AC SUSCEPTIBILITY BEHAVIOUR OF $Ce_2Pd_{1-x}Co_xSi_3$*

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We report the results of ac susceptibility (χ) measurements for various compositions of the series, $Ce_2Pd_{1-x}Co_xSi_3$, in the polycrystalline form, which has been previously reported to undergo a transformation from magnetic ordering to non-magnetic Kondo lattice behaviour as x is varied from 0 to 1. A finding of emphasis here is that the features in ac χ data for the compositions, $x = 0.6$ and 0.7, in the close vicinity of quantum critical point (QCP), are typical of long range magnetic ordering systems, whereas the compositions far away from QCP at the Pd-end including Ce_2PdSi_3 exhibit spin–glass characteristics. This trend is different from hitherto known behaviour among Kondo alloys.

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It has been generally assumed in the literature that there may be an intimate relationship between disorder, SG ordering and NFL behaviour at QCP. In this article, we present ac magnetic susceptibility ($\chi_{\rm ac}$) behaviour of a pseudo-ternary series, $Ce_2Pd_{1-x}Co_xSi_3$ (Ref. [2]), crystallising in a AlB_2-derived hexagonal structure. The results suggest that the SG behaviour as revealed by the features in the ac χ data is actually seen only if one moves away from QCP in this series in contrast to hitherto known trends among Kondo lattices [1].

For previous work on these alloys, the readers may see Refs. [1–5]. The results on the pseudo-ternary alloys revealed that there is a transformation from magnetic ordering for $x = 0.0$ to non-magnetic heavy fermion behaviour for $x = 1.0$. The magnetic ordering temperature ($T_{\rm o}$) falls [7] in the region 2 to 4 K (Ref. [2]) for $x<0.7$ reducing to a value close to 2.3 K for $x = 0.7$; for $x = 0.8$, $T_{\rm o}$ (if present) apparently dips to a value well below 2 K. Given [2,7] that Ce_2CoSi_3 is non-magnetic, it is clear that QCP lies at a composition

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close to 0.8. In other words, gradual Co substitution for Pd results in a shift towards QCP in this alloy series.

The samples, $Ce_2Pd_{1-x}Co_xSi_3$ (x = 0.0, 0.2, 0.4, 0.6, 0.7), employed here are the same as those in Ref. [1]. The temperature (T) dependence (1.8–20 K) of real and imaginary parts of ac susceptibility (χ' and χ'', respectively,) were obtained by a commercial superconducting quantum interference device (Quantum Design) (ac driving field 1 Oe) at several frequencies (1, 10, 100 and 1000 Hz).

The results of our measurements are shown in Figs. 1 and 2. It is obvious from Fig. 1 that ac χ' tends to peak for all the compositions and the peak positions (about 2.7, 3.3, 3.3, 3.1 and 2.3 for $x = 0.0$, 0.2, 0.4, 0.6 and 0.7, respectively,) mark the appearance of magnetic ordering in all these alloys [6]. For $x = 0.7$, the onset of magnetic ordering at 2.3 K is manifested as a tendency of χ to flatten both in ac χ' (Fig. 1) and dc χ (Ref. [1]). In order to understand whether the magnetic transitions are of a long range or of a SG character, it is important to look at the frequency dependence of ac χ' as well as the behaviour of χ''. It is now well established that long range magnetic ordering (LRMO) systems do not exhibit any frequency dependence of ac χ at T_o, unlike SG systems which show a marginal shift of the curves (as indicated by a reduction of the χ' values below the peak temperature) towards high temperature with increasing frequency with a well-defined cusp in the χ'–T plot at T_o; in addition, χ'' of SG systems may exhibit a sharp prominent upturn at T_o as T is lowered, a feature that is absent in LRMO systems.

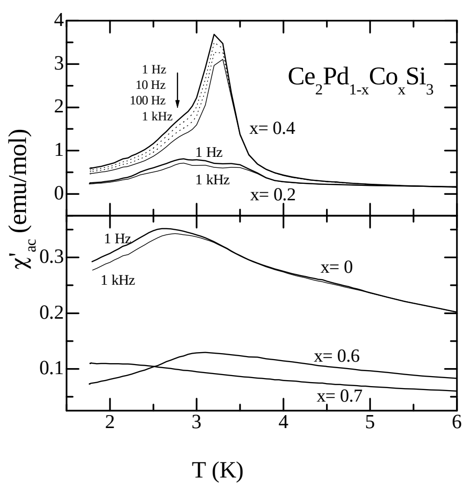

Fig. 1. Real part of ac susceptibility of the alloys, $Ce_2Pd_{1-x}Co_xSi_3$, as a function of temperature at various frequencies. For $x = 0.6$ and 0.7, the curves at all the four frequencies overlap. For the sake of clarity, the data points have been omitted and a line through the data points is only shown for each curve.

