

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

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Abstract

CePt_2 is an antiferromagnetic (AF) Kondo-lattice compound with $T_N \sim 1.7\text{ K}$ and $T_K \sim 4.6\text{ K}$. 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 $\sim 56\%$ of the Ce in CePt_2 is involved in Kondo interactions, with the rest involved in magnetic correlations. While 100% of the expected entropy ($R \ln 2$) 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 ($80\text{--}90\%$) and a smaller AF fraction ($15\text{--}3\%$) 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.

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PACS: 65.80.+n; 72.15.Qm; 75.30.Mb

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 $T_N = 2.5\text{ K}$, but with 50% La substitution for Ce, it becomes a heavy fermion with $\gamma = 1000\text{ mJ/mol Ce K}^2$ [1]. Gschneidner et al. [2] pointed out that large heat capacities can arise from non-magnetic atom disorder (NMAD) in compounds where Ce (U) atoms occupy a periodic lattice. 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 ($T_N = 1.7\text{ K}$) that grows in the C15 (MgCu_2) Laves phase. Neutron diffraction and X-ray spectra confirm that $\text{CePt}_{2.5}$ 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 interac-

tions evolve as x is varied from 0 to 1 . The present study was therefore performed on CePt_{2+x} with $x = 0\text{--}1$ through the measurements of specific heat at 0.4 to 30 K .

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 $0.4\text{--}30\text{ K}$ using a thermal-relaxation microcalorimeter in a ^3He refrigerator, with the mg-pellet sample attached to a sapphire holder on which a $\text{RuO}_2\text{--Al}_2\text{O}_3$ film thermometer and a Ni–Cr heater were deposited.

3. Results and analysis

Diffraction measurements indicated that the lattice constant a decreases from 7.73 \AA to 7.64 \AA as x increases from 0 to 1 (Fig. 1). The decrease of lattice constant is

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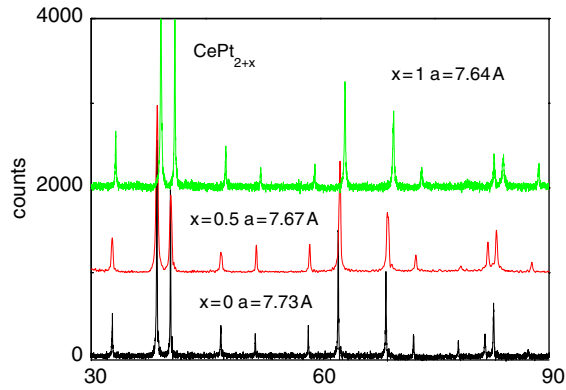


Fig. 1. X-ray 2θ (degree) diffraction patterns for CePt_{2+x} with $x = 0, 0.5$ and 1.

likely due to the smaller ionic size of Pt compared to Ce. The increased line width of $\text{CePt}_{2.5}$ 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 $T = 0.4\text{--}15\text{ K}$ 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 $\text{CePt}_{2.5}$ 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 $\sim 200\text{--}300\text{ K}$, for $T < 15\text{ K}$ 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 $R \ln 2$ 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 ($T_{\text{K}} = 4.6\text{ K}$ for 0.56% Ce) with the assumption of negligible contribution of magnetic correlations for $T > 8\text{ K}$.

Applying this same data analysis to $\text{CePt}_{2.5}$ and CePt_3 the entropy of $\text{CePt}_{2.5}$ and CePt_3 are estimated to be 0.95 and 0.93 $R \ln 2$ respectively, possibly indicating the appearance of some nonmagnetic Ce^{4+} or that some ceriums

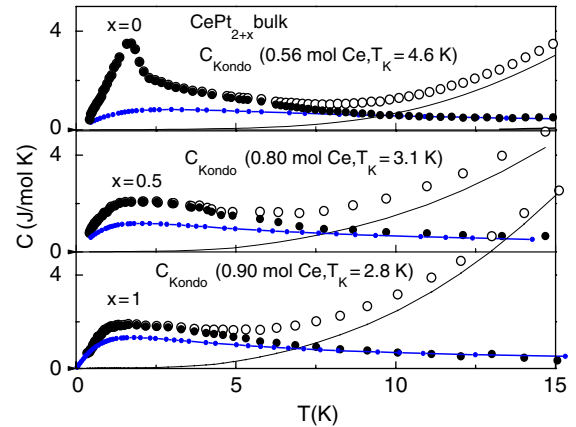


Fig. 2. Specific heat for CePt_{2+x} with $x = 0, 0.5$ and 1 (open circles), other notations are lattice phonon C_{ph} (solid lines), $C(T)-C_{\text{ph}}$ (solid circles), and Kondo model fit (dashed lines)

participate in higher-temperature interactions. The contributions of Kondo interactions C_{K} were obtained for $\text{CePt}_{2.5}$ with $T_{\text{K}} = 3.1\text{ K}$ for 80% Ce and for CePt_3 with $T_{\text{K}} = 2.8\text{ K}$ 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 $\text{CePt}_{2.5}$ 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

This work was supported by the National Research Council of the Republic of China under Grant no. NSC 93-2112-M-001-022 and by the US Department of Energy under Contract no. DE-AC02-05CHI231.

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