

Magnetization and electrical resistivity measurements on CeCuAl₃ single crystal

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Abstract.

We present the electronic properties of CeCuAl₃ studied by means of magnetization and electrical resistivity measurements in well characterized single crystal. CeCuAl₃ crystallizes in the ordered non-centrosymmetric tetragonal BaNiSn₃-type structure. The antiferromagnetic phase transition around 2.5 K was revealed by magnetization measurement. The clearly pronounced anomaly on electrical resistivity data was observed the same temperature. A relatively strong magnetocrystalline anisotropy with the magnetization easy-basal plane and hard c-axis was observed. The ferromagnetic correlation stronger along the c-axis than in the basal plane were found in paramagnetic state.

1. Introduction

CeCuAl₃ belongs to the family of CeTX₃ compounds revealing such physical phenomena as valence fluctuations in CeRuSi₃ [1], spin-glass order in CePtAl₃ [2] or the pressure induced superconductivity in CeRhSi₃ and CeIrSi₃ [3, 4]. Recently, the strong electron-phonon interaction leading to the formation of a new quantum quasi-bound state, so called vibron state, was observed in CeCuAl₃ [5]. The exceptional and often exotic behavior of these Ce-based intermetallics originates in vicinity of the energy of cerium 4f shell level to the 5d and 6s levels. The different magnetic properties then arise mainly from the competition between the long-range magnetic order of the RKKY type and the screening of the localized cerium 4f moments by conduction electrons.

CeCuAl₃ adopts the ordered non-centrosymmetric tetragonal BaNiSn₃-type structure as was unambiguously revealed by our recent study [6]. The antiferromagnetic order with sample dependent Néel temperature $T_N \cong 2.5 - 2.9$ K [7, 8, 2, 9] is realized in the compound. The preliminary results of neutron diffraction experiment led to observation of antiferromagnetic peak described by the $(\frac{1}{2} \frac{1}{2} 0)$ propagation vector [10]. We plan to perform (accepted for D10 instrument in ILL) a detailed neutron diffraction experiment to reveal the magnetic structure of CeCuAl₃. We present the results of magnetization, electrical resistivity and magnetoresistivity measurements performed on structurally well defined CeCuAl₃ single crystal.

2. Experimental details

Single crystalline CeCuAl₃ sample was prepared by Czochralski growing method and additionally annealed in quartz tubes at 900°C for 8 days. The details of sample preparation as well as its



structural and chemical characterization by differential scanning calorimetry, x-ray, electron and neutron scattering are published in our separate paper [6].

The magnetization and electrical resistivity measurements were performed on Magnetic Property Measurement System (MPMS, Quantum design) and Physical Property Measurement System (PPMS, Quantum design), respectively. The prism-shape samples with ≈ 8 mg mass were cut along the significant crystallographic directions of the tetragonal crystal structure, i.e. [100], [110] and [001], and oriented with the longest side along the direction of the magnetic field for magnetization experiments. The electrical resistivity was measured using classical four-point schema in transversal current-field mode as clarified in the corresponding figures.

3. Results and discussion

3.1. Magnetization measurements

The antiferromagnetic order in CeCuAl_3 is indicated by a maximum in the temperature dependence of magnetization measured in low magnetic fields, see Figure 1. Clear sharp maximum at $T_N \approx 2.5$ K appears when the magnetic field is applied within the basal plane (no significant differences between basal plane [100] and [110] directions were observed). The maximum is less pronounced and takes place at lower temperature of ≈ 2.4 K when the field is applied along the [001] direction. The difference between zero field-cooled (ZFC) and field-cooled (FC) regimes is rather small in measurements along all significant crystallographic directions (see Figure 1) and completely vanishes in the field of 0.05 T.

