

**Electronic states of single crystal CeAl<sub>2</sub> near the pressure-induced quantum critical point**

Hidenori Miyagawa and Gendo Oomi

*Department of Physics, Kyushu University, Ropponmatsu, Chuo-ku, Fukuoka 810-8560, Japan*

Masashi Ohashi

*Faculty of Environmental Design, Kanazawa University, Kanazawa, Ishikawa 920-1192, Japan*

Isamu Satoh

*Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

Takemi Komatsubara

*Center for Low Temperature Science, Tohoku University, Sendai 980-8577, Japan*

Masato Hedo

*Faculty of Science, University of the Ryukyus, Nishihara, Okinawa 903-0213, Japan*

Yoshiya Uwatoko

*Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan*

(Received 25 February 2008; revised manuscript received 29 May 2008; published 5 August 2008)

The electrical resistivity and magnetoresistance (MR) of single crystal CeAl<sub>2</sub> have been measured up to 8 and 3.2 GPa, respectively. The magnetic part of the electrical resistivity  $\rho_{\text{mag}}$  increases as temperature decreases until it shows two maxima around 5 K(=T<sub>1</sub>) and 63 K(=T<sub>2</sub>) at ambient pressure. As pressure increases, T<sub>1</sub> and T<sub>2</sub> decrease up to 2 and 3 GPa, respectively, but they begin to increase above these pressures. MR changes from negative to positive with increasing pressure. It is suggested that Kondo temperature T<sub>K</sub> increases as pressure increases. Above 3 GPa, Fermi-liquid behavior obeying T<sup>2</sup> dependence of the electrical resistivity is observed and the coefficient of T<sup>2</sup> dependence diverges around 2.7 GPa. The Néel temperature of CeAl<sub>2</sub> at ambient pressure is 3.8 K(=T<sub>N</sub>) and it is suppressed to zero around 2.7 GPa. In the pressure range of 2.5 GPa ≤ P ≤ 2.7 GPa,  $\rho_{\text{mag}}(T)$  deviates largely from the normal Fermi-liquid behavior, which suggests the so-called non-Fermi-liquid behavior around 2.7 GPa. Pressure-induced superconductivity was not observed down to 50 mK around 2.7 GPa.

DOI: [10.1103/PhysRevB.78.064403](https://doi.org/10.1103/PhysRevB.78.064403)

PACS number(s): 75.20.Hr, 73.43.Nq, 74.62.Fj

**I. INTRODUCTION**

The 4*f* electrons in Ce compounds have been well known as showing various interesting physical properties such as magnetic ordering, valence fluctuation, heavy fermion owing to competition between the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction, and Kondo effect.<sup>1,2</sup> The former interaction enhances a magnetic ordered state and the 4*f* electron is treated as a localized one. The latter gives rise to a screening of the 4*f* local moment by the conduction electrons and leads to the itinerant nonmagnetic state. It has been well known that RKKY interaction and Kondo effect are sensitive to pressure.<sup>3</sup> Through the investigation under high pressure, we can get a lot of useful information about the mechanism of Kondo or heavy fermion states.

In several Ce compounds, the unconventional physical properties have been observed around quantum critical point (QCP) which is the point that the ordering temperature is tuned to zero by adjusting a control parameter such as high pressure or chemical composition and so on.<sup>4–6</sup> Around QCP, the anomalous temperature dependence of the specific heat and the resistivity, which deviate strongly from normal Fermi-liquid behavior, have been observed. This is called as non-Fermi-liquid behavior. On the other hand, Fermi-liquid behavior is recovered in the nonmagnetic region obtained by

changing parameter further across QCP. Until now, we have reported several examples showing QCP in Ce compounds by measuring the electrical resistivity under high pressure.<sup>7–10</sup>

CeAl<sub>2</sub> with the cubic Laves phase MgCu<sub>2</sub> structure is well known as a typical heavy fermion compound, which has the large electronic specific-heat coefficient  $\gamma = 135$  mJ/mol K<sup>2</sup>.<sup>11</sup> Antiferromagnetic order is observed at T<sub>N</sub>=3.8 K,<sup>12–15</sup> which is due to the formation of spin-density wave (SDW). The Ce ion in CeAl<sub>2</sub> is trivalent at ambient pressure. Kondo temperature T<sub>K</sub> and the crystal electric field (CEF) splitting energies were estimated to be 6 K,  $\Delta_1 = 100$  K, and  $\Delta_2 = 180$  K from the neutron-scattering measurement.<sup>11,16</sup> Although the CEF of cubic Ce compounds should split the J=5/2 ground state into a  $\Gamma_7$  doublet and a  $\Gamma_8$  quartet, the CEF splitting in CeAl<sub>2</sub> shows three levels. This was explained by assuming an existence of bound state between a crystal-field excitation and a low-lying phonon by strong magnetoelastic coupling.<sup>17</sup>

Electrical resistivity under high pressure has been reported by several authors.<sup>7,8,18,19</sup> T<sub>N</sub> decreases with increasing pressure. T<sub>K</sub> increases as pressure increases and the magnitude of CEF splitting decreases with increasing pressure.<sup>7,18</sup> T<sup>2</sup> dependence in electrical resistivity which is a characteristic of Fermi-liquid behavior was observed above 3

GPa. The coefficient  $A$  of  $T^2$  term diverges around 3 GPa, which suggests an existence of QCP around 3 GPa. The electrical resistivity at various temperatures was measured up to 13 GPa by using quasihydrostatic pressure,<sup>19</sup> but details of the electronic states have not been reported.

In the present work, we have measured the electrical resistivity up to 8 GPa and magnetoresistance (MR) up to 3.2 GPa to examine the stability of Kondo or heavy fermion states of  $\text{CeAl}_2$  by controlling the competition between RKKY interaction and Kondo effect using high pressure. Since  $T_N$  is decreased by applying pressure, we made an attempt to induce QCP at high pressure to look for the development of different ground states such as superconductivity or non-Fermi-liquid state by carrying out the detailed analysis of the electrical resistivity.

