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Disorder effects on Kondo behavior in $CePt_{2+x}$

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Abstract

$CePt_2$ is an antiferromagnetic (AF) Kondo-lattice compound with T_N ~1.7 K and T_K ~4.6 K. The evolution of AF and Kondo interactions in $CePt_{2+x}$ with x= 0, 0.5 and 1 is observed by analysis of the temperature-dependent specific heat. This analysis shows that ~56% of the Ce in $CePt_2$ is involved in Kondo interactions, with the rest involved in magnetic correlations. While 100% of the expected entropy (Rln2) is recovered by 15 K in $CePt_2$, only 95% and 93% is recovered for x=0.5 and 1.0, respectively. Meanwhile a larger Kondo fraction (80-90%) and a smaller AF fraction (15-3%) is observed, while T_K decreases from 4.6 K to 2.8 K as x increases from 0 to 1. This trend in T_K is opposite that expected from the measured lattice contraction. We conjecture that lattice disorder induced by Pt alloying is responsible for these results.

Keywords: Kondo lattice, Antiferromagnetic order , Disorder

1. Introduction

It is known that nonmagnetic substitutions can suppress the magnetic ordering and enhance Kondo effects in Ce (U) based compounds. For instance Ce_3Al is an antiferromagnet with T_N=2.5 K, but with 50% La substitution for Ce, it becomes a heavy fermion with γ= 1000 mJ/ mol Ce K^2 [1]. K. A. Gschneidner *et. al.* pointed out that large heat capacities can arise from non-magnetic atom disorder (NMAD) in compounds where Ce (U) atoms occupy a periodic lattice [2]. In order to clarify the role of disorder in determining physical properties, Pt alloying in $CePt_{2+x}$ has been studied. $CePt_2$ is a cubic antiferromagnet (T_N =1.7 K) that grows in the C15 ($MgCu_2$) Laves phase. Neutron diffraction and x-ray spectra confirm that $CePt_{2.5}$ and $CePt_3$ alloys are single phase with the C15 and C15b structures respectively [3]. Alloying Ir or Rh on the Pt-sites in $CePt_2$ results in a decrease of T_N [4]. It is of interest to see how substitutional disorder influences a system like $CePt_{2+x}$ in which the magnetic order and Kondo interactions evolve as x is varied from 0 to 1. The present study was therefore performed on $CePt_{2+x}$ with x=0-1 through the measurements of specific heat at 0.4 to 30 K.

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2. Experimental details

Polycrystalline samples of bulk $CePt_{2+x}$ for x= 0, 0.5 and 1 were prepared by arc melting high-purity constituent elements in an argon atmosphere. Cu K_α X-ray diffraction measurements demonstrate that these alloys have the Laves–phase structure and no visible impurity phase (Fig. 1). The specific heat was measured in the range 0.4-30 K using a thermal-relaxation microcalorimeter in a ^{3}He refrigerator, with the mg-pellet sample attached to a sapphire holder on which a RuO_2-Al_2O_3 film thermometer

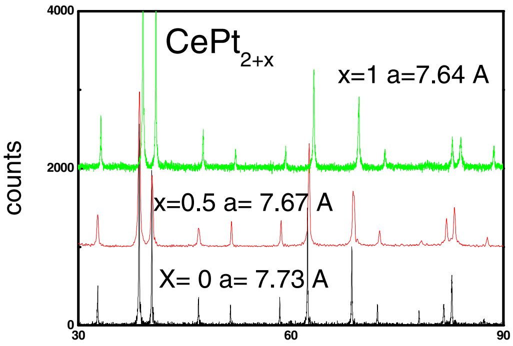

FIG. 1 X-ray 2θ(degree) diffraction patterns for $CePt_{2+x}$ with x=0,0.5 and 1.

