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Authors
Chen, Y.Y.
Huang, P.H.
Booth, C.H.
et al.

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

Y. Y. Chen, a,* P. H. Huang, a C. H. Booth, b J. M. Lawrence c

aInstitute of Physics, Academia Sinica, Taipei, Taiwan 115 ROC
bChemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley CA 94720-8175 USA
cDepartment of Physics and Astronomy, University of California, Irvine CA 92717 USA

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Abstract

CePt$_2$ is an antiferromagnetic (AF) Kondo-lattice compound with $T_N \approx 1.7$ K and $T_K \approx 4.6$ 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 ($R \ln 2$) 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 (80-90%) 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.

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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$_5$Al is an antiferromagnet with $T_N=2.5$ K, but with 50% La substitution for Ce, it becomes a heavy fermion with $\gamma=1000$ 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$_x$ 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.

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$_x$ 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$_2$O$_3$ film thermometer...
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 contributions of the lattice phonon $C_{ph}$, magnetic correlations $C_{mag}$, Kondo interactions $C_K$ and crystal field splittings $C_{cf}$. 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 contributions $S = \int (C/T) \, dT$ is found to be about Rhn2 between 0 to 15 K, the result is consistent with a $\Gamma_7$ 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 Rhn2 respectively, possibly indicating the appearance of some nonmagnetic Ce$^{3+}$ 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$_1$ 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.

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