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### Pressure-enhanced magnetoresistance of $\alpha$ -Ce single crystal

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#### Abstract

The electrical resistivity and magnetoresistance (MR) of single crystalline  $\alpha$ -Ce have been measured under high pressure and low temperature. It is found that the temperature dependence of resistivity  $\rho(T)$  at low temperature is described as  $\rho(T) = \rho_0 + AT^2$  below 10 K, where  $\rho_0$  is the residual resistivity and A is the constant. A decreases significantly as pressure increases, which corresponds to the increase of Kondo temperature  $T_K$  at high pressure. A seems to be divergent from higher pressures toward 0.3 GPa, which suggests the existence of magnetic phase boundary. MR ratio of  $\alpha$ -Ce is positive and enhanced by applying pressure. These results are discussed on the basis of theoretical model presented by Kawakami–Okiji.

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### 1. Introduction

Ce metal has various phases depending on the temperature and pressure. Below 5 GPa and room temperature, three phases have been reported, which are called as  $\alpha$  (f.c.c.),  $\beta$ (dhcp) and  $\gamma$  (f.c.c.) phases [1]. At room temperature and ambient pressure,  $\gamma$ -Ce is stable having a local magnetic moment of 2.52 $\mu$ B.  $\gamma$ -Ce shows an isostructural phase transition to  $\alpha$ -Ce at 0.7 GPa at room temperature or at about 110 K at ambient pressure. This transition is accompanied by a drastic change in volume, resistance and magnetic susceptibility [2– 4].  $\alpha'$ -Phase is stabilized above 5 GPa at room temperature and shows a superconductivity near 1.7 K [5]. The electronic properties of  $\alpha$ -Ce show the intermediate properties between  $\gamma$ -Ce and  $\alpha'$ -Ce.  $\alpha$ -Ce has been considered as an exchange enhanced paramagnet such as Pd [4].

However it is very difficult to obtain a single phase of  $\alpha$ -Ce at ambient pressure since there is another phase below

\* Corresponding author. E-mail address: miyagawa@gemini.rc.kyushu-u.ac.jp (H. Miyagawa). room temperature and below about 0.3 GPa, which is called  $\beta$ -Ce showing an antiferromagnetic order at 12.5 K. Since a contamination of  $\beta$ -Ce in  $\alpha$ -Ce influences strongly the magnetic and transport properties of  $\alpha$ -Ce, there have been many discrepancies between experimental results on  $\alpha$ -Ce [6,7]. For this reason a cooling under hydrostatic pressure above at least 0.3 GPa is needed to obtain a single phase and non-strained  $\alpha$ -Ce [6,7].  $\alpha$ -Ce has been considered as a Kondo substance showing an intermediate valence and having high Kondo temperature  $T_{\rm K}$  of several hundreds K [8].

In the present work, we have measured the electrical resistance and MR of Ce under high pressure in detail by using single crystal and newly designed high pressure apparatus in order to get deep insight into the electronic properties of  $\alpha$ -Ce.

### 2. Experimental method

Single crystal of  $\gamma$ -Ce was grown by Czochralski pulling method. Hydrostatic pressure up to 2.8 GPa was generated by

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piston-cylinder device and a 1:1 mixture of Fluorinert, FC 70 and FC 77, as a pressure transmitting medium [9]. Pressure was kept constant within  $\pm 0.1$  GPa throughout the experiment by controlling the oil pressure of hydraulic press. The electrical resistance was measured in the temperature range between 2 and 285 K by a standard dc four probe method in which four copper wires (diameters 50 µm) were spotwelded on the sample. The measurements were performed using superconducting magnet with field up to 9 T. The direction of current was parallel to [100]. The magnetic field was perpendicular to the direction of current.

