Magnetic and Transport Properties of BaNiSn$_3$ type CeCuAl$_3$ under Pressure

Y. Kawamura$^1$, T. Nishioka$^1$, H. Kato$^1$, M. Matsumura$^1$ and K. Kodama$^2$

$^1$Graduate School of Integrated Arts and Sciences, Kochi University, Kochi 780-8520, Japan
$^2$Kochi Core Center (KCC), Kochi University, Nankoku, 783-8502, Japan
E-mail: b08d6a02@s.kochi-u.ac.jp.

Abstract. We have measured ac susceptibility $\chi_{ac}$ and electric resistivity $\rho$ of polycrystalline CeCuAl$_3$ under pressure. At ambient pressure, $\chi_{ac}(T)$ shows a peak and $\rho(T)$ also a kink, the temperature of which increases gradually with pressure. These anomalies are attributed to a localized antiferromagnetic transition. The $\rho(T)$ shows another broad peak at around 10 K at ambient pressure, which increases monotonically with pressure. We suggest that this peak comes from the small crystalline electric field splitting combined with Kondo effect and the increase is due to enhancement of the splitting by applying pressure.

1. Introduction
Recently, BaNiSn$_3$-type heavy fermion antiferromagnet CeRhSi$_3$, CeCoGe$_3$ and CeIrSi$_3$ were reported to become superconductors under pressure [1, 2, 3]. These discoveries have received considerable attention, because BaNiSn$_3$-type crystal structure lacks the center symmetry that was considered to be necessary for heavy-fermion superconductors [4]. CeCuAl$_3$ also crystallizes in the BaNiSn$_3$-type structure, which was revealed by high-resolution neutron diffraction experiments [5]. The electric resistivity, specific heat and ac susceptibility measurements suggested that CeCuAl$_3$ is a heavy fermion antiferromagnet with the Néel temperature $T_N \approx 3$ K [6]. The NMR study on single crystals grown by the Czochralskii method under pressure revealed that $T_N$ decreases rapidly with increasing pressure [7]. This result implies quantum critical point (QCP) exists at about 10 kbar. In addition, magnetization measurements of a CeCuAl$_3$ single crystal revealed crystalline electric field level (CEF) scheme with three doublets at 0, 10, and 180 K [8].

In our previous paper, we reported dc magnetic susceptibility $\chi_{dc}$ measurement of a polycrystalline CeCuAl$_3$ under pressure up to 7.4 kbar [9]. The temperature dependence of $\chi_{dc}$ indicates a broad peak at $T_0 \approx 2.5$ K, and $T_0$ increases slightly with pressure. This pressure dependence of $T_0$ is quite different from that of $T_N$ obtained from the above NMR measurements. The purpose of this paper is to clarify what the peak in $\chi_{dc}(T)$ indicates and relationship between $T_0$ and $T_N$. In order to achieve this, we have measured the ac susceptibility $\chi_{ac}$ and the electric resistivity $\rho$ of polycrystalline CeCuAl$_3$ under pressure.
2. Experimental

Polycrystalline samples were prepared by argon-arc melting appropriate amounts of the constituent elements (3N-Ce, 4N-Cu and 4N-Al). Each sample was sealed in an evacuated quartz ampoules and annealed for 7 days at 800 °C followed by quench into water. The powder X-ray diffraction experiments confirmed that CeCuAl$_3$ is tetragonal BaNiSn$_3$-type (I4mm) structure with $a = 4.256$ Å, $c = 10.636$ Å. The ac susceptibility was measured using a standard Hartshorn Bridge circuit for powdered sample. The electrical resistivity was measured using standard ac four-probe method in a frequency of 40 Hz and in an effective current of 3 mA using a lock-in-amplifier for a sample with the shape of 1 × 1 × 5 mm$^3$. Pressure was generated by a piston cylinder type pressure cell with Daphne 7373 oil as a pressure transmitting medium and calibrated by the superconducting transition temperature of Pb and In for ac susceptibility and electrical resistivity measurements, respectively.

3. Results and Discussion

Figure 1 shows the temperature dependence of $\chi_{ac}$ of CeCuAl$_3$ under pressure. The magnitude of $\chi_{ac}$ decreases with increasing pressure. We can see from Fig. 1 (b) that there is a shoulder at $T_1 \sim 3.6$ K. $T_1$ increases with pressure. This pressure dependence is similar to that of $T_0$ in $\chi_{dc}$ [9]. It is difficult to declare by only susceptibility measurements whether $T_1$ corresponds to $T_N$.

