

Effect of pressure on the electrical resistivity of the heavy fermion CeAl_3

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Abstract

The electrical resistivity $\rho(T)$ of the heavy fermion compound CeAl_3 has been measured under high pressure up to 1.5 GPa in order to clarify the volume dependence of the electronic state in the heavy fermion. It is found that the temperature showing a maximum in the $\rho(T)$ curve increases with increasing pressure. Compressibility was also measured using the X-ray diffraction technique at high pressure. On the basis of the data, a quantitative analysis of the volume dependent energy scale is examined using a simple theoretical model.

1. Introduction

It is well known that the application of pressure to Ce or U-based concentrated Kondo (CK) compounds enhances the Kondo temperature T_K [1], which dominates the characteristic electronic state in the CK system. The temperature dependence of the electrical resistivity $\rho(T)$ under high pressure revealed that there is a pressure-induced crossover from the well localized heavy fermion (HF) state to the intermediate valence state for the HF compounds such as CeInCu_2 [2,3] or CeCu_6 [4].

CeAl_3 is well known as a prototypical HF compound, having a large specific-heat coefficient $\gamma \approx 1500 \text{ mJ/mol K}^2$ [5]. We have reported [6] that the substitution of the Ce-site by La reduces T_K and also the density of state at the Fermi level, $N(0)$. This result was interpreted as an interplay between the negative chemical pressure and the decrease in the number of 4f-electrons by alloying with La. We developed a phenomenological treatment of the pressure (or volume) dependence of electrical resistivity of CK or HF compounds, and found that it explains well the results obtained for CeInCu_2 , CeCu_6 [7], etc.

Here we turn our attention to CeAl_3 as an application of our method, since there are few systematic investigations of the effect of pressure (or volume change) on the electrical properties of CeAl_3 [8,9].

The high pressure measurement of the electrical resistivity and lattice constant for the HF compound CeAl_3 were carried out and analyzed quantitatively on the basis of phenomenological theory.

2. Experimental procedure

The polycrystalline CeAl_3 and LaAl_3 samples were prepared by arc melting weighed amounts of the constituent elements in an argon atmosphere and annealing at 900 °C for 10 days.

Electrical resistance under high pressure was measured by using a standard four-probe method up to 1.5 GPa. Hydrostatic pressure was generated using a Cu–Be piston-cylinder device and a 1:1 mixture of Fluorinert, FC 70 and 77 as a pressure transmitting medium. The pressure was changed only at room temperature to minimize an internal strain in the specimen and the load was controlled to within $\pm 1\%$ throughout the measurement [10].

X-Ray diffraction measurements under high pressure were carried out to obtain the pressure–volume relation. Hydrostatic pressure for the X-ray study was generated using Bridgman-type tungsten-carbide opposed anvils with a face of 3 mm in diameter. The powdered sample and NaCl were placed in a 0.3-mm hole at the center of a beryllium disk 0.5 mm in thickness. A 4:1 methanol/ethanol mixture was used as the pressure transmitting medium. The pressure was determined by Decker's equation of state for NaCl [11].

3. Results and discussion

The temperature dependence of the electrical resistivity $\rho(T)$ at various pressures up to 1.5 GPa and $\rho(T)$ of LaAl_3 at ambient pressure are shown in Fig. 1. ρ of LaAl_3 has a normal temperature dependence as observed in non-magnetic metal. At ambient pressure, ρ of CeAl_3 has a maximum at 35 K and shows a shoulder

