Specific heat study of non-Fermi liquid behavior in $Ce_xLa_{1-x}Pd_2Al_3$

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Abstract

We have measured specific heat of $Ce_xLa_{1-x}Pd_2Al_3$ ($0 \le x \le 1.0$) from 0.4 to 40 K. The magnetic specific heat normalized to a mole of Ce divided by temperature, C_m/T, for $x = 1.0$ shows a peak at 3.0 K which is attributed to the antiferromagnetic transition. For $x = 0.7$, a logarithmic diverdence in C_m/T is observed below 7 K simultaneously with the disappearance of magnetic transition. This suggests that the non-Fermi liquid behavior appears due to the magnetic instability.

Key words: $Ce_xLa_{1-x}Pd_2Al_3$, Specific heat, Non-Fermi liquid behavior, Magnetic instability

1. Introduction

The hexagonal $PrNi_2Al_3$-type compound $CePd_2Al_3$ is characterized as an antiferromagnetic (AF) heavy fermion material with large electronic specific heat coefficient $\gamma(0) \simeq 340$ mJ/molK^2 [1],[2]. The magnetic structure was determined for polycrystalline samples; the Ce moments align in the basal plane and are stacked antiferromagnetically along the *c*-axis [3]. The interesting point of this compound is strong sample dependence of the magnetic ordering. AF ordering below $T_N = 2.7$ K was found in annealed polycrystals, whereas as-cast polycrystals and single crystals do not exhibit long-range magnetic order [2]. This sample dependence is believed to be responsible for displacements and/or vacancies of Al sites destroying the weak interplane magnetic interactions [4],[5].

Previously we reported the low temperature properties of $Ce_xLa_{1-x}Pd_2Al_3$ with $x \le 0.15$ [6]. Non-Fermi liquid (NFL) behavior is observed in specific heat, electric resistivity and magnetic susceptibility below 7 K. The origin of the anomalous behavior has not been clarified, however the characteristic situation of the magnetic order in $CePd_2Al_3$ is likely to be significant. In the present work, we extend our study on the NFL behavior to wide range of x between 0.3 and 1.0 including the critical concentration, at which T_N becomes to be 0 K.

The polycrystalline samples of $Ce_xLa_{1-x}Pd_2Al_3$ ($0 \le x \le 1.0$) were prepared by arc-melting stoichiometric amounts of the four elements in an argon atmosphere, and careful annealing at ahout 900 °C follows for 5 days in vacuum. The X-ray diffraction experiment at room temperature confirms that the samples are crystallized into a single phase. The specific heat measurements were carried out between 0.4 and 40 K by using an adiabatic calorimeter, which was mounted in a ^{3}He and ^{4}He cryostats with a mechanical heat switch.

The magnetic contribution of the 4f electrons to the specific heat C_m was evaluated by subtracting the specific heat of $LaPd_2Al_3$ from that of $Ce_xLa_{1-x}Pd_2Al_3$ ($0.3 \le x \le 1.0$). In fig. 1 we show the temperature dependence of C_m for $x \ge 0.3$. The data for $x = 1.0$ in this work exhibit almost the same behavior as those of ref. [2], except sharper peak observed at higher T_N of 3.0 K. The result seems to indicate good quality of our samples. With decreasing x, T_N is lowered to 1.8 and 0.8 K for $x = 0.9$ and 0.8, respectively and consequently disappears in $x = 0.7$.

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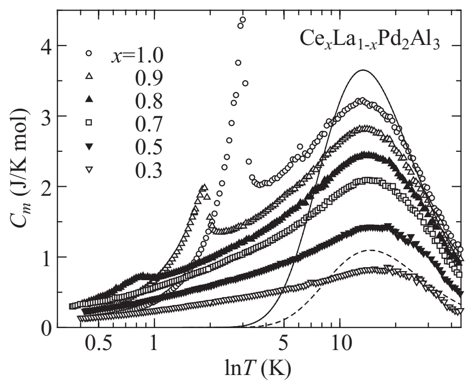

Fig. 1. Magnetic specific heat C_m of $Ce_xLa_{1-x}Pd_2Al_3$ in C_m vs. ln$T$ plot. The solid and dashed curves are calcurated Schottky specific heat with Δ_1=32K and 35K, respectively.

A broad maximum around 13-15 K observed for all samples is attributed to the Schottky-type anomaly due to the crystalline electric field (CEF) splitting of 4f states in Ce^{3+} ions. From the maximum value, it is considered that the first excited doublet mainly contributes to the anomaly. To explain the maximum, we calcurated the Shottky specific heat by assuming a small doublet-doublet splitting with $\Delta_1 = 32$ and 35 K (the solid and dashed lines in fig. 1, respectively). The lines agree with the maximum for $x = 1.0$ and 0.3. Thus the strength of CEF is insensitive to x.

In fig. 2 we normalize the magnetic specific heat to a mole of Ce and plot in C_m/T vs. ln$T$. For $x = 1.0$, the extrapolation of C_m/T leads to $\gamma(0) = 260$ mJ/molK^2 which is smaller than that of ref. [2]. Since the sample having sharper peak at T_N exhibits lower $\gamma(0)$, the enhanced value of $\gamma(0)$ is thought to arise from not only the strong Kondo interaction but also the randomized magnetism. The $\gamma(0)$ monotonously increases with decreasing x. Then C_m/T shows a logarithmic divergence for $x = 0.7$ simultaneously with the disappearance of the AF transition. This suggests that the NFL behavior appears due to the magnetic instability. With further decreasing x, the data of C_m/T exhibit surprisingly identical temperature dependence in the wide Ce concentration range between $x = 0.3$ and 0.7. Such concentration dependence characteristic of $Ce_xLa_{1-x}Pd_2Al_3$ has not been found in other systems so far.

To the NFL behavior appearing around the magnetic instability, the SCR theory [7] has been applied frequently [8]. However the invariant slope of the divergence in C_m/T for $0.3 \leq x \leq 0.7$ cannot be explained by the theory. To elucidate the origin of the NFL behavior, it aeems to take into account the magnetic order sensitive to the randomness of Al sites in $CePd_2Al_3$. Systematic measurements of the NQR spectrum on our samples are required to clarify the disorder of the Al sites as in ref. [4] and [5].

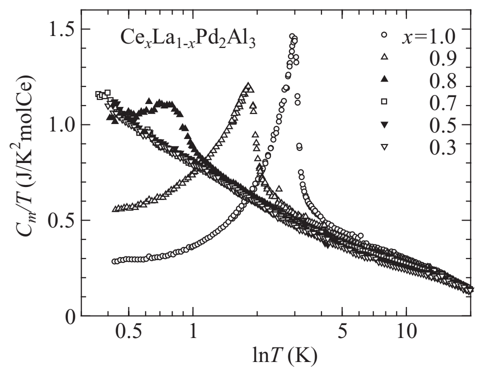

Fig. 2. Magnetic specific heat C_m of $Ce_xLa_{1-x}Pd_2Al_3$ in C_m/T vs. ln$T$ plot.

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