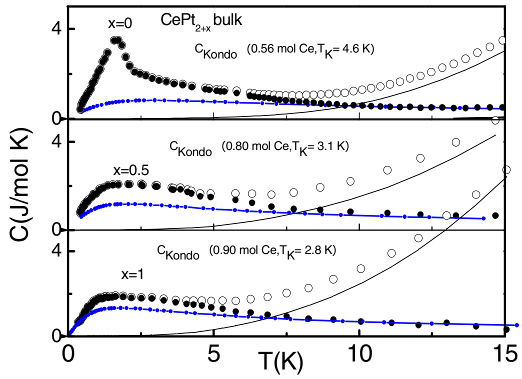

FIG. 2 Specific heat for $CePt_{2+x}$ with x = 0, 0.5 and 1 (open circles), other notations are lattice phonon C_{ph} (solid lines), C(T)-C_{ph} (solid circles), and Kondo model fit (dashed lines)

and a Ni-Cr heater were deposited.

3. Result and analysis

Diffraction measurements indicated that the lattice constant *a* decreases from 7.73 Å to 7.64 Å as x increases from 0 to 1 (Fig. 1). The decrease of lattice constant is likely due to the smaller ionic size of Pt compared to Ce. The increased line width of $CePt_{2.5}$ and $CePt_3$ relative to $CePt_2$ reflects increased lattice disorder and the increasing number of Pt atoms on the normally Ce sites.

The temperature dependence of the specific heat for T= 0.4 -15 K is shown in Fig. 2. These results are in good agreement with an earlier report [3], although the present data extend to lower temperatures. For $CePt_2$ a sharp peak near 1.7 K is superimposed with a low-temperature bump, reflecting the coexistence of Kondo interactions and magnetic correlations; the profiles of alloys $CePt_{2.5}$ and $CePt_3$ do not have as clear a peak as $CePt_2$, instead exhibiting stronger Kondo-like anomalies. We account for the specific heat of $CePt_2$ by the contribution of the lattice phonon C_{ph}, magnetic correlations C_m, Kondo interactions C_K and crystal field splittings C_{cry}. Since the crystal field splitting T_{CF} in the alloys is ~ 200-300 K, for T < 15 K its contribution is obviously insignificant [3]. After lattice phonon subtraction referred to separate measurements on their non-magnetic counterparts $LaPt_{2+x}$, the integrated entropy of magnetic contribution $S = \int (C/T)\, dT$ is found to be about Rln2 between 0 to 15 K, the result is consistent with a Γ_7 doublet with S=1/2 for one mole of trivalent cerium [2]. The individual contributions of the Kondo interactions and magnetic correlations were further resolved by fitting high temperature data to a Kondo model (T_K=4.6 K for 0.56% Ce) with the assumption of negligible contribution of magnetic correlations for T > 8 K.

Applying this same data analysis to $CePt_{2.5}$ and $CePt_3$ the entropy of $CePt_{2.5}$ and $CePt_3$ are estimated to be 0.95 and 0.93 Rln2 respectively, possibly indicating the appearance of some nonmagnetic Ce^{4+} or that some ceriums participate in higher-temperature interactions. The contributions of Kondo interactions C_K were obtained for $CePt_{2.5}$ with T_K=3.1 K for 80 % Ce and for $CePt_3$ with T_K=2.8 K for 90% Ce. Pt alloying in $CePt_{2+x}$ from x= 0 to 1 not only enhances the fraction of ceriums involved in Kondo interactions from 56 % to 90%, but also suppresses the fraction involved in magnetic correlations from 44% to 3%. The enhancement of Kondo interactions with increasing x explains the more Kondo-like profile of $CePt_{2.5}$ and $CePt_3$. Furthermore, Pt alloying effects on decreasing T_K from 4.6 K (x=0) to 2.8 K (x=1) are also revealed. Since this latter trend is opposite to that expected from the measured lattice contraction, site exchange of Ce-Pt and structural variation from C15 -C15b created by Pt alloying are factors attributed to the consequences.

Acknowledgment

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