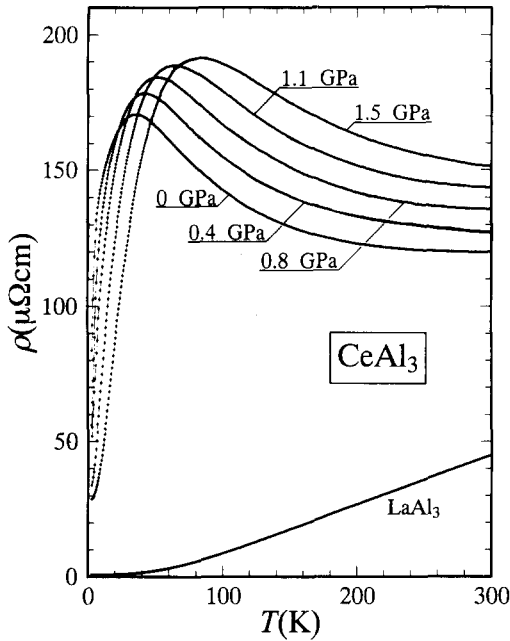


Fig. 1. The electrical resistivity ρ of CeAl₃ under high pressure as a function of temperature. ρ of LaAl₃ at ambient pressure is also shown for comparison.

near 6 K. This behavior is typically observed in non-cubic HF systems because of the crystalline electric field, implying an appearance of the two temperature regions where a logarithmic resistivity is observed. The maximum is shifted towards the higher temperature, which is in qualitative agreement with previous studies [8,9], and the height is enhanced as the pressure is increased. Since the ρ of the CK system is considered to have a maximum at a temperature that is roughly proportional to the Kondo temperature T_K [12], increase in T_K is associated with increasing hybridization between the 4f- and conduction electrons by applying pressure.

The magnetic contribution to the resistivity by 4f-electrons was estimated by the relation $\rho_{\text{mag}} = \rho(\text{CeAl}_3) - \rho(\text{LaAl}_3)$. Figure 2 shows ρ_{mag} as a function of $\log T$. It is assumed that $\rho(\text{LaAl}_3)$ shows the phonon contribution and its pressure dependence is negligibly small. At high pressure above 1.1 GPa, the shoulder-like anomaly levelled out and a typical T^2 -term in ρ appears instead. It is reported that the T^2 -dependence is obtained below 0.3 K and its coefficient equals $35 \mu\Omega\text{-cm/K}^2$ at ambient pressure [5]. At 1.5 GPa, the temperature range of the T^2 -dependent resistivity is extended up to 10 K, while the coefficient is estimated as $0.17 \mu\Omega\text{-cm/K}^2$, about 200 times smaller than that at ambient pressure. The result suggests that the $N(0)$ decreases markedly at high pressure because the coefficient of the T^2 -term is proportional to $N(0)^2$. The decrease in $N(0)$ by pressure was also reported for CeAl₃ in the measurement of the thermal expansion coefficient under high pressure [13].

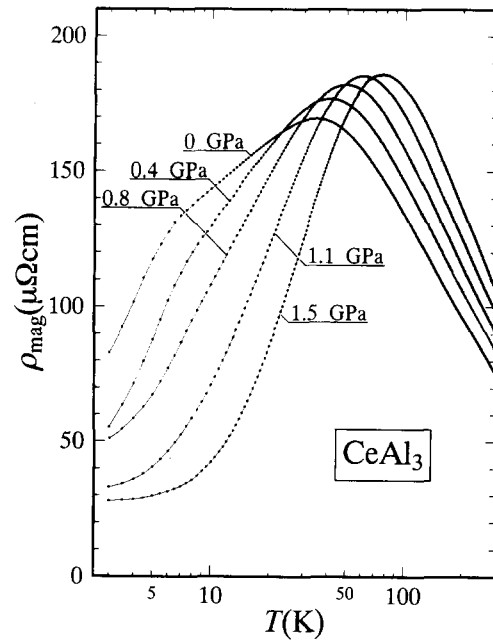


Fig. 2. The magnetic part ρ_{mag} as a function of $\log T$ at various pressures.

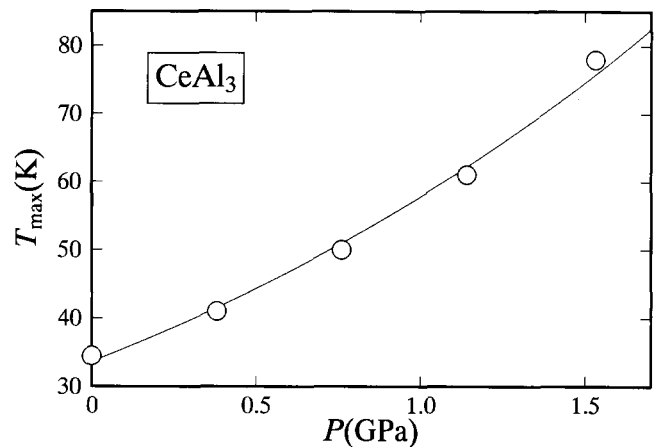


Fig. 3. The pressure dependence of the temperature T_{max} at which ρ_{mag} has a maximum.