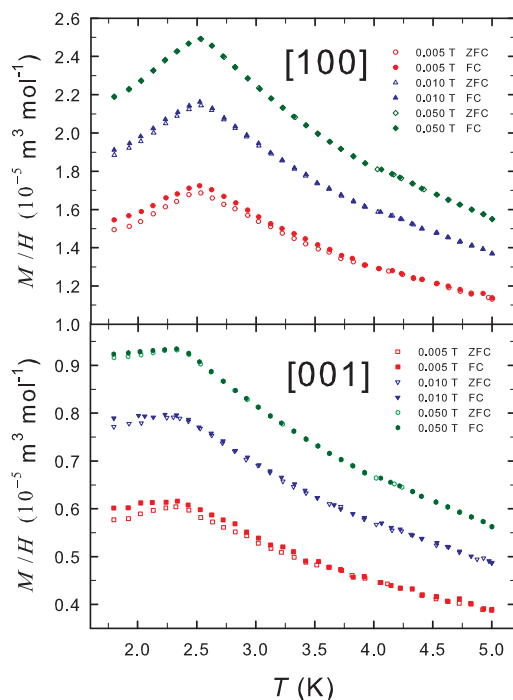


Figure 1. (Color online) The temperature dependence of magnetization along [100] and [001] in small magnetic fields. The measured curves overlap significantly, the shift of two of them (0.01 and 0.05 T) was done for better lucidity. ZFC means zero field-cooled and FC field-cooled regimes.

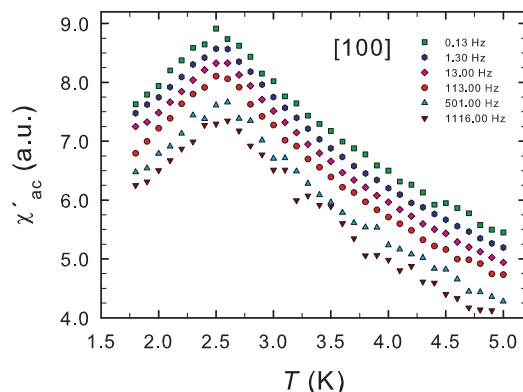


Figure 2. (Color online) The real part of ac-susceptibility measured along [100] direction. The curves for all frequencies overlap because of which each frequency curve is shifted by -0.2 from the previous one for better lucidity. The curve for frequency of 0.13 Hz is not shifted.

To bring further evidence of a long range magnetic order and to exclude some kind of spin-glass behavior, the alternate current (ac-) susceptibility measurements were performed. Figure 2 shows the temperature dependence of ac-susceptibility with a clear phase transition at T_N , in agreement with the static magnetization. The measured dependencies are frequency independent, what points to a long-range magnetic order in CeCuAl₃.

The magnetization curves measured for external field applied along significant crystallographic directions are plotted in Figure 3. We observe relatively strong anisotropy between the curves measured with field parallel and perpendicular to the tetragonal c-axis. The larger saturation magnetization for field applied perpendicular to the c-axis reflects the fact that the moment of the ground state crystal field (CF) level is larger for the z-component than for the xy-component of magnetic moments. This does not imply necessarily, that the moments order in the basal plane. The magnetization is not saturated in fields up to 7 T and further increase is expected. Assuming a smooth increase with external field, the saturated magnetization of Ce³⁺ free ion ($= 2.16 \mu_B$) would be reached in magnetic field higher than 50 T.

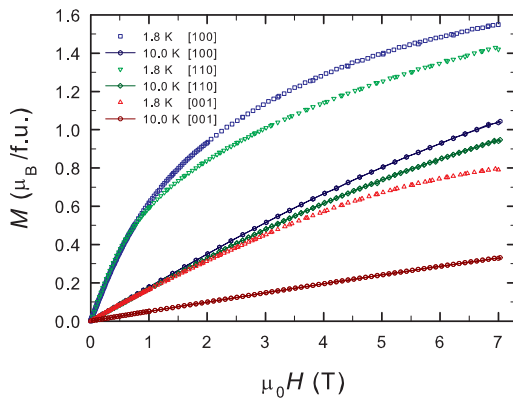


Figure 3. (Color online) The magnetic field dependence of magnetization along significant crystallographic directions [100], [110] and [001] measured at temperatures 1.8 and 10 K.