### 3. Experimental result

# 3.1. Temperature dependence of electrical resistivity at high pressure

Fig. 1 shows the temperature dependence of electrical resistivity  $\rho(T)$  of  $\alpha$ -Ce at various pressures. The  $\gamma \rightarrow \alpha$ phase transition is clearly observed around 210 K at 0.4 GPa.  $\rho(T)$  on cooling shows a sudden decrease at this transition having the large hysteresis about 80 K characteristic to the first-order phase transition.  $\rho$  of Ce at room temperature is shown in the inset of Fig. 1 as a function of pressure, in which the  $\gamma \rightarrow \alpha$  transition is clearly seen at 0.7 GPa as a discontinuous change in  $\rho(P)$ . The resistance of  $\gamma$ -Ce at room temperature increases with increasing pressure with the rate  $(1/R) dR/dP = 6.1 \times 10^{-2} \text{ GPa}^{-1}$ . Considering the compressibility  $\kappa(\gamma - \text{Ce}) = 4.8 \times 10^{-2} \,\text{GPa}^{-1}$ [10], the pressure coefficient of  $\rho$  is  $4.5 \times 10^{-2} \,\text{GPa}^{-1}$ . On the other hand, the resistance of  $\alpha$ -Ce at room temperature decreases with increasing pressure with the rate  $(1/R) dR/dP = -10.5 \times 10^{-2} \text{ GPa}^{-1}$ . From the compressibility of  $\alpha$ -Ce is  $\kappa = 5.0 \times 10^{-2} \text{ GPa}^{-1}$  [10], the pressure coefficient of  $\rho$  is  $-12.2 \times 10^{-2} \text{ GPa}^{-1}$ .

 $\rho(T)$  of Kondo substances shows  $T^2$ -dependence at low temperature,  $T \ll T_{\rm K}$  [11]. In order to examine a scattering mechanism in the temperature dependence of  $\rho$ , we analyzed the present data by assuming the equation  $\rho(T) = \rho_0 + AT^n$ , where  $\rho_0$  is the residual resistivity and A is the constant.  $\rho(T) - \rho_0$  versus T is plotted in the logarithmic scale as shown in Fig. 2. The values of n were determined as n = 2.1and 2.0 at 0.4 and 2.8 GPa, respectively. n is almost independent of pressure. Thus the existence of the  $T^2$ -term in  $\alpha$ -Ce is confirmed, which indicates that  $\alpha$ -Ce is described as a Fermi liquid below 10 K.

A is plotted as a function of pressure in Fig. 3. A decreases rapidly with increasing pressure below 1 GPa and tends to saturate above it. Since A is proportional to the square of density of states of conduction electrons at the Fermi level  $N(0)^2$  and  $T_{\rm K}^{-2}$  [12], the large decrease of A corresponds to a large enhancement of  $T_{\rm K}$  by applying pressure. This result is consistent with other heavy Fermion Kondo compounds [13]. From the extrapolation of A versus P curve, A diverges around  $P_{\rm C} \simeq 0.3$  GPa indicating some kind of phase boundary.

## 3.2. Magnetic field dependence of electrical resistivity of $\alpha$ -Ce under high pressure

MR ratio, defined as  $\Delta \rho / \rho(0) = [\rho(H) - \rho(0)] / \rho(0)$ , is plotted in Fig. 4 as a function of *H* at 4.2 K, where  $\rho(0)$ and  $\rho(H)$  are the resistivities at magnetic fields, 0 and *H*, respectively.  $\alpha$ -Ce has the positive MR:MR ratios under 9 T at 0.4, 1 and 2.8 GPa are 23, 36 and 35%, respectively. MR ratio at 1 GPa is almost the same as that of 2.8 GPa. MR ratio at 4.2 K and 9 T is shown in Fig. 5 as a function of pressure. The positive MR is enhanced by applying pressure below 1 GPa, and tends to saturate above 1 GPa. It approaches zero around 0.3 GPa, which corresponds well to  $P_{\rm C}$  showing  $A \rightarrow \infty$ .

MR seems to show almost quadratic behavior against H and may be written as  $\Delta \rho / \rho(0) = BH^m$ . In order to determine the value of m, we plotted  $\Delta \rho / \rho(0)$  as a function of H in the logarithmic scale in Fig. 6. The values of m at 9 T and 4.2 K under 0.4, 1.0 and 2.8 GPa are 1.6, 1.8 and 1.7,



Fig. 1. Temperature dependence of the electrical resistivity of  $\alpha$ -Ce at high pressures. Pressure dependence of the electrical resistivity at 280 K is shown in the inset.