## II. EXPERIMENTAL METHOD

Single crystals of  $\text{CeAl}_2$  and  $\text{LaAl}_2$  were grown by Czochralski pulling method with the typical size of 5 mm in diameter and 50 mm in length. The electrical resistivity was measured with the applying current parallel to the  $a$  axis in the temperature range between 2 and 300 K by a standard four-probe method using copper wires (diameters 50  $\mu\text{m}$ ) or gold wires (diameters 20  $\mu\text{m}$ ). Hydrostatic pressures up to 3.2 and 8.0 GPa were generated using a piston-cylinder device<sup>20</sup> and a cubic anvil device,<sup>21</sup> respectively. A 1:1 mixture of Fluorinert, FC70, and FC77 was used as a pressure transmitting medium. The pressure inside the cell was kept constant in all temperature ranges in the present work by automatic controlling of the load. The electrical resistivity around 3 GPa down to 50 mK was measured by means of dilution refrigerator and clamp-type pressure cell. The magnetic field was applied perpendicularly to the current direction up to 9 T.

## III. EXPERIMENTAL RESULTS

### A. Kondo scattering under high pressure

Figure 1(a) shows the temperature dependent electrical resistivity  $\rho(T)$  of  $\text{CeAl}_2$  at various pressures and nonmagnetic  $\text{LaAl}_2$  at ambient pressure for comparison. At ambient pressure,  $\rho(T)$  of  $\text{CeAl}_2$  decreases with decreasing temperature and shows a peak around 5 K and a shoulder around 60 K. The peak shifts to lower temperature and disappears above 2.5 GPa. On the other hand, the shoulder is enhanced at high pressure becoming a broad peak centered around 50 K at 3 GPa. The broad peak becomes less prominent by applying higher pressure.

The magnetic part of electrical resistivity  $\rho_{\text{mag}}(T)$  was estimated by subtracting phonon contribution using the  $\rho(\text{LaAl}_2)$ ,  $\rho_{\text{mag}}(T) = \rho(\text{CeAl}_2) - \rho(\text{LaAl}_2)$ , where we assume that the phonon part of  $\rho(\text{CeAl}_2)$  is approximated by  $\rho(\text{LaAl}_2)$ .  $\rho_{\text{mag}}(T)$  curves are shown in Fig. 1(b) as a function of temperature in the logarithmic scale.  $T_N$  at ambient pressure was shown by an arrow.  $\rho_{\text{mag}}(T)$  at ambient pressure has two maxima due to combination of Kondo effect and CEF splitting, which were observed up to 2.4 GPa. These peaks become a single peak above 2.5 GPa through a shoulder in

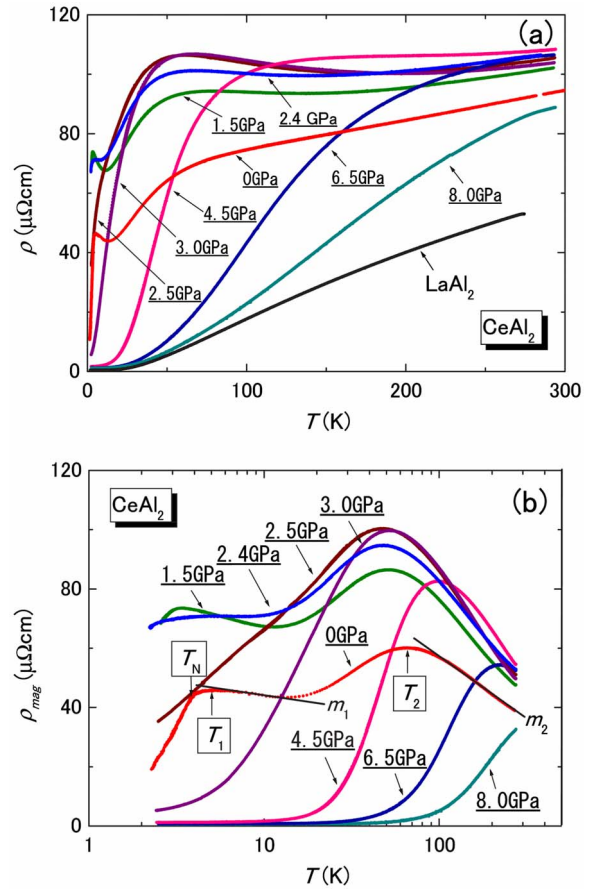


FIG. 1. (Color online) (a) Temperature dependence of the electrical resistivity under high pressure. (b) The temperature dependence of  $\rho_{\text{mag}}$ .  $T_N$  at ambient pressure was shown by an arrow.  $T_1$  and  $T_2$  show the temperature of two maxima.

the  $\rho(T)$  around 2.5 GPa.  $\rho_{\text{mag}}(T)$  is found to show  $-\log T$  dependence characteristic to Kondo effect in the temperature ranges  $7 \text{ K} \leq T \leq 20 \text{ K}$  and  $100 \leq T \leq 300 \text{ K}$ . The coefficients of  $-\log T$  are defined to be  $m_1$  and  $m_2$  as shown in Fig. 1(b). Figure 2(a) shows the pressure dependence of  $T_1$ ,  $T_2$ , and  $T_N$ . Here,  $T_1$  and  $T_2$  were determined by the temperature dependence of first derivative of  $\rho_{\text{mag}}(T)$  as the temperatures of  $d\rho_{\text{mag}}/dT=0$ , which are shown in Fig. 2(b). Above 2.4 GPa, a shoulder in  $d\rho_{\text{mag}}/dT$  is still observed around 10 K up to 2.7 GPa. The temperature showing shoulder  $T_s$  was defined as the minimum or the point showing a sudden increase of  $d\rho_{\text{mag}}/dT$ .  $T_s$  [shown by the triangle in Fig. 2(a)] is found to fall on the curve extrapolated from low pressure and shifts to higher temperature as pressure increases. It is interesting to note that  $\rho_{\text{mag}}(T_2)$  increases below 2.5 GPa with pressure, while it decreases above 2.5 GPa. It is found that  $T_2$  decreases with pressure below 3 GPa, but begins to increase above 3 GPa. The similar behavior for  $T_1$  and  $T_2$  has been reported in other Ce compounds such as  $\text{CeCu}_2\text{Si}_2$  and  $\text{CeCu}_2\text{Ge}_2$ .<sup>4,22</sup> The details of this pressure effect will be discussed later.

