic field (H) up to 12 T were also tracked at low temperatures (0.6, 2, 5 and 10 K).

3. Results and discussion

In figure 1, we show the ac χ data as a function of (logarithmic) T below 12 K for $x =$ 0.0. There is a peak at 3 K consistent with the observation in the heat capacity (C) and dc χ data [3] due to the onset of magnetic ordering. The new finding is that there is an additional peak at about 1.2 K (which could not be tracked in earlier studies [2,3] due to non-accessibility of this T range), establishing the existence of another magnetic transition. In fact two transitions are expected [5] from two crystallographically inequivalent Ce sites for an ordered arrangement of Au and Si ions. There is a frequency dependence of ac χ for both the transitions, which suggests that this material is a spin-glass.

In order to further substantiate our earlier proposal [2] that this compound lies at the low coupling limit, we show in figure 2 the ρ data (normalized to 2 K value) below 20 K at various external pressures. There is a drop in ρ setting in around 10 K, well above T_0 due to the well-known [6] interplay between indirect exchange interaction and the Kondo effect in magnetically ordered Kondo lattices. The main point of emphasis is that there is a shoulder at about 5 K, which arises as an artifact of long range magnetic ordering setting in at 3 K; this shoulder moves to a higher temperature with increasing pressure, say to 6 K at 17 kbar. If this compound lies at the peak or at the right-hand side of the Doniach's diagram, one would have expected that this shoulder goes to a lower T with increasing pressure. Thus this data may be consistent with our earlier proposal that this compound is situated at the left-hand side of the Doniach's magnetic phase diagram. This naturally means that, with the Co end member being a non-magnetic Kondo lattice, the present solid solution spans a wide range of 4f-conduction band coupling strength.

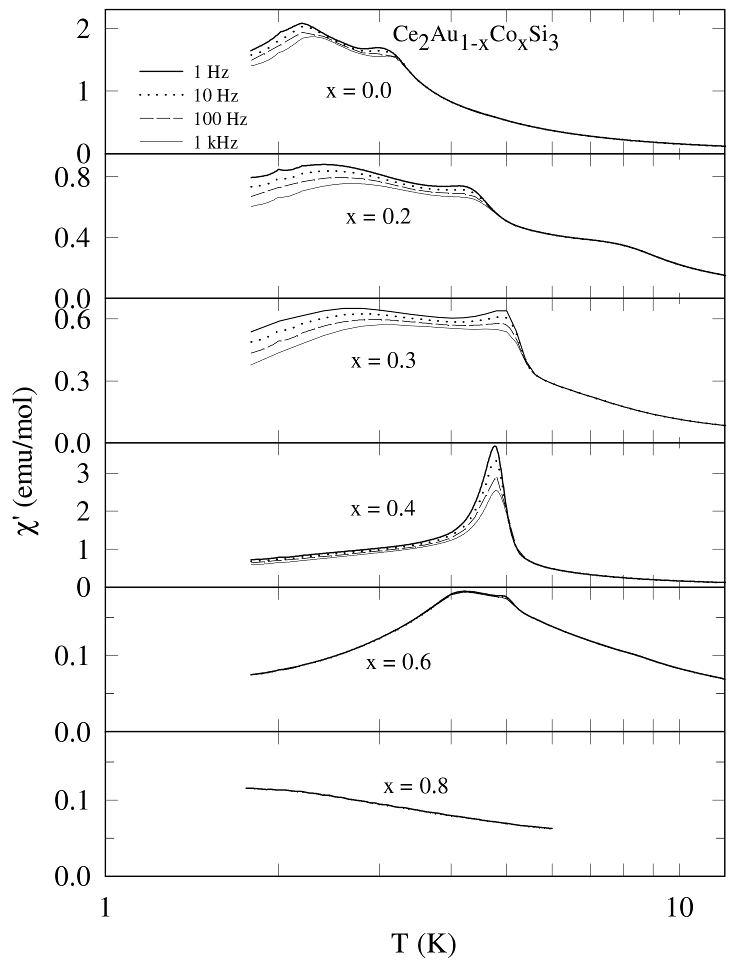

Figure 1. ac Susceptibility (real part) as a function of logarithmic temperature (below 12 K) for the alloys, $Ce_2Au_{1-x}Co_xSi_3$, at various frequencies.