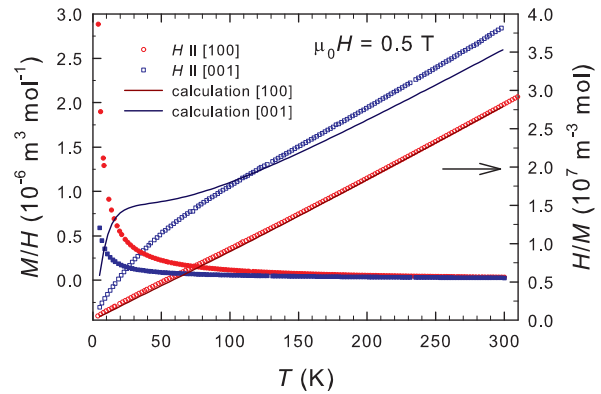


Figure 4. (Color online) The temperature dependence of M/H along [100] and [001] directions in magnetic field of 0.5 T. The H/M together with the dependencies calculated from crystal field parameters given in Ref. [5] (solid lines) are presented as well. See text for more details.

In the paramagnetic region, the temperature dependence of M/H , shown in Figure 4, follows the Curie-Weiss law at least down to 60 K for measurement with $H \parallel [001]$ and even to much lower temperatures for field perpendicular to [001]. The fitting of measured data reveals the effective magnetic moment, μ_{eff} , in good agreement with theoretical value for Ce³⁺ ion, i.e. $\mu_{\text{eff}} = 2.54 \mu_B$. The Curie paramagnetic temperature is then -52.0 K for $H \parallel [001]$, -6.2 K for $H \parallel [100]$ and -4.7 K for $H \parallel [110]$ direction. Figure 4 shows also comparison of the measured $H/M(T)$ dependencies and calculated curves based on the CF parameters determined from the inelastic neutron scattering data [5]: $B_2^0 = 0.611(17)$, $B_4^0 = -0.015(1)$ and $|B_4^4| = 0.317(4)$ meV. The calculated dependencies are plotted in Figure 4 as lines of a corresponding color. Very good agreement of calculated and measured data is observed for $H \parallel [100]$, but clear discrepancy occurs for $H \perp [001]$. Calculated curve shows a kink around 30 K, which is not reproduced in the measured data. We note that the measurements were performed several times comparing as-cast samples, annealed samples and also on two sets of independent single crystals, in order to verify the reproducibility of observed M/H dependence. All of these measurements led to almost identical results. Any small changes of CF parameters do not lead to significant improvement

of the calculated and measured curves. Presumably some common analysis of susceptibility and neutron scattering data might lead to a set of CF parameters which would satisfactory describe both experiments.

3.2. Electrical resistivity

The electrical resistivity measured with electrical current along all three main crystallographic directions is plotted in Figure 5. The relatively broad maximum around 10 K is clearly observable in resistivity along all crystallographic directions in perfect agreement with previously published data [8]. The decrease below 10 K can be caused by the development of Kondo lattice state and crystal field effect at low temperatures as speculated in Ref. [8]. The kink at low temperatures shows clearly the temperature of magnetic phase transition around 2.5 K in good agreement with magnetization and ac-susceptibility measurements (see Figure 5).

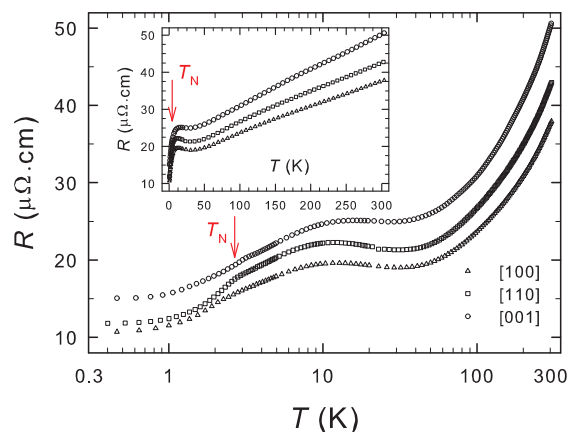


Figure 5. The temperature dependence of electrical resistivity with the current along significant crystallographic directions. The arrows corresponding to magnetic phase transitions are included.

The magnetoresistivity measurements were performed in several arrangements with respect to the electrical current and magnetic field directions. There were 4 types of mutual arrangements of crystallographic directions, electrical current flow j and magnetic field H as shown in Figures 6 and 7. In all cases, the transversal current-field configuration ($j \perp H$) was used. Temperature dependencies of resistivity in several static magnetic fields are plotted in Figure 6 and the magnetoresistance (MR) curves in Figure 7. Several differences can be traced depending on the experimental arrangement.

