Competition between Kondo effect and RKKY interaction modified by Cu concentration in $Ce(Pd_{1-x}Cu_x)_2Al_3$

Peijie Sun [a,1], Yosikazu Isikawa [a], Qingfeng Lu [b], Tomohiko Kuwai [a], Dexuan Huo [a], Junji Sakurai [a]

[a] *Department of Physics, Toyama University, Toyama 930-8555, Japan*
[b] *Department of Physics, Henan Normal University, Henan 453002, China*

Abstract

It was found that introducing Cu into Pd-site of $CePd_2Al_3$ drives down the antiferromagnetic temperature and brings about a ferromagnetic-like state, then further introduction leads to a revival of the antiferromagnetic state again. The nature of the conduction electron band changed by Cu substitution plays an essential role in such $Ce(Pd_{1-x}Cu_x)_2Al_3$ system. Focusing on the Cu-poor regime of $x \leq 0.1$, we found from specific heat results that T_N decreases rapidly with growing Cu concentration, while Sommerfeld coefficient γ shows an increase tendency. These behaviors do not follow the Doniach's phase diagram and are contrasted with the previous studies on $Ce(Pd_{1-x}Ni_x)_2Al_3$ system.

Key words: Kondo effect; RKKY interaction; $Ce(Pd_{1-x}Cu_x)_2Al_3$; specific heat

As a typical antiferromagnetic Kondo compound, $CePd_2Al_3$, which exhibits antiferromagnetic ordering at $T_N = 2.8$ K and Sommerfeld coefficient $\gamma = 380$ mJ/mol K^2 [1], has been investigated by a variety of experimental techniques so far. The magnetic moments in $CePd_2Al_3$ have a value about 0.38 μ_B [2], and the entropy released up to T_N is no more than 40 % of R ln 2 from the doublet ground state [1], indicating a heavy competition between magnetic ordering and Kondo effect. As recently reported, T_N is decreased and the Kondo temperature T_K is enhanced by introducing Ni to Pd-site [3–5], and the Doniach's phase diagram [6]is reproduced in this case. Similar behaviors were also observed from the experimental results under pressure [7]. We know now, decreasing of unit-cell volume provides a rather good explanation to the evolution of both T_N and T_K in the above two cases, in which, hybridization between conduction electrons and localized 4f electrons is strengthened.

[1] Corresponding author. E-mail: d023014@ems.toyama-u.ac.jp

We investigated a similar system $Ce(Pd_{1-x}Cu_x)_2Al_3$ in this work. Introducing Cu provides an extra d electron to the conduction electron band in comparison to Pd/Ni. Continuous substitution of Pd by Cu leads to a complicated magnetic phase diagram as shown in Fig. 1. Primarily, T_N decreases abruptly with substitution in the Cu-poor regime of $x < 0.1$, passing a concentration region without long range magnetic order, a ferromagnetic-like state is developed around $x = 0.1 \sim 0.4$. The antiferromagnetic state is established again at $x = 0.5$, then T_N goes up with x. A more complete description of the magnetic and transport properties for this system will be published elsewhere [8]. However, here we will pay our attentions on the specific heat measurements of $x \leq 0.1$.

Polycrystalline samples employed were prepared by arc-melting under an argon atmosphere and were subsequently annealed at 900 ˚C for 100 $\sim$ 150 hours. The crystal structure was confirmed to be a hexagonal $PrNi_2Al_3$-type one by analyzing the powder x-ray diffraction patterns. The specific measurements were

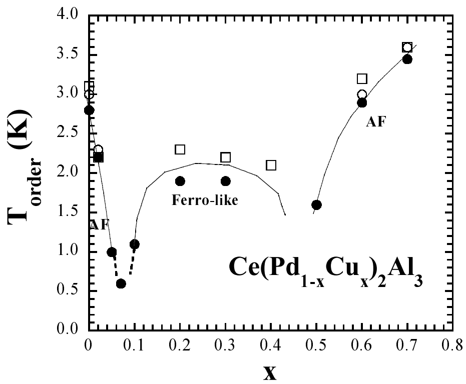

Fig. 1. Magnetic phase diagram of $Ce(Pd_{1-x}Cu_x)_2Al_3$ system. Closed circles, open circles, and squares denote magnetic transition temperatures defined by peaks in specific heat, magnetic susceptibility and electrical resistivity, respectively [8].