Figure 3 shows  $\rho_{\text{mag}}$  above 2.5 GPa as a function of  $T^2$ . Above 3 GPa,  $T^2$  dependent electrical resistivity is observed below 10–20 K. This means that Fermi-liquid state is stable above 3 GPa and stabilized more as pressure increases since

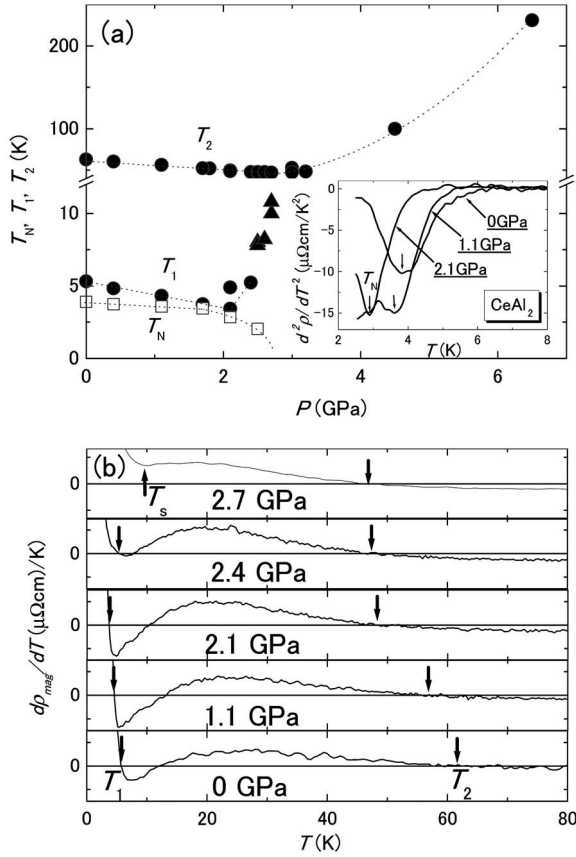


FIG. 2. (a)  $T_1$ ,  $T_2$ , and  $T_N$  as a function of pressure. The closed circles show the pressure dependence of  $T_1$  and  $T_2$ , and the closed triangles show that of the temperature where a shoulder is observed in  $d\rho_{\text{mag}}/dT$ . The open squares show  $T_N$  as a function of pressure. The dashed line is a guide to the eyes. Second derivative of  $\rho(T)$  at high pressures is shown in the inset as a function of temperature. (b) The temperature dependence of first derivative of  $\rho_{\text{mag}}(T)$ . The arrows show  $T_1$ ,  $T_2$ , and  $T_s$ . Here,  $T_1$  and  $T_2$  were determined as the temperatures of  $d\rho_{\text{mag}}/dT=0$ , and  $T_s$  was determined as the temperature showing a minimum of  $d\rho_{\text{mag}}/dT$ .

the temperature range showing  $T^2$  dependence increases with pressure. The pressure dependence of the coefficient  $A$  of  $T^2$  term is shown in Fig. 4.  $A$  decreases rapidly as pressure increases and seems to diverge around 3 GPa. The previous data in CeAl<sub>2</sub> are plotted also in Fig. 4.<sup>7</sup>

### B. Effect of pressure on the antiferromagnetic ordering temperature $T_N$

A sudden decrease due to antiferromagnetic ordering is observed in  $\rho(T)$  as shown in Fig. 1(b). The antiferromagnetic ordering temperature  $T_N$  was defined as the minimum in the temperature dependence of second derivative of  $\rho(T)$  since  $T_N$  at ambient pressure ( $T_N=3.8$  K) in this work is in agreement with that obtained by neutron-scattering experiment ( $T_N=3.8$  K).<sup>13</sup> The temperature dependence of  $d^2\rho(T)/dT^2$  is shown in the inset of Fig. 2(a).

The pressure dependence of  $T_N$  is shown in Fig. 2(a).  $T_N$  decreases slightly up to 2 GPa as pressure increases and decreases rapidly above 2 GPa. Considering that the anomaly

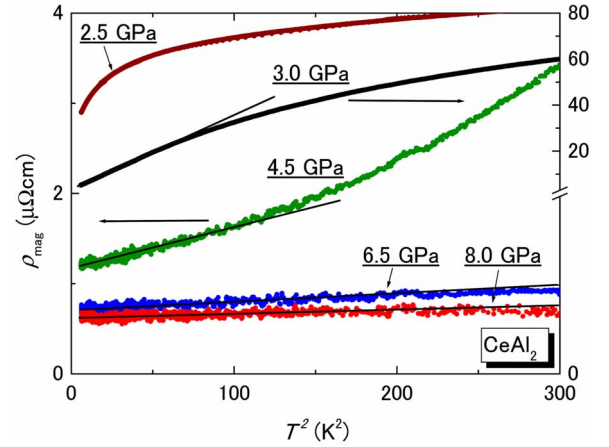


FIG. 3. (Color online)  $T^2$  dependence of  $\rho_{\text{mag}}$  under high pressure.

in the  $\rho(T)$  due to antiferromagnetic ordering becomes unclear above 2.5 GPa,  $T_N$  may disappear above 2.5 GPa. It has been reported that the positive pressure coefficient of  $T_N$  is predicted using the data of thermal-expansion measurement<sup>23</sup> in which  $T_N$  shows a maximum around 0.5 GPa and then  $T_N$  is decreased by applying higher pressure. However,  $T_N$  in the present work decreases as pressure increases without any peak in the pressure dependence. More experimental data are settled this point. The pressure where  $T_N=0$  is obtained to be 2.7 GPa by a smooth extrapolation of the data as shown by dotted curve which indicates that QCP in CeAl<sub>2</sub> exists around 2.7 GPa.

### C. Temperature dependence of the electrical resistivity around QCP

As was mentioned in Sec. III B, the pressure-induced QCP is considered to be observed around 2.7 GPa. In order to investigate the detailed behavior of  $\rho(T)$  at low temperature below 2 K, we have measured  $\rho(T)$  down to 50 mK, which is shown in Fig. 5.  $\rho(T)$  changes significantly as pressure increases: the  $\rho(T)$  at 2.1 GPa has a broad peak around 4 K followed by a rapid decrease below that due to magnetic

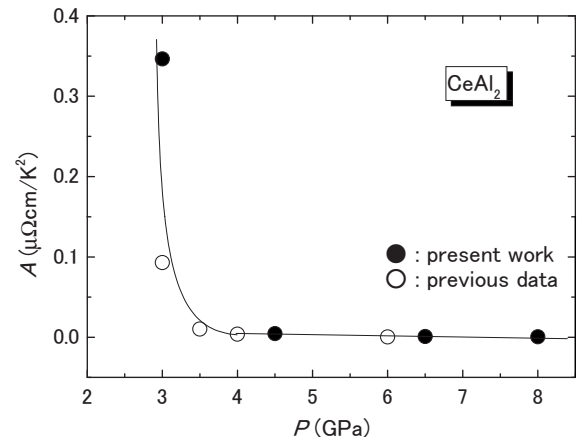


FIG. 4. Pressure dependence of the coefficient  $A$  of  $T^2$ . The solid line is a guide to the eyes.