We now address the question whether any of these disordered alloys is a spin-glass as predicted in [1]. For this purpose, we show in figure 1 the frequency dependence of ac χ for several compositions ordering magnetically in the T range of ac χ investigation. It has already been stated that the parent Au compound is a spin-glass. The alloy $x = 0.2$ exhibits two magnetic transitions, one at about 8 and the other at 3 K, manifesting as a broad shoulder and a peak in ac χ, respectively; in addition, there is another peak at about 1.5 K, the origin of which could be due to spin reorientation effects or chemical inhomogeneities; while there is absolutely no frequency dependence for the 8 K transition, there is a marginal decrease of χ near the peak temperature with increasing frequency as in the case of $x = 0.0$. Similar features are seen for $x = 0.3$ with two prominent transitions at 4 and 1.5 K, respectively, due to crystallographically inequivalent Ce sites. For $x = 0.4$, however, there is only one spin-glass-like magnetic transition at 5 K agreeing with C and

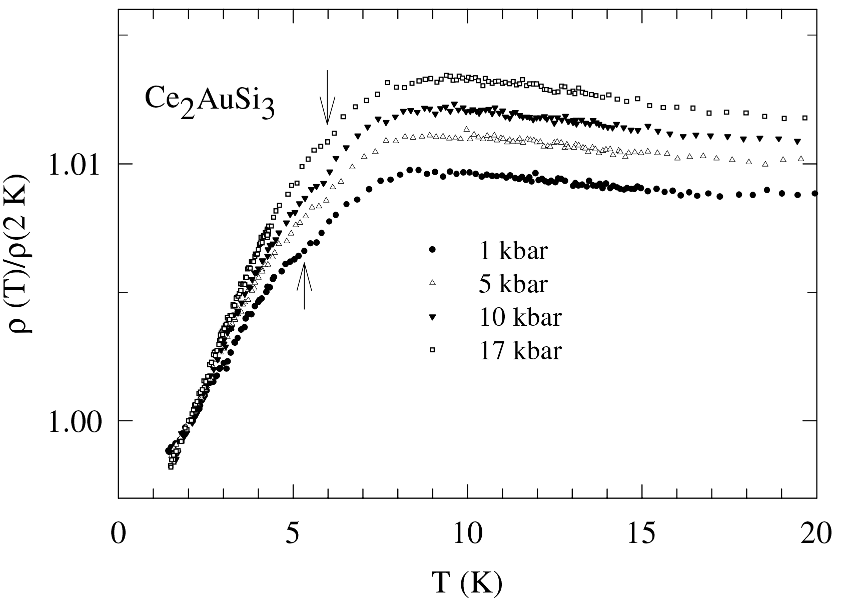

Figure 2. Electrical resistivity as a function of temperature below 20 K at various externally applied pressures for Ce_2AuSi_3. Vertical arrows mark the feature due to magnetic ordering.

dc χ data [3]. For $x = 0.6$, not only the peak temperature (4.5 K) but also the values are frequency independent indicating that this alloy is not a spin-glass. For $x = 0.8$, χ tends to show a flattening below 2.5 K, as though there is a tendency for magnetic ordering at a lower temperature.

