Heat Capacity of Heavy Fermion Compound CeCu$_4$Ga in High Magnetic Fields

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The heat capacity in the applied magnetic field up to 9 T, susceptibility and magnetization of polycrystalline CeCu$_4$Ga are presented. Magnetic ordering was not observed down to 2 K. For temperature $T < 200$ K a Curie–Weiss behavior is observable giving an effective magnetic moment $\mu_{\text{eff}} = 2.53 \, \mu_B$/f.u. The experimental value of $\mu_{\text{eff}}$ is close to the calculated one for a free Ce$^{3+}$ ion ($\mu_{\text{eff}} = 2.54 \, \mu_B$/f.u.), thus indicating the presence of well localized magnetic moments carried by the stable Ce$^{3+}$ ions. At low temperatures the electronic heat capacity coefficient value depends strongly on the temperature range used for the extrapolation and applied magnetic field. We observe a typical heavy fermion behavior with $\gamma$ value of about 380 mJ mol$^{-1}$ K$^{-2}$ obtained from extrapolation to $T = 0$ K of the temperature range above 7 K. However, extrapolation of the lowest temperatures range yields the $\gamma$ value of 3.3 J mol$^{-1}$ K$^{-2}$.

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1. Introduction

Cerium based ternary compounds demonstrate different phenomena depending on the valence of the Ce ion. It is believed that the hybridization between the conducting electrons and the 4$f$ Ce electrons should be responsible for the valence state of Ce. Depending on the strength of the $f$-ligand hybridization one observes in these compounds phenomena such as magnetic ordering, heavy fermion behavior, the Kondo effect, valence fluctuations and superconductivity.

We have previously studied the CeNi$_4$X (X = Al, Ga, Si) [1–3], and CeCu$_4$Al [4] compounds crystallizing in the hexagonal CaCu$_5$-type structure, space group $P$6/mmm. The CeNi$_4$X compounds are of special interest due to the nearly filled Ni (3$d$) band implying a negligible contribution of Ni atoms to the resultant magnetic moment [1–3]. In the temperature dependence of electrical resistivity we have observed a shallow minimum for CeNi$_4$X below 20 K. It has been ascribed to a Kondo-like behavior. Both the susceptibility and X-ray photoelectron spectra (XPS) show that Ce ions in CeNi$_4$X are in the intermediate valence state. The CeCu$_4$Al is known as the heavy fermion compound [4] and it is the derivate of CaCu$_5$. The estimated value of the electronic coefficient $\gamma$ was of about 280 mJ mol$^{-1}$ K$^{-2}$.

In this paper we describe our studies of the magnetic (ac/dc magnetic susceptibility) and thermodynamic (heat capacity) properties of the CeCu$_4$Ga compound.

2. Experimental details

The CeCu$_4$Ga compound was prepared by induction melting of stoichiometric amounts of the constituent elements in a water-cooled boat, under an argon atmosphere. The ingots were inverted and melted several times to ensure homogeneity. The crystal structure was established by a powder X-ray diffraction technique, using Cu $K_\alpha$ radiation. The CeCu$_4$Ga compound crystallizes in the hexagonal CaCu$_5$-type structure, space group $P$6/mmm.

Heat capacity measurements were performed by PPMS commercial device (Quantum Design) in the temperature range 2–300 K by relaxation method using two-tau model.

Magnetic measurements were carried out using a vibrating sample magnetometer in a magnetic field up to 9 T using the System MagLab 2000 magnetometer.

3. Results and discussion

In Fig. 1 the temperature dependence of the inverse magnetic susceptibility $\chi^{-1}(T)$ is presented. Magnetic ordering was not observed down to 2 K. Above about 2 K, $\chi^{-1}(T)$ follows the Curie–Weiss law with the effective magnetic moment $\mu_{\text{eff}} = 2.53 \, \mu_B$/f.u. and the paramagnetic Curie temperature $\theta_p = -4.8$ K. The experimental value of $\mu_{\text{eff}}$ is close to the calculated one for a free Ce$^{3+}$ ion $\mu_{\text{eff}} = g(j(j+1))^{-1/2} = 2.54 \, \mu_B$/f.u., thus indicating a presence of well localized magnetic moments carried by
The stable Ce$^{3+}$ ions. The negative paramagnetic Curie temperature is known to enable a rough estimation of the Kondo temperature $T_K$ as $T_K = \theta_p/2 = 2.4$ K.