Fig. 2.  $\rho - \rho_0$  vs. T in logarithmic scale at high pressure.

respectively. *m* is found to be independent of pressure. The MR of normal metals shows  $H^2$  dependence. The small deviation from 2 implies that  $\alpha$ -Ce is in an intermediate valence state(IVS) having moderate electron correlation.

### 4. Discussion

## 4.1. Pressure dependence of Kondo temperature and the proximity to the phase boundary

Here we estimate  $T_{\rm K}$  of  $\alpha$ -Ce using the values of A. Kadowaki and Woods reported the universal relationship between A and  $\gamma$  for typical heavy Fermion compounds such as  $A/\gamma^2 = 1.0 \times 10^{-5} \ \mu \ \Omega \ {\rm cm} ({\rm mol} \ {\rm K/mJ})^2$  [14]. Using this and Rajan relation [15], the relationship between A( $\mu \ \Omega \ {\rm cm}/{\rm K}^2$ ) and  $T_{\rm K}$  is obtained

$$T_{\rm K} = \frac{17.9}{\sqrt{A}}.\tag{1}$$



Fig. 3. The coefficients A of the equation,  $\rho = \rho_0 + AT^2$  as a function of pressure.



Fig. 4. MR ratio of  $\alpha$ -Ce at 4.2 K at high pressure.

From the results in Fig. 3, the values of  $T_{\rm K}$  are estimated to be 440 and 663 K at 0.4 and 2.8 GPa, respectively.  $T_{\rm K}$  at 2.8 GPa is about 1.5 times larger than that at 0.4 GPa.

As is mentioned in Section 3.1, *A* becomes infinite near  $P_{\rm C} \simeq 0.3$  GPa. Usually *A* shows a divergence at the critical point for the magnetic instability [16]. In the present case,  $P_{\rm C}$  suggests the existence of the phase boundary between  $\beta$ -Ce and  $\alpha$ -Ce.

#### 4.2. Pressure-induced enhancement of MR

 $\alpha$ -Ce shows the positive MR as shown in Fig. 4. The positive MR is enhanced by applying pressure. Generally speaking, the localized moment system shows a negative MR. In such sense the positive MR of  $\alpha$ -Ce indicates that there is no local moment in  $\alpha$ -Ce. This consideration is consistent with the fact that  $\alpha$ -Ce is in IVS.

Kawakami and Okiji (K–O) explained the mechanism of MR of heavy Fermion substances on basis of the periodic Anderson model [17]. They calculated the field dependence



Fig. 5. MR ratio at 4.2 K and 9 T as a function of pressure.



Fig. 6. A log–log plot of MR ratio vs. H of  $\alpha$ -Ce at 4.2 K.

of MR of the Ce substances as a function of  $H/T_{\rm K}$  at various temperature  $T/T_{\rm K}$ . According to their calculation, the MR ratio increases with increasing  $H/T_{\rm K}$  for small values of  $T/T_{\rm K}$ . Since  $T_{\rm K}$  of  $\alpha$ -Ce is high ( $T_{\rm K} \sim$  several hundreds K) and the present MR was measured at T = 4.2 K, positive MR is observed because  $H/T_{\rm K}$  and  $T/T_{\rm K}$  are of the order of  $10^{-2}$ .

In the present work, MR ratio of  $\alpha$ -Ce at 4.2 K and 9 T is enhanced by applying pressure as shown in Fig. 5. Since  $T_{\rm K}$ rapidly increases with increasing pressure above 0.3 GPa, the value of  $H/T_{\rm K}$  decreases at high pressure. This result gives rise to an increase of MR ratio. Thus the present results are explained qualitatively on the basis of K–O model.

### 5. Conclusion

In this paper, we have measured the electrical resistance at high pressure and magnetic field. The main results are summarized as follows:

- (1) MR ratio of  $\alpha$ -Ce is positive and is increased strongly by applying pressure.
- (2) The existence of  $T^2$ -term in  $\alpha$ -Ce indicates that  $\alpha$ -Ce is described as a Fermi liquid at low temperature.
- (3) A diverges around 0.3 GPa, which suggests an existence of the magnetic phase boundary between  $\alpha$ -Ce and  $\beta$ -Ce.

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