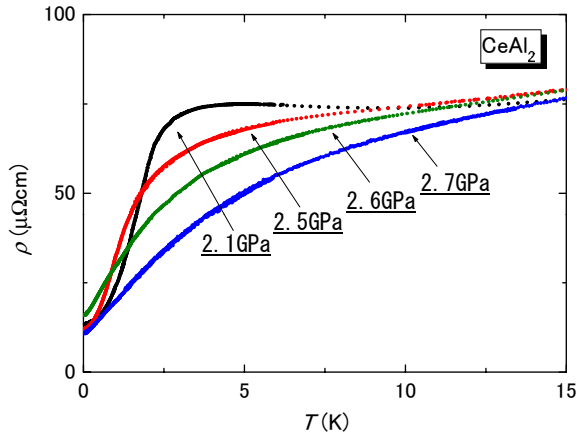


FIG. 5. (Color online)  $\rho(T)$  under high pressure down to 50 mK.

ordering, but as pressure increases, the peak disappears and then  $\rho(T)$  above 2.6 GPa shows a smooth increase with temperature.  $\rho_{\text{mag}}(T)$  is shown in Fig. 6. At 2.1 GPa, a peak is observed around 5 K.  $T_1$  becomes unclear above 2.5 GPa and the shoulder is observed. Figures 1(b) are in good agreement within experimental error.

In some Ce compounds, the superconductivity has been observed around QCP,<sup>3,24</sup> which are known as the unconventional superconductivity such as  $d$  or  $p$  wave superconductivities. In the present work, no superconductivity was observed down to 50 mK at 2.7 GPa. Recently, it has been reported that the superconductivity is induced around  $P_v$  by valence fluctuation, where  $P_v$  is the pressure showing it. Since  $P_v$  of CeAl<sub>2</sub> may be higher than 2.7 GPa, a valence fluctuation mediated superconductivity is expected at higher pressure than 3 GPa.

**D. Magnetoresistance under high pressure**

Figure 7 shows the electrical resistivity  $\rho(H)$  at 4.2 K as a function of magnetic field  $H$  under high pressure. Extremely large MR is observed, in which  $\rho$  is decreased about 40% by applying 9 T. All  $\rho(H)$  curves show smooth change against

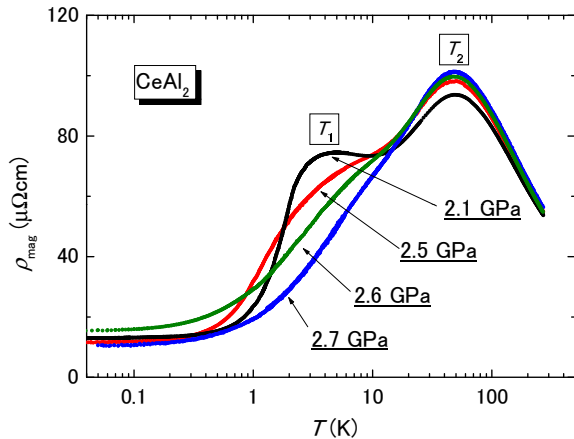


FIG. 6. (Color online)  $\rho_{\text{mag}}(T)$  at high pressure in the range  $2.1 \leq P \leq 2.7$  GPa.

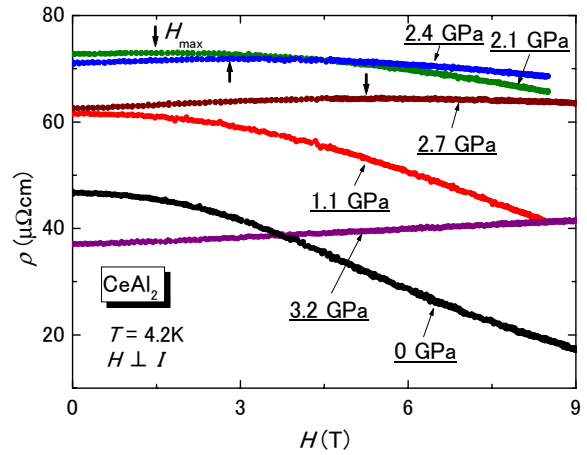


FIG. 7. (Color online) The electrical resistivity as a function of magnetic field.

$H$ . This is due to the fact that no metamagnetic transition is observed at 4.2 K because the antiferromagnetic ordering occurs at 3.8 K at ambient pressure. Below 1.1 GPa, negative MR is observed up to 9 T, while at 2.1, 2.4, and 2.7 GPa, MR is positive at low magnetic field followed by a broad maximum around  $H_{\text{max}}$ , which is determined as the magnetic field of  $d\rho/dH=0$  as shown in Fig. 8.  $\rho(H)$  at 3.2 GPa shows a positive MR up to 9 T. Figure 8 shows the  $H$  dependence of the first derivative of  $\rho(H)$ ,  $d\rho/dH$ . At ambient pressure, the values of  $d\rho/dH$  show a minimum around 5 T, which indicates that MR tends to saturate above 5 T. At 1.1 GPa, the values of  $d\rho/dH$  decrease up to 8.5 T without any minimum and the change of sign at  $H_{\text{max}}$  is observed around 1.5 T at 2.1 GPa.  $H_{\text{max}}$  shifts to high magnetic field as pressure increases, which is not observed up to 9 T at 3.2 GPa. Thus the sign of MR in CeAl<sub>2</sub> changes from negative to positive at high pressure. In order to examine the change of the resistivity by applying  $H$ , the values of  $d\rho/dH$  are plotted in Fig. 9 as a function of pressure at  $H=3, 6,$  and  $8.5$  T. The signs of  $d\rho/dH$  are found to change around 2.5, 2.7, and 2.9 GPa, respectively, which indicate that a crossover in the electronic state takes place around these pressures.