Large negative magnetoresistance is observed for $j \parallel [001]$, resembling measurements reported for substituted $\text{CeCu}_x\text{Ag}_{1-x}\text{Al}_3$ compounds [11]. Significant difference in resistivity development takes place for $j \perp [001]$ regardless the field direction. In these arrangements, positive MR appears for temperatures below T_N in magnetic fields up to 2-3 T (0.45 and 1.0 K curves in Figure 7). We observe a clear maximum before the resistivity starts to decrease with further field increase. It suggest suppression of the antiferromagnetic order and arise of induced ferromagnetic state, similarly as was observed for instance in YbNiAl_2 [12].

In paramagnetic state, negative magnetoresistance is observed for all arrangements, but clear difference can be traced for $H \parallel [001]$ and $H \perp [001]$. In the former case, the effect of 9 T field becomes negligible above 20 K, whereas in the latter case relatively large resistivity reduction remains up to 30 K (see Figure 6). The decrease of resistivity in rising field could be a result of probably ferromagnetic correlations between the cerium 4f moments. Other reason could be a suppression of the Kondo scattering with increasing field. Following the approach discussed also for $\text{CeCu}_x\text{Ag}_{1-x}\text{Al}_3$ compounds [11], we can use the single-ion Kondo model with Bethe-ansatz

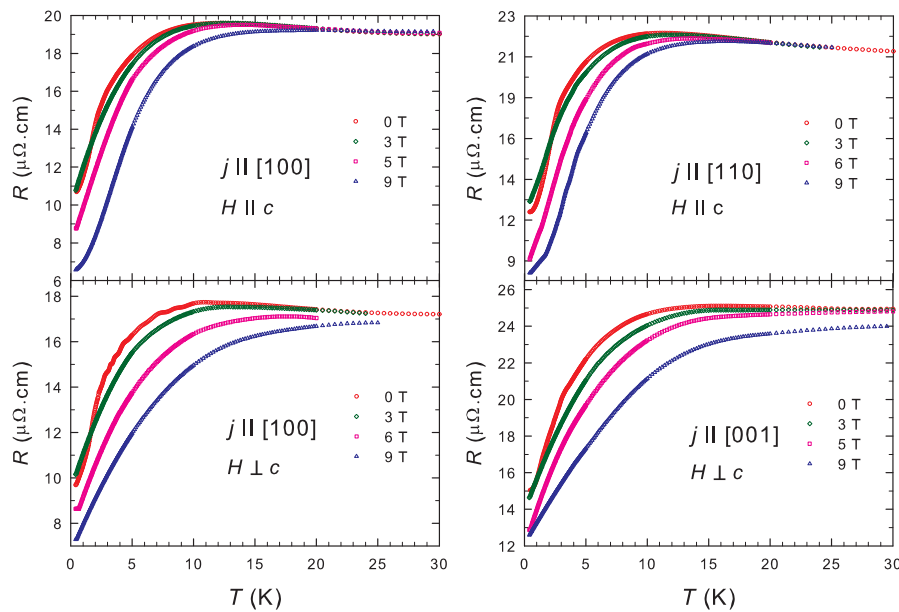


Figure 6. (Color online) The temperature dependence of electrical resistivity in magnetic field measured in several experimental arrangements with respect to current j and magnetic field H directions. The transverse configuration ($j \perp H$) of current and magnetic field was used.