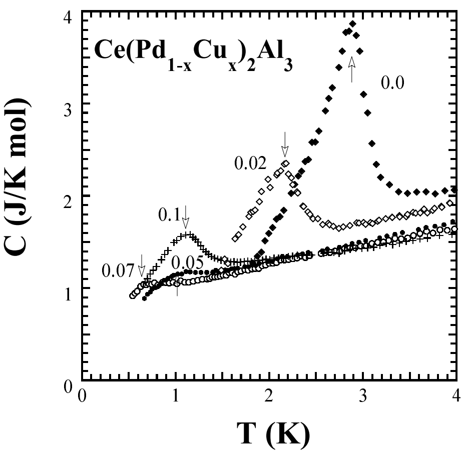

Fig. 2. Specific heat as a function of temperature. The arrows indicate the phase transitions.

performed in an adiabatic method using a ^{4}He cryostat or a ^{3}He -^{4}He dilution refrigerator.

Fig. 2 shows the specific heat of $Ce(Pd_{1-x}Cu_x)_2Al_3$ as a function of temperature, for $x = 0.0$, 0.02, 0.05, 0.07 and 0.1. Introducing of slight amount of Cu into Pd-site lowers the antiferromagnetic ordering temperature rapidly. A well defined peak regarded as T_N was observed at 2.2 K for $x = 0.02$, however, the peak for $x = 0.05$ and 0.07 drops down to 1.0 K and 0.6 K respectively, and is somewhat broad and tiny. Nevertheless, for $x = 0.1$, a well defined peak with considerably large intensity was observed. This indicates a revival of magnetic transition as a ferromagnetic-like one, if the results of magnetization measurement are considered [8]. It should be noted that, for the concentration in the crossover of magnetic types as $x = 0.05$ and 0.07, a complex magnetic state, e.g. spin glass, rather than a well arrayed magnetic ordering maybe realize and further investigation is required.

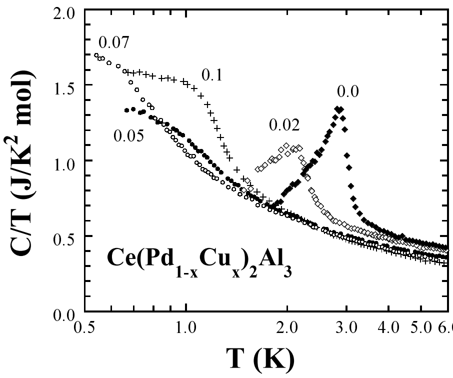

Fig. 3. Specific heat divided by temperature as a function of temperature on a log T scale.

C/T curves presented in Fig. 3 show that C/T value starts increasing with decreasing temperature from at least 6 K for all concentrations. Peak corresponding to T_N was observed for $x = 0.0$ and 0.02, while sharp upturn over a value of 1 J/mol K^2 was observed for $x =$ 0.05, 0.07 and 0.1. Moreover, we notice that C/T continues to increase even below the temperature of phase transition for x= 0.05, 0.07, and 0.1. This is attributed to the competition between Kondo effect and RKKY interaction, since the former promotes and the latter suppresses the spin fluctuations, which enhance the γ value. In fact, a decrease tendency of T_K with introducing Cu was inferred from some facts, e.g. the entropy released up to T_N for $x = 0.6$ and 0.7 reaches to about 80 % of R ln 2 [8]. Obviously, such a tendency is also supported by the specific heat of Cu-poor regime, and is contrary to the case of $Ce(Pd_{1-x}Ni_x)_2Al_3$, although a decrease of unit-cell volume was observed in both systems with introducing Ni/Cu and a similar volume effect is expected for the present case.

In summary, the specific heat data of $x \le 0.1$ for $Ce(Pd_{1-x}Cu_x)_2Al_3$ shows that, introducing Cu into Pd-site of $CePd_2Al_3$ modifies Kondo effect and RKKY interaction sensitively, and the nature of conduction band changed by the extra d electron overwhelms the volume effect and then dominates in the physical properties. The results do not follow the Doniach's diagram.

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