Negative MR indicates the incoherent Kondo state but positive MR indicates the coherent one.<sup>25</sup> Although 4.2 K is

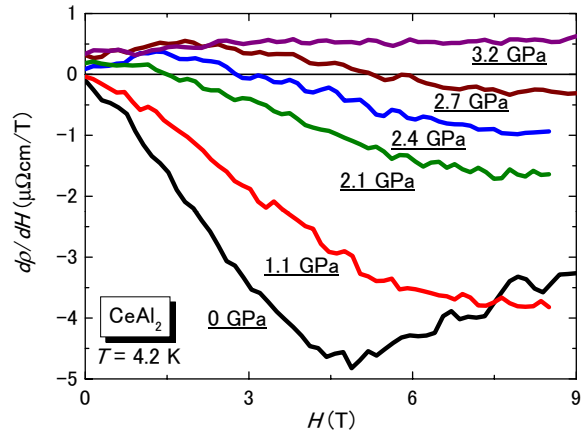


FIG. 8. (Color online) The magnetic-field dependence of first derivative of  $\rho(H)$ .



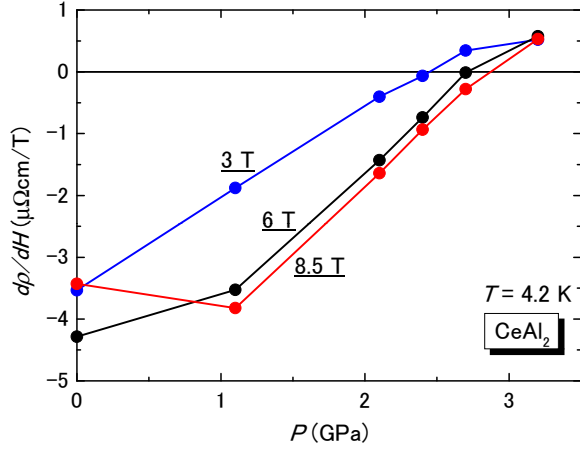


FIG. 9. (Color online) Pressure dependence of the value of  $dp/dH$  at 3, 6, and 8.5 T.

below  $T_K=6$  K, the electronic state in CeAl<sub>2</sub> at 4.2 K and ambient pressure is considered not to be stabilized as the coherent Kondo state. The mechanism of MR of heavy fermion substances has been explained on the basis of the periodic Anderson model by Kawakami and Okiji.<sup>26</sup> They calculated MR on the Ce substances as a function of  $H/T_K$  at various temperatures  $T/T_K$ . According to their results, the reduction of  $T/T_K$  induces the change from negative to positive MR. In this work,  $T/T_K$  decreases with increasing pressure since positive MR becomes stable at high pressure above 2.1 GPa. The reduction of  $T/T_K$  corresponds to an increase of  $T_K$  since  $T$  is constant (4.2 K) in the present work. The coherent Kondo state is considered to be stable above 3.2 GPa and 4.2 K since positive MR is observed without a maximum. The change from negative MR to positive MR is explained as a crossover from the incoherent Kondo state to the coherent one with increasing  $T_K$ .

#### IV. DISCUSSION

##### A. Effect of pressure on the Kondo scattering

$T_1$  decreases below 2.1 GPa as shown in Fig. 2(a). In general,  $T_1$  in the concentrated Kondo substance corresponds roughly to  $T_K$ . Although  $T_K$  of CeAl<sub>2</sub> increases with pressure as indicated by the result described in  $D$  of Sec. III,  $T_1$  below 2.1 GPa decreases. The antiferromagnetic order with spin-density wave is observed around 3.8 K at ambient pressure. Since SDW has a large effect on the electrical resistance and  $T_K$  is close to  $T_N$ ,  $T_1$  is influenced not only by Kondo effect but also by SDW. The reduction of  $T_1$  may be connected to a decrease of  $T_N$  at high pressure. In other words, it appears that an increase of  $T_1$  or  $T_K$  is masked by a decrease of  $T_N$ . However,  $T_1$  begins to increase above 2.1 GPa. In CeAl<sub>2</sub>, the coexistence of the sinusoidally modulated SDW phase and the simple type-II antiferromagnetic phase below 1.5 GPa has been reported. Above 2.0 GPa, SDW disappears and only the simple type-II antiferromagnetic phase exists.<sup>13,14</sup> The sudden change of  $T_1$  around 2.1 GPa is due to disappearance of SDW, and  $T_1$  is influenced only by Kondo effect above 2.1 GPa.

The Kondo temperature in the substance having two CEF splitting energies was calculated by using periodic Anderson model.<sup>27</sup> In this model,  $T_K$  is modified by CEF splitting and  $\rho_{\text{mag}}(T)$  shows two maxima at  $T_K$  and  $T_K^h$ .  $T_K^h$  is described as follows:

$$T_K^h = (\Delta_1 \Delta_2 T_K)^{1/3}, \quad (1)$$

where  $\Delta_1$  and  $\Delta_2$  are the CEF splitting energies.  $T_K$  at ambient pressure is 6 K and magnitude of the CEF splitting is obtained to be  $\Delta_1=100$  K and  $\Delta_2=180$  K by the inelastic neutron scattering.<sup>16</sup> By using these values and Eq. (1),  $T_K^h$  is estimated to be 48 K, which is comparable with  $T_2=62$  K. Although  $T_K$  increases at high pressure,  $T_2$  decreases below 3 GPa. Thus the fact implies that the value of  $\Delta_1 \Delta_2$  decreases with pressure. Above 3 GPa, the peak corresponding to  $T_1$  merges with that of  $T_2$  and then the  $\rho_{\text{mag}}(T)$  shows a single peak.