Figure 2 shows the temperature dependence of $\rho$ of CeCuAl$_3$ at ambient pressure and 17.8 kbar together with non magnetic LaCuAl$_3$. The dominant scattering mechanism in LaCuAl$_3$ is thought to be electron-phonon scattering $\rho_{ph}$, and elastic impurity and defect scattering $\rho_{imp}$ that is identical with residual resistivity. We assume that the phonon contribution to the resistivity of CeCuAl$_3$ is the same as $\rho_{ph}$ of LaCuAl$_3$, and deduced magnetic contribution of $\rho_{mag} (= \rho - \rho_{ph})$ in CeCuAl$_3$ is also plotted in Fig. 2. The temperature dependence of $\rho_{mag}$ shows a kink at 3.5 K and a broad peak at 10 K. Since the kink is sharp and the temperature is close to $T_1$, the former anomaly is attributed to a Néel transition. The temperatures of both resistivity anomalies increase at 17.8 kbar.

Figure 3 shows a pressure-temperature phase diagram of CeCuAl$_3$ and a volume-temperature phase diagram of CeCu$_{x}$Ag$_{1-x}$Al$_3$ and CeAuAl$_3$. All the temperatures of the peak in $\chi_{dc}(T)$, kink in $\chi_{ac}T(T)$ and $\rho(T)$ are about the same and increase with pressure, which suggest

![Figure 1](image_url). (a) $\chi_{ac}$ vs $T$ and (b) $\chi_{ac}T$ vs $T$ for CeCuAl$_3$ at several pressures.
Figure 2. Temperature dependence of $\rho$ of CeCuAl$_3$ at ambient pressure and 17.8 kbar together with non magnetic reference LaCuAl$_3$. The magnetic contribution of CeCuAl$_3$ $\rho_{\text{mag}}$ is also shown. The inset shows $\rho(T)$ of CeCuAl$_3$ below 10 K. Both the kink and the broad peak temperatures indicated by arrows increase at 17.8 kbar compared to ambient pressure.

Figure 3. Pressure dependence of the temperature of the peak observed at $\chi_{\text{dc}}(T)$ ($\triangle$), the shoulder observed at $\chi_{\text{ac}}(T)$ (■), the kink observed at $\rho(T)$ (▽) and $T_N$ obtained by NMR [7] (○) and volume dependence of $T_N$ in CeCu$_x$Ag$_{1-x}$Al$_3$ ($x = 1, 0.95, 0.9, 0.8$)(□), in which the volume of the lattice increase monotonically with decreasing $x$ [10], and in CeAuAl$_3$ (◇) [11]. The inset shows the temperature of broad peak observed at $\rho(T)$ (●). The dashed line (- - - - ) is a guide for eyes.

that these anomalies represent Néel transition and $T_N$ increases with pressure. The $T_N$ of CeAuAl$_3$ or CeCu$_x$Ag$_{1-x}$Al$_3$ increases with contracting the cell volume, which is the same dependence as CeCuAl$_3$. This supports that it is appropriate that $T_N$ in CeCuAl$_3$ increases with pressure. As $T_N$ increases with pressure, CeCuAl$_3$ is in the localized electric region in Doniach’s magnetic phase diagram. On the other hand, the $T_N$ obtained by NMR study decreases rapidly with pressure. Since Al can be easily substituted by Cu in CeCuAl$_3$, the difference between the pressure dependence of $T_N$ in our study and in NMR study may be ascribed to sample dependence.
The dotted line in Fig. 2 shows the calculated spin-disorder electric resistivity attributed to CEF with three doublet \(| \pm \frac{3}{2} >, | \pm \frac{3}{2} >, | \pm \frac{5}{2} >\) at 0, 10, 180 K, respectively. Because only the magnetic moment in the ground state is effective below 10 K, the calculated line drops at about 10 K. We suggest that the broad peak in \(\rho(T)\) comes from the small CEF splitting combined with Kondo effect. The inset in Fig. 3 shows pressure dependence of the peak temperature in \(\rho(T)\). The peak temperature in \(\rho(T)\) increases monotonically with pressure, which indicates the first excited state of CEF is enhanced with pressure. Since the enhancement of the first excited states suppresses the magnetic moment at low temperature, our suggestion is consistent with the magnitude of \(\chi_{ac}\) decreasing with pressure in Fig. 1.