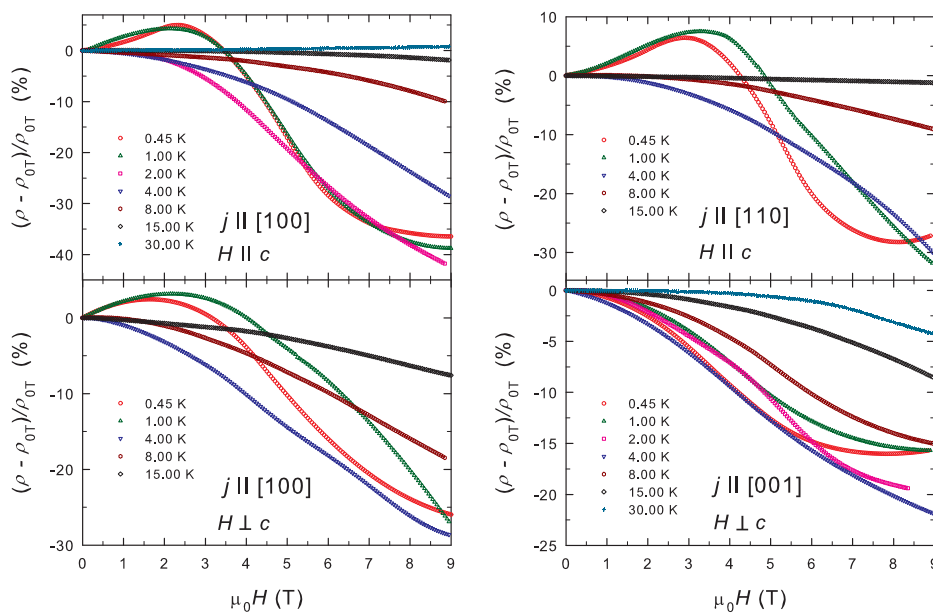


Figure 7. (Color online) The magnetoresistance ($MR = \frac{\rho(H,T) - \rho(0,T)}{\rho(0,T)}$) at several temperatures below and above antiferromagnetic phase transition with the current along significant crystallographic directions. The transverse configuration $j \perp H$ was used.

studies [13]. Figure 8 is obtained by re-scaling the x-axis as $\frac{\mu_0 H}{T + T^*}$, where T is the temperature for the MR measurement and T^* is a certain measure of the strength of ferromagnetic correlations in the material [12, 14]. The dependencies measured at temperatures above T_N overlap with each other for characteristic temperature $T^* = 1.0$ and 4.5 K for $[001] \parallel H$ and $[001] \perp H$, respectively

(the change of T^* by ± 0.5 K leads to the much worse overlap between curves measured at $T > T_N$). The former value is close to that found for polycrystalline $\text{CeCu}_{0.9}\text{Ag}_{0.1}\text{Al}_3$ [11]. Positive values of T^* imply that the ferromagnetic correlations between magnetic moments are rather weak. Lower value of T^* for magnetic field along [001] direction suggests that ferromagnetic correlations are stronger along the c-axis than in the basal plane. The development between magnetic and paramagnetic state is also demonstrated in Figure 8.

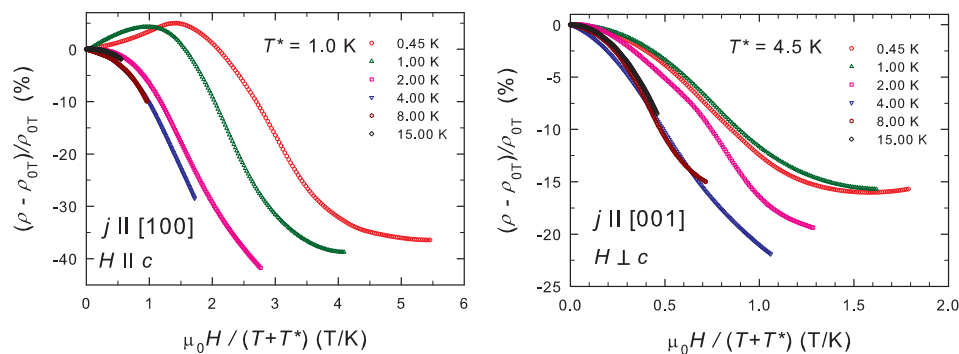


Figure 8. (Color online) The magnetoresistance ($\text{MR} = \frac{\rho(H,T) - \rho(0,T)}{\rho(0,T)}$) in dependence of $\frac{\mu_0 H}{T + T^*}$ in several temperatures with the current along crystallographic directions [100] and [001] and magnetic field parallel and perpendicular to the c-axis, respectively.

4. Conclusions

The electronic properties of CeCuAl_3 single crystal were investigated by means of magnetization, ac-susceptibility, electrical resistivity and magnetoresistivity measurements. CeCuAl_3 orders antiferromagnetically with $T_N = 2.5(1)$ K. The long-range magnetic order was verified. The magnetization measurements revealed magnetization easy-basal plane and hard c-axis. The ferromagnetic correlations stronger along the c-axis than in the basal plane were observed.

Acknowledgement

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