With these criteria in mind, one can make relevant conclusions looking at Figs. 1 and 2. Though we have collected the data at four frequencies, for the sake clarity, we show the plots in Fig. 1 at two frequencies only for $x = 0$ and 0.2. For $x = 0.6$ and 0.7, the plots at the four frequencies overlap with each other even in the magnetically ordered state. Thus, these two alloys are definitely not spin-glasses; as a firm support, there is no upturn in χ'' at T_0; therefore, these two compositions, which are closer to QCP, can be classified as LRMO systems. The magnetic structure of these compositions are presumably of an anti-ferromagnetic type and not of a ferromagnetic type as indicated by the isothermal M behaviour at 2 K and dc χ behaviour reported in Ref. [1].

As one decreases x to 0.4, there is a well-defined cusp in χ' at T_0 (Fig. 1, top). Apparently there is a significant frequency dependence of the values of χ' as well at a given T below and near the peak temperature. In addition, there is an upturn of χ'' below 3.5 K, which is most prominent for this composition (see Fig. 2). χ'' is also found to show marked frequency dependence as in the case of χ'; however, for the sake of clarity, we have shown the plot only at one frequency (1 Hz). These features reveal that this alloy undergoes SG freezing below T_0. As the composition is varied towards Pd rich end, say for $x = 0.2$, the peaks are broadened (Figs. 1 and 2) with an observable, but a relatively weak, frequency dependence. The features, particularly in χ'' are, however, relatively suppressed for $x = 0.0$. We would like to add that ac χ measurements were carried on the single crystals (same crystal as in Ref. [3]) and we are able to see spin-glass anomalies not only in χ' but also in χ'' (not shown here); in accordance with this, we have also noted the separation of zero-field-cooled and field-cooled dc χ curves ($H = 100$ Oe) below about 2.3 K in this crystal. Hence, we believe that these features are intrinsic to this compound and it, therefore, appears that the feature in χ'' in polycystals may be sometimes suppressed due to random orientations of the crystallites. It may be recalled that neutron diffraction pattern [3], however, revealed the features due to anti-ferromagnetic ordering, but not extending to the entire crystal for $x = 0.0$; magnetic cluster size has been found to be of the order of 100 Å. Therefore, we believe that the frequency dependence, particularly for the Pd end member, arises from some degree of randomness of magnetic coupling among such clusters. In short, the Pd-rich alloys may be classified as cluster spin-glass systems.

To conclude, the results presented in this article suggest that, in the series, $Ce_2Pd_{1-x}Co_xSi_3$, the compositions which are far away from QCP, appear to behave like (cluster) spin-glasses. It is thus interesting to note that, as one approaches QCP, the SG freezing in fact disappears in favour of LRMO. It is of interest to perform neutron diffraction measurements to verify this observation considering its implications. [8]

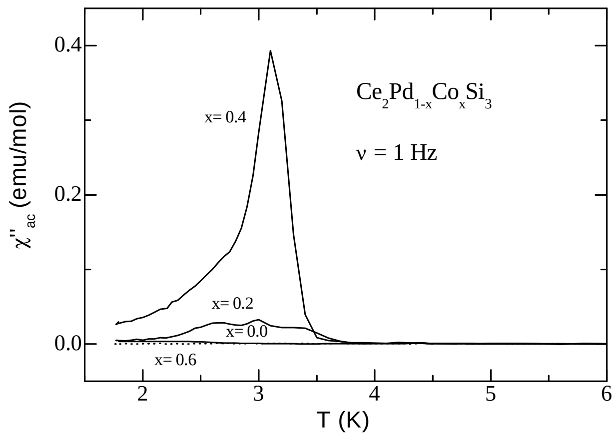

Fig. 2. Imaginary part of ac susceptibility of the alloys, $Ce_2Pd_{1-x}Co_xSi_3$, as a function of temperature measured at a frequency of 1 Hz. For $x = 0.6$ and 0.7, the curves overlap. For the sake of clarity, the data points have been omitted and a line through the data points is only shown for each curve.

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