Recently, we have succeeded in growing single crystals of CeCu\(_{x}\)Al\(_{4-x}\) (\(x \sim 0.7\)) by the Al self flux method [12]. The CEF calculation for the susceptibility measurement of the single crystal revealed that it has three doublet \(| \pm \frac{3}{2} >, | \pm \frac{3}{2} >, | \pm \frac{5}{2} >\) in this order. The ground and first excited state are exchanged to each other from those of CeCuAl\(_3\). The ratio of lattice constant \(c/a\) of this single crystal is larger than that of CeCuAl\(_3\) and the stretch of \(c\) axis enhances the energy of \(| \pm \frac{3}{2} >\). On the other hand, CeAuAl\(_3\) has the similar \(c/a\) to that of CeCuAl\(_3\) and has ground state \(| \pm \frac{1}{2} >\) and first excited state \(| \pm \frac{3}{2} >\) [13]. Since the \(c/a\) of CeCu\(_{x}\)Al\(_{4-x}\) becomes large with increasing \(x\), CeCu\(_{x}\)Al\(_{4-x}\) may be in the system occurring crossover of exchanging the ground and first excited state of CEF. The single crystal of CeCuAl\(_3\) grown by Czochralski method, whose \(x\) value is a little less than 1, may be in the critical point of crossover. In order to clarify this hypothesis, the electric resistivity measurements of a single crystal grown by the Czochralskii method under pressure are now in progress.

4. Summary
We have performed ac susceptibility and electrical resistivity measurements of polycrystalline CeCuAl\(_3\) under pressure and CeCu\(_{x}\)Ag\(_{1-x}\)Al\(_3\) at ambient pressure. CeCuAl\(_3\) shows a shoulder in \(\chi_{ac}T(T)\) and a kink in \(\rho(T)\) at around 3.5 K at ambient pressure, and the temperature increases gradually with pressure like a peak in \(\chi_{dc}(T)\) of previous magnetization measurements [9]. These anomalies are attributed to a localized antiferromagnetic transition from Doniach’s phase diagram. The increase in \(T_N\) by applying pressure is appropriate when \(T_N\) of CeAuAl\(_3\) and CeCu\(_{x}\)Ag\(_{1-x}\)Al\(_3\) increase with contracting its cell volume. The temperature of a broad peak in \(\rho(T)\) at around 10 K increases monotonically with pressure. We suggest that this peak is caused by the small CEF splitting combined with Kondo effect and the increase in the broad peak temperature is due to enhancement of the first excited state of CEF by applying pressure. CeCu\(_{x}\)Al\(_{4-x}\) may be a system whose CEF ground changes by \(x\) substitution.

References
[1] Kimura N, Ito K, Saitoh K, Umeda Y and Aoki H 2005 Phys. Rev. Lett. 95 247004
[2] Sugitani I et al. 2006 J. Phys. Soc. Jpn. 75 043703
[3] Settai R, Sugitani I, Okuda Y, Thamizhavel A, Nakashima M, Onuki Y and Harima H 2007 J. Magn. Magn. Mater. 310 84
[4] Anderson P W 1984 Phys. Rev. B 30 400
[5] Moze O and Buschow K H J 1996 J. Alloys Comp. 245 112
[6] Mock S, Pfeilerer C and Löhneysen H v 1999 J. Low. Temp. Phys. 115 1
[7] Kontani M, Sugihara N, Murase K and Mori N 1996 Czech. J. Phys. 46 (S4) 2067
[8] Kontani M, Ido H, Ando H, Nishioka T and Yamaguchi Y 1994 J. Phys. Soc. Jpn 63 1652
[9] Nishioka T, Kawamura Y, Kato H, Matsumura M, Kodama K and Sato N K 2007 J. Magn. Magn. Mater. 310 e12
[10] Werner D, Bauer E, Martin J M and Lees M R 1999 Physica B 259 10
[11] Paschen S, Felder E and Ott H R 1998 Eur. Phys. J. B 2 169
[12] Oe K, Kobayashi R, Nishioka T, Kato H, Matsumura K and Kodama K 2008 J. Phys.: Conf. Series to be published
[13] Sugawara H, Saha S R, Matsuda T D, Aoki Y, Sato H, Gavilano J L and Ott H R 1999 Physica B 259 16