Fig. 1. The temperature dependence of the inverse magnetic susceptibility for CeCu$_4$Ga. Solid line: a fit with the Curie-Weiss law.

The magnetic field dependences of magnetization for CeCu$_4$Ga are presented in Fig. 2. The temperature evolution confirms the previous observations and the absence of any ordering down to 2 K.

Fig. 2. Magnetic field dependence of the magnetic moment in different temperatures.

Figure 3 shows the temperature dependence of the heat capacity $C_p(T)$ of CeCu$_4$Ga in the temperature range 2–300 K and in zero magnetic field. We have not observed any real sign of the magnetic order down to 2 K.

Fig. 3. The temperature dependence of the heat capacity $C_p(T)$ of CeCu$_4$Ga in the temperature range 2–300 K and in zero magnetic field. Inset — the low temperature part of $C_p(T)/T$ as a function of $T^2$.

low temperatures ($T < 5$ K) the electronic heat capacity coefficient value depends strongly on the temperature range used for the extrapolation and on the applied magnetic field.

Fig. 4. The specific heat of CeCu$_4$Ga as the $C_p/T$ vs. $T^2$ dependence in magnetic fields up to 9 T.

We tried to estimate the distance from the quantum critical point using the formula 1 from [7]:

$$\frac{C_p}{T} = \frac{\gamma_0}{2} - a \sqrt{\frac{\pi}{2}} \int_0^{\infty} \frac{d\epsilon}{\sinh^2 \epsilon} \left( \frac{r}{T} \right)^{1/2} \left( \frac{r}{T} \right)^{1/2} + (2\epsilon)^2,$$

where $a$ is a constant, $t = T/T^*$ and $r$ is a control parameter tuning the material through the $T = 0$ transition [7, 8]. $T^*$ is of the order of the Kondo temperature. We have found that without studies of various doping of the compound we cannot determine $r$ and it can be put arbitrary, therefore we use $r = 0$, which in principle makes the formula equivalent to the $T^{1/2}$ dependence for critical fluctuations close to an antiferromagnetic critical point for $T \to 0$. Simultaneously, it is found that $T^*$ has to be varied to fit the $C_p/T$ vs. $T$ dependence for
various magnetic fields. Inset of Fig. 5 shows such a fit on the example of $H = 0$ T, 3 T and 6 T. Figures 5 and 6 show the field dependence of $T^*$ and $\gamma_0$, respectively. The values of $\gamma_0$ behave in a manner consistent with the Anderson model based on the theoretical predictions of Kim et al. [9] and other calculations [10], i.e., it depends strongly on the applied magnetic field.

**Fig. 5.** $T^*$ as a function of magnetic field. Inset — an example of the $C_p/T$ vs. $T$ fit.

**Fig. 6.** Electronic specific heat coefficient as a function of the applied magnetic field.

The value of $T^* = 2.4$ K is in good agreement with results of paper [5] and $T_K$ derived above from the paramagnetic Curie temperature.

### 4. Conclusions

CeCu$_4$Ga is paramagnetic and follows a Curie–Weiss law with the effective magnetic moment $\mu_{\text{eff}} = 2.53 \, \mu_B/\text{f.u.}$ and the paramagnetic Curie temperature $\theta_p = -4.8$ K. We observe a typical heavy fermion behavior with $\gamma$ value of about 380 mJ mol$^{-1}$ K$^{-2}$. Extrapolation of the lowest temperatures range of $C_p/T$ yields the value of 3.3 J mol$^{-1}$ K$^{-2}$. At low temperatures ($T < 5$ K) the electronic heat capacity coefficient depends strongly on the applied magnetic field.

The Kondo temperature estimated both from the magnetic susceptibility and the specific heat measurements is about 2.4 K.

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### References