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Low Temperature Properties of the $Ce_{1-x}La_xNiAl_4$

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Measurements of the heat capacity in ultralow temperatures (down to 350 mK) have been carried out for $Ce_{1-x}La_xNiAl_4$. The paramagnetic behavior above about 30 K can be well described by the Curie–Weiss magnetic susceptibility. The undoped $CeNiAl_4$ compound is a known heavy fermion system with a large electronic specific heat coefficient ($\gamma = 0.5$ J mol^{-1} K^{-2}) and the Kondo temperature in the range 30–80 K. In the case of the $Ce_{0.8}La_{0.2}NiAl_4$ and $Ce_{0.6}La_{0.4}NiAl_4$ compounds a peak in C/T appears below 2 K, which is strongly damped by the magnetic field. It is probably connected with the Kondo and/or magnetic interactions and the electronic specific heat coefficient is 0.19 J mol^{-1} K^{-2} (0.43 J mol^{-1} K^{-2}) for $x = 0.2$ ($x = 0.4$) at $T \to 0$. The value determined above the peak, at temperature for which the magnetic field starts to decrease γ (≈ 3 K), is about 0.5 J mol^{-1} K^{-2} and the effect of the magnetic field can be well analyzed in frames of the single-ion Kondo model.

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1. Introduction

$CeNiAl_4$ is a well known heavy fermion system [1–4] with the electronic specific heat coefficient reaching value of $\gamma = 0.5$ J mol^{-1} K^{-2}, as determined by the extrapolation of the specific heat C_p/T down to $T = 0$ in the region of its steep increase in the lowest temperatures. The Kondo temperature has been found in the range 30–40 K, depending on the method of measurements. Especially, in the previous studies we have measured the electrical resistivity and thermopower, which confirmed the Kondo lattice state in $CeNiAl_4$.

As the properties of the Ce-based Kondo systems are sensitive to the pressure (external or internal), it is often practiced to dilute Ce with a nonmagnetic element. In the case of $CeNiAl_4$ the dilution with yttrium has been studied by Ragel et al. [5]. In this paper, we discuss the low temperature properties of the $Ce_{0.8}La_{0.2}NiAl_4$ and $Ce_{0.6}La_{0.4}NiAl_4$ compounds, with a special emphasis on the effect of the magnetic field on the behavior of the heat capacity. The previous studies of the magnetic properties of these compounds in the standard temperature range (2–300 K) have shown logarithmic increase of the electrical resistivity at low temperatures and the Curie–Weiss type behavior of the magnetic susceptibility [6]. A transition from the Kondo dense regime ($x = 0$) to the single-ion Kondo regime ($0.05 \le x \le 0.8$) has been observed [6] for $Ce_{1-x}La_xNiAl_4$. The heat capacity measurements did not include the low temperature range (below 2 K) and the influence of the magnetic field was not analyzed. In the present study the single-ion Kondo model is tested to explain the evolution of the electronic specific heat coefficient with the magnetic field value.

2. Experimental

The $Ce_{1-x}La_xNiAl_4$ compounds were prepared by induction melting of the stoichiometric amounts of the constituent elements in a water-cooled boat, under an argon atmosphere. Magnetic measurements were carried out using the VSM option of the PPMS system (Quantum Design) in the magnetic field up to 9 T.

Heat capacity measurements were carried out by PPMS commercial device (Quantum Design) in Prague, in the temperature range 350 mK–300 K.

3. Results and discussion

The reciprocal magnetic susceptibility (Fig. 1) shows linear dependence on temperature in wide temperature range. The Curie–Weiss model leads to close to localized values of the magnetic moments (2.54 μ_B for Ce) and the electrical resistivity shows logarithmic growth [6] at low temperatures (inset of Fig. 1), which is typical of the Kondo systems.