The  $\rho_{\text{mag}}(T)$  in Fig. 1(b) shows  $-\log T$  dependence in the two temperature ranges at ambient pressure,  $7 \text{ K} \leq T \leq 20 \text{ K}$  and  $100 \leq T \leq 300 \text{ K}$ , which is a characteristic behavior of Kondo effect. The coefficients of  $-\log T$  are defined to be  $m_1$  and  $m_2$  at low and high temperatures, as shown in Fig. 1(b). From the model presented by Cornut and Coqblin<sup>28</sup> (C-C),  $\rho_{\text{mag}}(T)$  is described by the following equation:

$$\rho_{\text{mag}}(T) = \frac{3m^2 \pi v_0 c}{e^2 \hbar^3 k_F^2} \left[ A + 2J_{sf}^2 D(\epsilon_F) \frac{\lambda^2 - 1}{2j + 1} \ln \frac{k_B T}{W} \right], \quad (2)$$

where  $A$  is the constant,  $c$  the cerium concentration,  $\lambda$  is equal to the total degeneracy of the  $4f$  level,  $W$  is the cutoff energy, and other parameters have literal meaning. The degeneracy factor at low and high temperatures is defined as  $\lambda_L$  and  $\lambda_H$ , respectively.  $m_2/m_1$  is written as

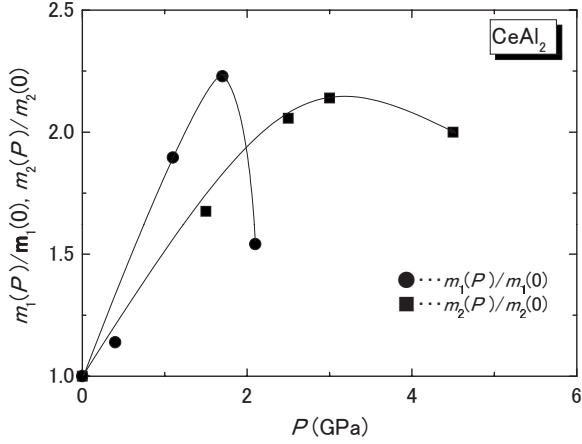
$$\frac{m_2}{m_1} = \frac{\lambda_H^2 - 1}{\lambda_L^2 - 1}. \quad (3)$$

Since the sixfold degenerate levels of Ce<sup>3+</sup> ion are split by CEF effect into doublet and quartet in Ce compounds with cubic structure, the degeneracy factors  $\lambda_L$  for ground state and  $\lambda_H$  for thermally excited state are 2 and 6, respectively, i.e.,  $m_2/m_1=35/3 \sim 12$  from Eq. (3).  $m_2/m_1$  in the present work is obtained  $43/9 \sim 5$  at ambient pressure by the data in Fig. 1(b), which is smaller than the theoretical value. The value of  $m_2/m_1$  in the previous data is almost the same as that in the present result.<sup>29,30</sup> Taking into account that  $m_2/m_1=5$  and the ground state is a doublet,  $\lambda_H$  should be 4.<sup>14</sup> This supports that the CEF splitting in CeAl<sub>2</sub> is three levels, which is in agreement with the results obtained by neutron scattering.<sup>16</sup>

In Eq. (2),  $\lambda$ ,  $W$ , and  $k_f$  are almost constant but a change in  $J_{sf}$  and  $D(\epsilon_F)$  at high pressure are dominant variation of  $m_1$  and  $m_2$ , and these are related to  $J_{sf}$  and  $D(\epsilon_F)$  as follows:

$$m_1 \propto m_2 \propto |J_{sf}^2 D(\epsilon_F)|. \quad (4)$$

The pressure dependence of  $m_1$  and  $m_2$  is shown in Fig. 10. Both  $m_1$  and  $m_2$  at low pressure increase with pressure, but they show a peak around 2 and 3 GPa, respectively. The pressure showing the peak of  $m_1$  corresponds to that where

FIG. 10.  $m_1$  and  $m_2$  as a function of  $P$ .

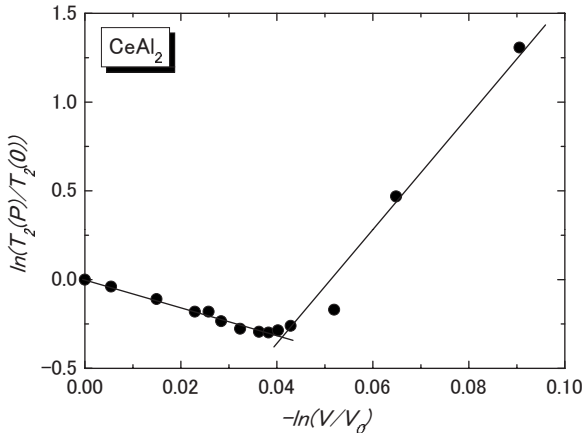
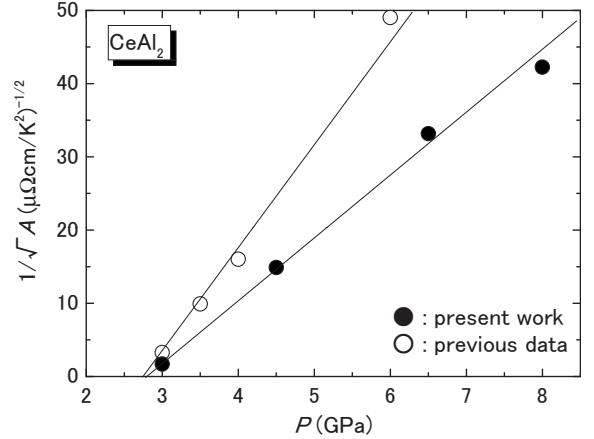
the pressure derivative of  $T_1$  changes from negative to positive as shown in Fig. 2(a). This fact implies that  $T_1$  and  $m_1$  are affected by the disappearance of SDW around 2 GPa. On the other hand, the pressure dependence of  $m_2$  shows a peak around 3 GPa and the pressure corresponds well to that showing QCP. This may be due to the change of the electronic state around QCP, but the details of the origins are not known at present.

### B. Grüneisen parameter of $T_K$ and CEF splitting energy

Next we consider the present results in connection with Grüneisen parameter of  $T_2$ . Since  $T_2$  corresponds to  $T_K^h$ , the Grüneisen parameter for  $T_K^h$ ,  $\Gamma_K^h$  is written as follows:<sup>31</sup>

$$\ln \left[ \frac{T_K^h(P)}{T_K^h(0)} \right] = \ln \left[ \frac{T_2(P)}{T_2(0)} \right] = -\Gamma_K^h \ln \frac{V}{V_0}, \quad (5)$$

where  $V$  and  $V_0$  are the volume at high and ambient pressures. Figure 11 shows the plot of  $\ln[T_2(P)/T_2(0)]$  versus  $-\ln(V/V_0)$ . The two linear relationships are found as shown by the two solid lines and the values of  $\Gamma_K^h$  are  $-8$  below 3 GPa and  $33$  above 3 GPa, respectively.  $\Gamma_K^h$  is also described by using Eq. (1) as follows:

FIG. 11.  $[T_2(P)/T_2(0)]$  as a function  $-\ln(V/V_0)$  in logarithmic scale.FIG. 12.  $1/\sqrt{A}$  as a function of pressure.

$$\Gamma_K^h = -\frac{1}{3} \frac{\partial \ln \Delta_1}{\partial \ln V} - \frac{1}{3} \frac{\partial \ln \Delta_2}{\partial \ln V} - \frac{1}{3} \frac{\partial \ln T_K}{\partial \ln V} = \frac{2}{3} \eta + \frac{1}{3} \Gamma_K, \quad (6)$$

where  $\eta$  is the Grüneisen parameter of crystalline field splitting energy. Here we assume  $\eta = -\partial \ln \Delta_1 / \partial \ln V = -\partial \ln \Delta_2 / \partial \ln V$ . The value of  $\Gamma_K$  in  $\text{CeAl}_2$  is  $34$ .<sup>32</sup> Below 3 GPa,  $\eta$  is estimated to be  $-29$ , which indicates that the magnitude of CEF splitting decreases with increasing pressure. Above 3 GPa,  $\Gamma_K^h$  and  $\Gamma_K$  are almost the same. This result suggests that  $T_2$  above 3 GPa, where a single peak is observed, corresponds to  $T_K$  and the degeneracy relevant to  $T_K$  changes from twofold to four- or sixfold.

### C. Pressure-induced QCP

The coefficient  $A$  of  $T^2$  term shown in Fig. 6 is related to  $T_K$  as  $T_K \propto 1/\sqrt{A}$ .<sup>33</sup> Figure 12 shows  $1/\sqrt{A}$  as a function of pressure. Since  $1/\sqrt{A}$  increases linearly with pressure,  $T_K$  increases at high pressure.  $1/\sqrt{A}$  is found to be extrapolated to 0 around 2.7 GPa, which indicates that a phase boundary exists around  $P_C \sim 2.7$  GPa. The previous result is also plotted in Fig. 12 for comparison.<sup>7</sup> The pressure of  $1/\sqrt{A} = 0$  coincides each other. Since the divergence in the magnitude of  $A$  or  $T_K = 0$  corresponds to a quantum phase transition (QPT), this result indicates that the QPT takes place at 2.7 GPa. This is supported by the results obtained in the following paragraph.

The electronic state near QCP is different from the normal Fermi-liquid behavior showing  $T^2$  dependence in the electrical resistivity. Here we attempted to fit the present data illustrated in Fig. 6 to the following equation:

$$\rho_{\text{mag}} = \rho_0 + A_n T^n, \quad (7)$$

where  $\rho_0$  and  $n$  are the residual resistivity and exponent connecting with electronic state, respectively.

$\rho_{\text{mag}}(T)$  below 2 K is shown in Fig. 13. Fitting was carried out in the temperature range  $50 \text{ mK} \leq T \leq 2 \text{ K}$ . Solid curves in Fig. 13 show the result of fitting, in which these curves reproduce well the experimental data. Figure 14 shows the pressure dependence of  $\rho_0$  and  $n$ .  $\rho_0$  has a peak around 2.7

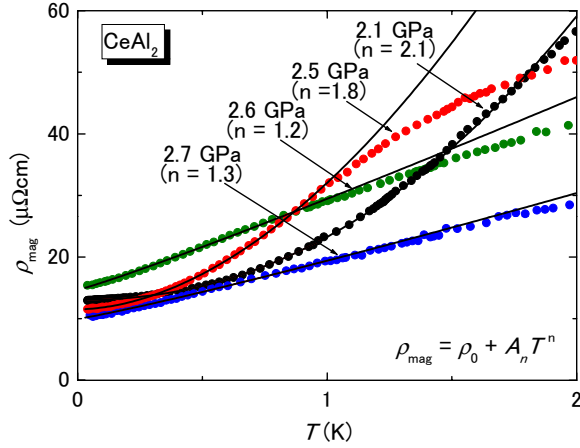


FIG. 13. (Color online)  $\rho_{\text{mag}}(T)$  below 2 K under high pressure. Solid curves show the results of fitting.

GPa.  $n$  is 2 below 2 GPa but around 2.7 GPa, it is 1.3  $\sim$  1.4, which significantly deviates from the normal Fermi-liquid behavior ( $n=2$ ). Judging from these facts, we conclude that CeAl<sub>2</sub> shows the QPT around 2.7 GPa. This indicates that the electronic state around 2.7 GPa is the so-called non-Fermi-liquid state. Furthermore,  $n$  becomes 2 above 3 GPa by applying pressure more as shown in Fig. 3, indicating that Fermi-liquid behavior is recovered.

#### D. $P$ - $T$ phase diagram in CeAl<sub>2</sub>

Finally, we make an attempt to construct the  $P$ - $T$  phase diagram for electronic and magnetic states of CeAl<sub>2</sub> on the basis of the present data. Figure 15 shows the pressure dependence of the electrical resistivity at several temperatures

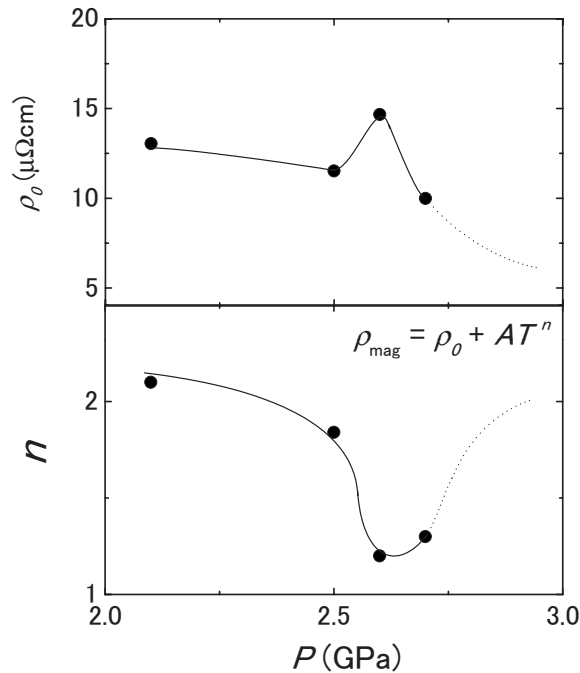


FIG. 14.  $\rho_0$  and  $n$  as a function of pressure. The solid curves are drawn as guide to the eyes.