The maximum temperature T_{max} in $\rho_{\text{mag}}(T)$ increases with increasing pressure as seen in Fig. 3. The increasing rate is larger at high pressures than at low pressures. This result implies a non-linear pressure dependence of T_K . T_K at 1.5 GPa is expected to be greater than twice that at ambient pressure because T_{max} is proportional to T_K . The solid line is drawn by a least-square fitting (see below).

As seen in Fig. 2, the logarithmic dependence of ρ_{mag} on temperature is observed in the wide range above the T_{max} at various pressures. Figure 4 illustrates the absolute value of the slope in ρ_{mag} vs. $\log T$ curve, $m = |\partial\rho_{\text{mag}}/\partial(\log T)|$. m increases with pressure by $\sim 35\%$ at 1.5 GPa. In the single magnetic impurity model, m is proportional to $|JN(0)|^3$, where J is the s-f exchange

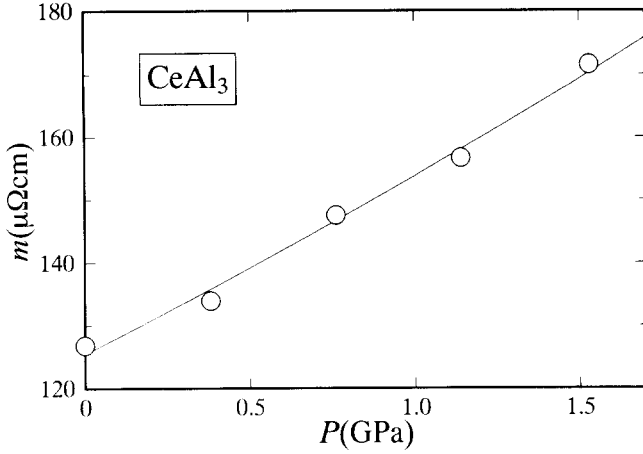
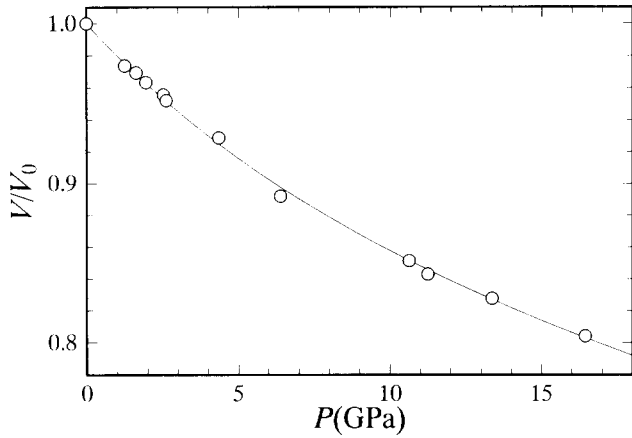
Fig. 4. The absolute value of the slope in ρ_{mag} vs. $\log T$ plot.

Fig. 5. The relative change of the volume as a function of pressure at room temperature.

interaction and $N(0)$ is the density of state at Fermi level. The increase in m results in the enhancement of $|JN(0)|$ and hence the enhancement of T_K assuming $T_K \propto \exp(-1/|JN(0)|)$. This is qualitatively consistent with the result in Fig. 3. Since $N(0)$ decreases with increasing pressure as mentioned above, the enhancement of $|JN(0)|$ indicates that the value of J increases more rapidly than the decrease in $N(0)$ with pressure.

