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Heat Capacity of Heavy Fermion Compound $CeCu_4Ga$ 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 $CeCu_4Ga$ 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 μ_{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 γ 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 γ 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 $4f$ 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 $CeNi_4X$ (X = Al, Ga, Si) [1–3], and $CeCu_4Al$ [4] compounds crystallizing in the hexagonal $CaCu_5$-type structure, space group $P6/mmm$. The $CeNi_4X$ compounds are of special interest due to the nearly filled Ni ($3d$) 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 $CeNi_4X$ 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 $CeNi_4X$ are in the intermediate valence state. The $CeCu_4Al$ is known as the heavy fermion compound [4] and it is the derivate of $CaCu_5$. The estimated value of the electronic coefficient γ 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 $CeCu_4Ga$ compound.

2. Experimental details

The $CeCu_4Ga$ 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_α radiation. The $CeCu_4Ga$ compound crystallizes in the hexagonal $CaCu_5$-type structure, space group $P6/mmm$.

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 μ_{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.

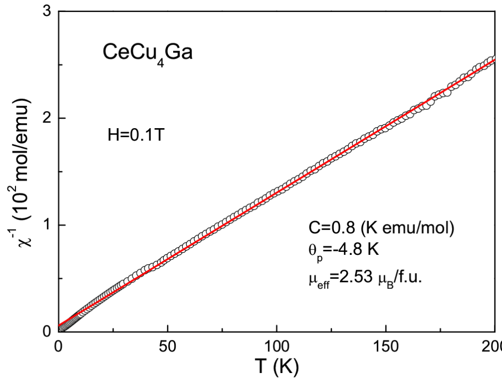

Fig. 1. The temperature dependence of the inverse magnetic susceptibility for $CeCu_4Ga$. Solid line: a fit with the Curie–Weiss law.

The magnetic field dependences of magnetization for $CeCu_4Ga$ are presented in Fig. 2. The temperature evolution confirms the previous observations and the absence of any ordering down to 2 K.

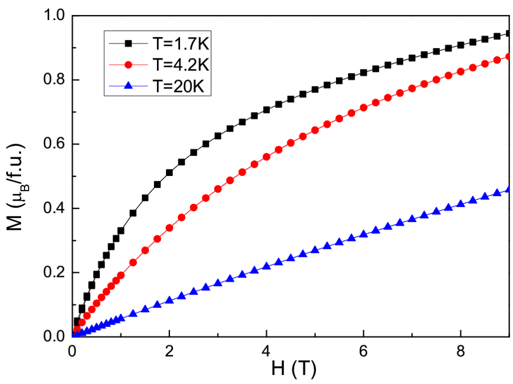

Fig. 2. Magnetic field dependence of the magnetic moment in different temperatures.

Figure 3 shows the temperature dependence of the heat capacity $C_p(T)$ of $CeCu_4Ga$ 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. In order to make comparisons with previous results [4, 5] we present in Fig. 3 (inset) the low temperature part of $C_p(T)/T$ as a function of T^2. An extrapolation to $T = 0$ K of the temperature range above 7 K yields a γ value of about 380 mJ mol^{-1} K^{-2}.

Extrapolation of the lowest temperatures range of $C_p/T(T^2)$ provides a large value of 3.3 J mol^{-1} K^{-2} as discussed below. In each case, these values confirm the presence of a heavy fermion state in this compound. This behavior is in a qualitative agreement with Refs. [5, 6].

Figure 4 presents the specific heat of $CeCu_4Ga$ as the C_p/T vs. T^2 dependence in magnetic fields up to 9 T. At

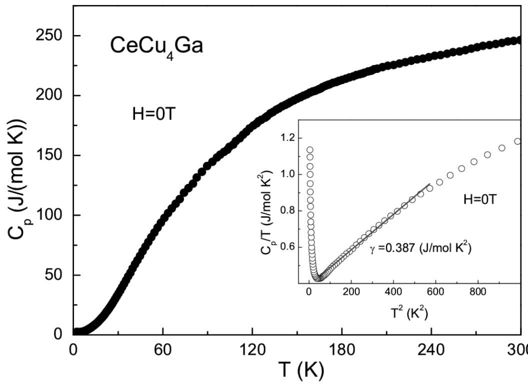

Fig. 3. The temperature dependence of the heat capacity $C_p(T)$ of $CeCu_4Ga$ in the temperature range 2–300 K and in zero magnetic field. Inset — the low temperature part of $C_p(T)/T$ as a function of T^2.

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.

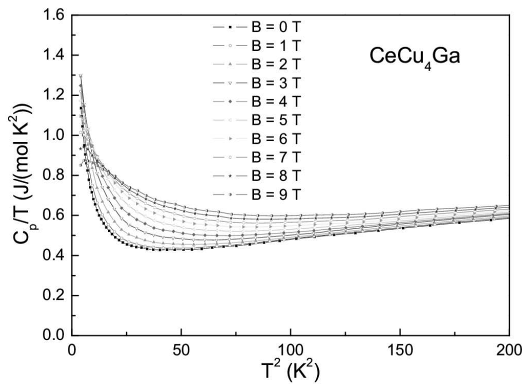

Fig. 4. The specific heat of $CeCu_4Ga$ as the C_p/T vs. T^2 dependence in magnetic fields up to 9 T.

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

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

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 aniferromagnetic 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 γ_0, respectively. The values of γ_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.

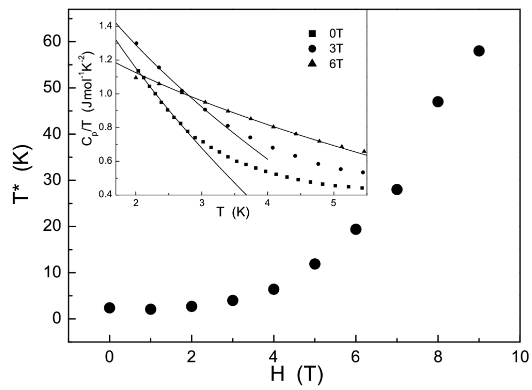

Fig. 5. T^* as a function of magnetic field. Inset — an example of the C_p/T vs. T fit.

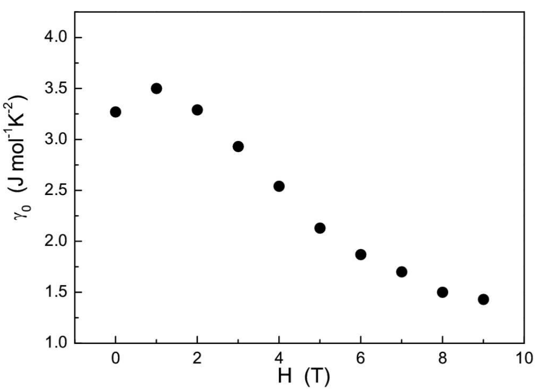

Fig. 6. Electronic specific heat coefficient as a function of the applied magnetic field.

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

$CeCu_4Ga$ is paramagnetic and follows a Curie–Weiss law with the effective magnetic moment μ_{eff} = 2.53 μ_B/f.u. and the paramagnetic Curie temperature $\theta_p = -4.8$ K. We observe a typical heavy fermion behavior with γ 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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