An example of the magnetic field effect on the temperature dependence of the specific heat is presented in Fig. 2. A maximum developed by antiferromagnetic correlations and/or the Kondo interactions is visible at low temperatures and is gradually damped for measurements in magnetic field. Therefore, we determine the electronic

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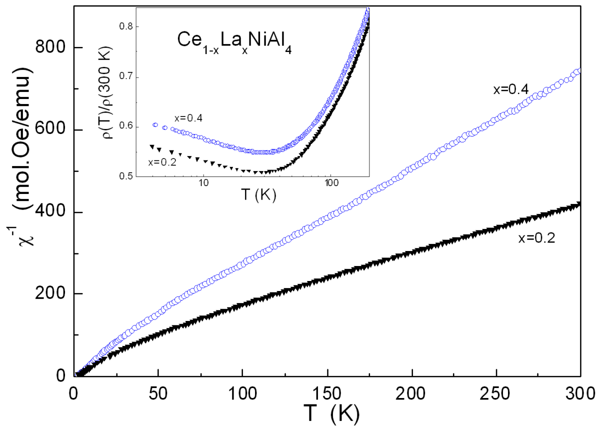

Fig. 1. The reciprocal magnetic susceptibility and the temperature dependence of the electrical resistivity (inset) for $Ce_{0.8}La_{0.2}NiAl_4$ and $Ce_{0.6}La_{0.4}NiAl_4$.

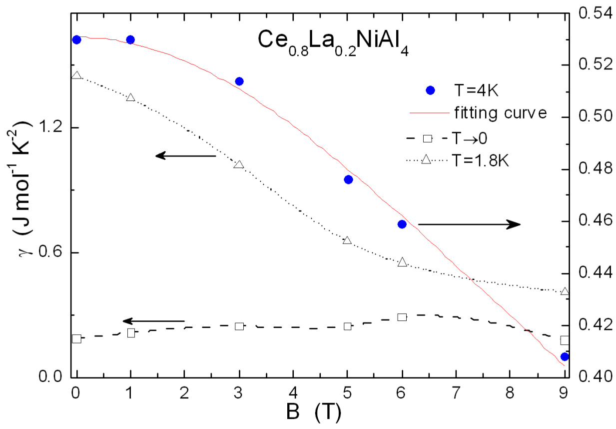

Fig. 3. Field dependence of the electronic specific heat coefficient for $Ce_{0.8}La_{0.2}NiAl_4$. Right axis: experimental points and fit (solid line) with Eq. (1) for γ_2. Left axis: γ_0 and γ_1 — the lines are a guide for the eye.

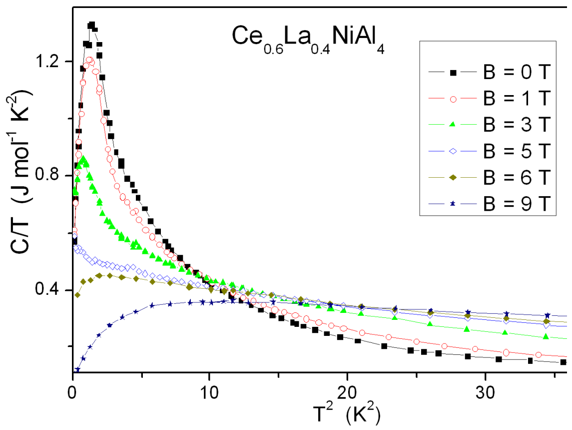

Fig. 2. The plot of C/T vs. T^2 for $Ce_{0.6}La_{0.4}NiAl_4$ in various values of the applied magnetic field.