We have also measured the magnetoresistance (MR) ($=[\rho(H) - \rho(0)]/\rho(0)$) as a function of H for many of these alloys in the vicinity of T_0. The results at 2 and 5 K and in some cases at 0.6 K, that is, in the vicinity of T_0, are shown in figure 3. The MR is negative for compositions $x < 0.6$, which is not inconsistent with spin-glass behavior. The ρ of the alloy $x = 0.8$, which is close to the QCP, and that of $x = 0.6$, however exhibits an increase for initial H below their respective T_0 (see for instance, 0.6 K data), characterizing that these alloys are antiferromagnets without magnetic Brillouin-zone gap effects. The fact that these compositions undergo long-range magnetic ordering is established by the observation of a well-defined heat-capacity anomaly at the transition temperature (not shown here due to space limitations), clearly revealing that the alloys (that is, as one approaches the QCP) are definitely not spin-glasses. The sign reversal at higher fields establish field-induced spin-reorientation effects. It should be mentioned that there are also qualitative changes in the shape of the plots as T (from 2 to 5 K) and x are changed, besides a sign change for $x = 0.6$ and 0.8 as H is varied, say, at 0.6 K. This observation suggests that there are changes in the magnetic structure both as a function of T as well as of x in this series.

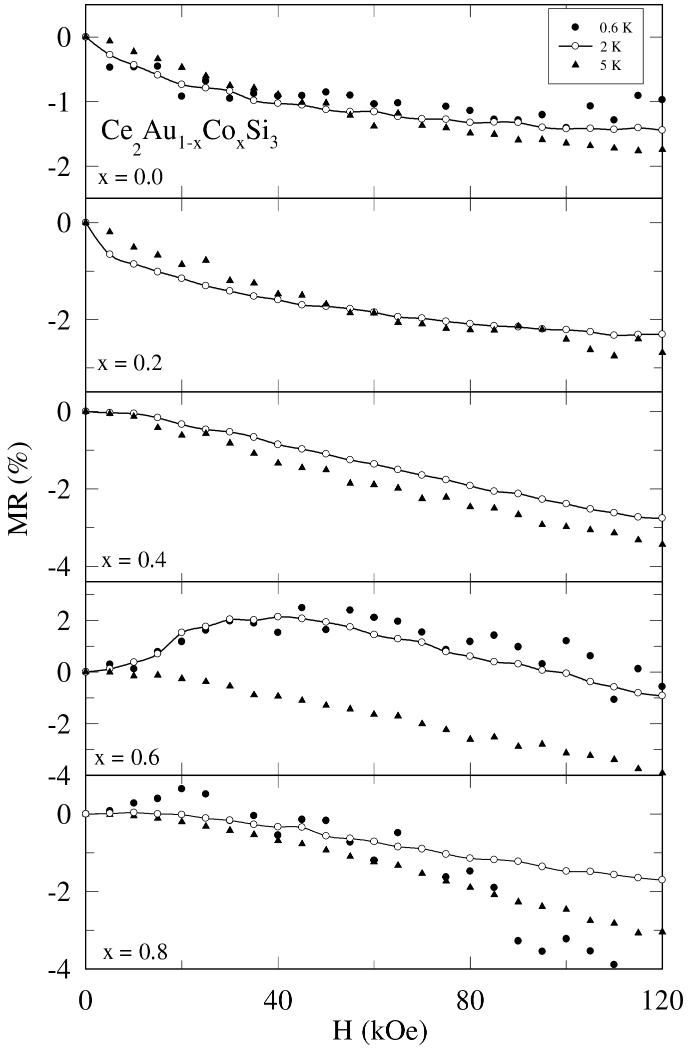

Figure 3. Magnetoresistance, $(=[\rho(H) - \rho(0)]/\rho(0))$ as a function of externally applied magnetic field at selected temperatures in the vicinity of magnetic ordering temperatures for $Ce_2Au_{1-x}Co_xSi_3$. The error bar in the values of MR is ±0.5%. The lines drawn through the data points serve as guides to the eyes.

4. Conclusion

To conclude, the results bring out rich magnetic behavior of this new class of Ce-based ternary alloys establishing an interplay between spin-glass, antiferromagnetism and the non-magnetic Kondo lattice behavior as the composition is varied.

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