Next we show the compression curve at room temperature in Fig. 5. The Ni₃Sn-type hexagonal structure is stable up to 16 GPa, because no new diffraction lines were observed. The relative change in volume, V/V_0 , decreases smoothly without discontinuity with increasing pressure, where V and V_0 are the volume at high and ambient pressure, respectively. The solid line in Fig. 5 shows the result of a least-square fit to the first order Murnaghan's equation of state,

$$P = \frac{B_0}{B_0'} \left[\left(\frac{V}{V_0} \right)^{-B_0'} - 1 \right] \quad (1)$$

where B_0 and B_0' are the values of the isothermal bulk modulus and its pressure derivative at ambient pressure, respectively. We obtained $B_0 = 47.8$ GPa and $B_0' = 3.8$. The B_0 of CeAl₃ is about half of that of CeInCu₂ but B_0' is nearly the same [14].

A brief analysis of the present data is shown in the following to elucidate the volume dependence of the characteristic energy scale. According to Yoshimori and Kasai [12], T_{max} is proportional to T_K . Let q denote the Grüneisen constant of $|JN(0)|$,

$$q = - \left. \frac{\partial \ln |JN(0)|}{\partial \ln V} \right|_{V=V_0} \quad (2)$$

and taking account of the relation $T_K \propto \exp[-1/|JN(0)|]$, the Grüneisen constant of T_K is $q/|JN(0)|_0$. Then we have

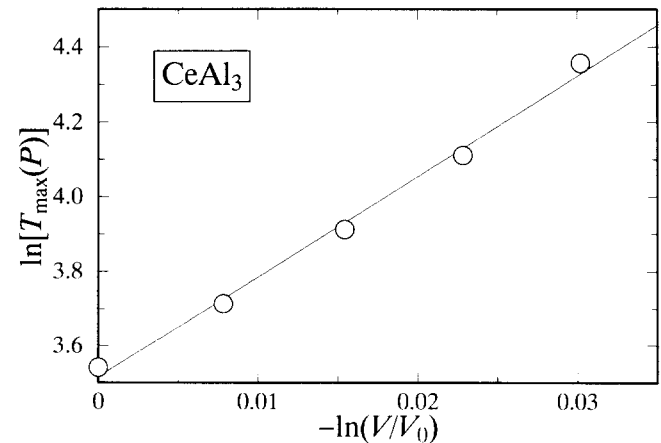
$$\ln \left[\frac{T_{\text{max}}(P)}{T_{\text{max}}(0)} \right] = \frac{q}{|JN(0)|_0} \ln \frac{V_0}{V} \quad (3)$$

where $|JN(0)|_0$ is the value of $|JN(0)|$ at ambient pressure. Figure 6 shows the least-square fitted $\ln[T_{\text{max}}(P)]$ vs. $-\ln(V/V_0)$ plot, where pressure is used as the implicit variable and $T_{\text{max}}(0)$ is the extrapolated value of $T_{\text{max}}(P)$ to ambient pressure, which was given by the intercept in Fig. 6. The slope of the solid line in Fig. 6 gives the Grüneisen constant $q/|JN(0)|_0 = 26.9$.

Furthermore, considering the relation $m \propto |JN(0)|^3$, the following equation is derived:

$$\frac{1}{3} \ln \left[\frac{m(P)}{m(0)} \right] = q \frac{V_0 - V}{V_0} \quad (4)$$

A similar analysis using eqn. (4) as carried out in Fig. 6 leads to a value of q of 2.66. Therefore, we obtain $|JN(0)|_0 = 0.12$, which is a typical value for CK or HF compounds such as CeInCu₂ [2,7], CeCu₆ [4,7] or CePtSi₂ [15]. Thus, the phenomenological treatment of

Fig. 6. The value of $\ln[T_{\text{max}}(P)]$ as a function of $-\ln(V/V_0)$.

the present results explains the characteristics of the HF compound CeAl₃.

4. Conclusion

The maximum resistivity temperature T_{\max} and the absolute value of the slope in the ρ_{mag} vs. $\log T$ plot increase with increasing pressure. This implies that the application of pressure enhances the Kondo temperature or $|N(0)|$. The volume dependence of the energy scale is explained well with this phenomenological treatment.

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