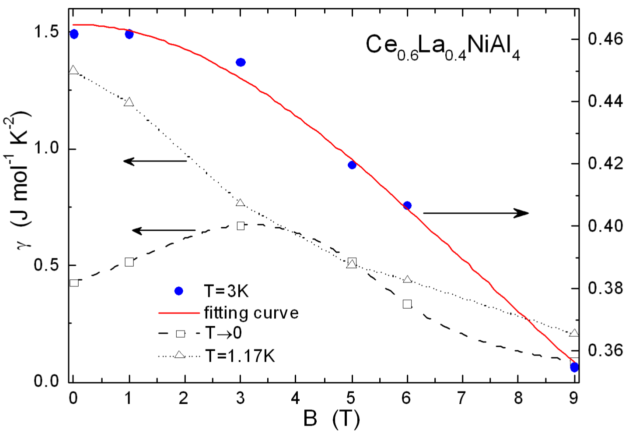

Fig. 4. Field dependence of the electronic specific heat coefficient for $Ce_{0.6}La_{0.4}NiAl_4$. Right axis: experimental points and fit (solid line) with Eq. (1) for γ_2. Left axis: γ_0 and γ_1 — the lines are a guide for the eye.

specific heat coefficient γ in three characteristic points: at $T \to 0$ below the peak (linear extrapolation of C/T vs. T^2), at the temperature of the peak for $B = 0$ T, and at temperature above the peak, for which the magnetic field starts to decrease γ (3 K for $x = 0.4$ and 4 K for $x = 0.2$). We label the values by γ_0, γ_1 and γ_2, respectively. In zero magnetic field γ_0 is 0.19 J mol^{-1} K^{-2} (0.43 J mol^{-1} K^{-2}) for $x = 0.2$ ($x = 0.4$), whereas γ_2 takes the value 0.53 J mol^{-1} K^{-2} (0.46 J mol^{-1} K^{-2}). Hence, γ_2 is in good agreement with the value determined previously for $x = 0$ [3, 4].

At low temperatures and after the application of the highest accessible magnetic field ($B = 9$ T) γ_0 drops down to 0.18 J mol^{-1} K^{-2} (0.09 J mol^{-1} K^{-2}) for $x = 0.2$ ($x = 0.4$).

Figure 3 and 4 show the electronic specific heat coefficients (γ_0, γ_1 and γ_2) as a function of the magnetic field for $Ce_{0.8}La_{0.2}NiAl_4$ and $Ce_{0.6}La_{0.4}NiAl_4$. As for $x \geq 0.05$ the incoherent Kondo regime is extended [6], it is justified to employ the single-ion Kondo model to describe the effect of the magnetic field on the γ values. To exclude the contribution due to the magnetic interactions we should operate above the peak of C/T. Indeed, as is visible in Figs. 3 and 4, only $\gamma_2(B)$ can be well analyzed by the relation [7–9]:

$$\gamma = \frac{\pi k_B R}{3\sqrt{\Gamma_0^2 + (\mu B)^2}}, \qquad (1)$$

where Γ_0 is the width of the Kondo resonance and is equal to the Kondo temperature T_K, μ is the effective magnetic moment and the other parameters have the usual meaning. From the fit with Eq. (1) we get $T_K = 16$ K and $\mu = 2.3\ \mu_B$ and $T_K = 19$ K and $\mu = 2.6\ \mu_B$ for $Ce_{0.8}La_{0.2}NiAl_4$ and $Ce_{0.6}La_{0.4}NiAl_4$, respectively.

As results from Figs. 3 and 4, $\gamma_0(B)$ exhibits a maximum, which may be developed by the presence of the magnetic correlations in the samples studied.

4. Conclusions

We have shown that $Ce_{0.8}La_{0.2}NiAl_4$ and $Ce_{0.6}La_{0.4}NiAl_4$ compounds exhibit large values of the electronic specific heat coefficient in spite of the significant dilution of the parent $CeNiAl_4$ compound with the nonmagnetic La and this enhancement of γ is observed in various analyzed temperature ranges. The analysis of the magnetic field effect on γ in frames of the single-ion Kondo model provides T_K of about 16–19 K for both compounds, which is much lower than the value estimated previously for $CeNiAl_4$ ($\approx$ 30 K). Close to the theoretically expected values of the effective magnetic moments are obtained within the model.

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