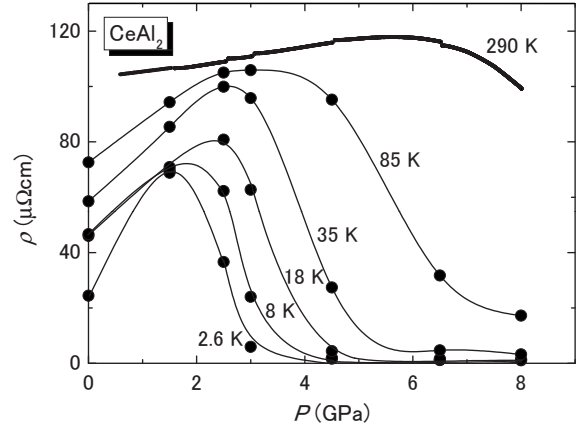


FIG. 15. Pressure dependence of the electrical resistivity at several temperatures. The full circles are obtained by isobaric temperature dependent data at several pressures. The thick line shows isothermal pressure dependence of the electrical resistivity at 290 K. Small discontinuities are observed in  $\rho(P)$  curve due to technical reasons, which does not affect the results in the present work.

in the range  $2.6 \text{ K} \leq T \leq 300 \text{ K}$  which was obtained from the results in Fig. 1(a). For all the isothermal  $\rho$ - $P$  curves,  $\rho$  increases with pressure followed by a maximum at  $P=P_{\text{max}}$  and  $P_{\text{max}}$  increases with temperature. The maximum seems to be clear at low temperature ( $T \leq 35 \text{ K}$ ) but to become broad at high temperature ( $T \geq 85 \text{ K}$ ). It has been reported that there is a crossover induced by pressure from weak Kondo to strong Kondo regime, which is largely different from the first-order  $\gamma$ - $\alpha$  phase transition in Ce metal.<sup>34</sup> In such a sense, the peaks observed in Fig. 16 may correspond to the crossover in the electronic state of CeAl<sub>2</sub>.  $P_{\text{max}}$ - $T$  curve indicates that the Kondo temperature  $T_K$  increases with increasing pressure.  $P_{\text{max}}$  is plotted in Fig. 16.  $P_{\text{max}}$  increases gradually up to 2.6 GPa and rapidly above the pressure where  $T_2$  also shows a similar change and then crosses room temperature at 5.5 GPa.

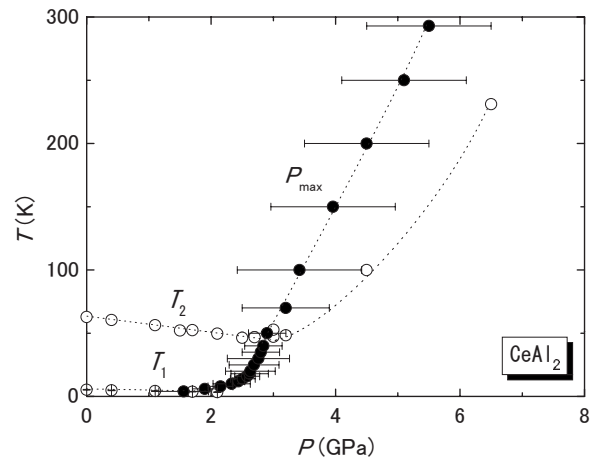


FIG. 16.  $P$ - $T$  phase diagram obtained from the present data. The closed circles show the pressure  $P_{\text{max}}$  showing the maximum of  $\rho$ - $P$  curves as shown in Fig. 15. The open circles show the pressure dependence of  $T_1$  and  $T_2$  for comparison with  $P_{\text{max}}$ .

## V. CONCLUSION

The electrical resistivity of CeAl<sub>2</sub> has been measured in detail at high pressures. The  $\rho(T)$  of CeAl<sub>2</sub> is changed dramatically by applying pressure. According to the analysis of pressure dependence of MR using Okiji-Kawakami model,  $T_K$  is found to increase with pressure.  $T_N$  is decreased by applying pressure and expected to disappear around 2.7 GPa.

Since the coefficient of  $T^2$  term diverges around 2.7 GPa, the magnetic phase boundary exists at 2.7 GPa. Non-Fermi-liquid behavior in  $\rho(T)$  was observed around 2.7 GPa. Above 3 GPa, Fermi-liquid behavior was recovered. The pressure effect on  $T_2$  changes around 3 GPa. Although CeAl<sub>2</sub> shows the QCP around 2.7 GPa, the superconductivity was not observed around 2.7 GPa above 50 mK.

- 
- <sup>1</sup>F. Steglich, J. Magn. Magn. Mater. **100**, 186 (1991).  
<sup>2</sup>S. Doniach, Physica B & C **91**, 231 (1977).  
<sup>3</sup>For the summary of recent progress in this research field, see J. Phys. Soc. Jpn. **76** (2007) Suppl., edited by T. Kagayama, M. Ohashi, and Y. Uwatoko.  
<sup>4</sup>A. T. Holmes, D. Jaccard, and K. Miyake, Phys. Rev. B **69**, 024508 (2004).  
<sup>5</sup>G. Knebel, D. Braithwaite, P. C. Canfield, G. Lapertot, and J. Flouquet, Phys. Rev. B **65**, 024425 (2001).  
<sup>6</sup>G. R. Stewart, Rev. Mod. Phys. **73**, 797 (2001).  
<sup>7</sup>G. Oomi, M. Ohashi, Y. Uwatoko, I. Satoh, and T. Komatsubara, Physica B **359-361**, 65 (2005).  
<sup>8</sup>H. Miyagawa, M. Ohashi, G. Oomi, I. Satoh, and T. Komatsubara, Physica B **378-380**, 771 (2006).  
<sup>9</sup>H. Miyagawa, Y. Fuchizaki, M. Ohashi, G. Oomi, I. Satoh, and T. Komatsubara, High Press. Res. **26**, 503 (2006).  
<sup>10</sup>M. Ohashi, G. Oomi, S. Koiwai, M. Hedo, and Y. Uwatoko, Phys. Rev. B **68**, 144428 (2003).  
<sup>11</sup>F. Steglich, C. D. Bredl, M. Loewenhaupt, and K. D. Schotte, J. Phys. Colloq. **C5**, 301 (1979).  
<sup>12</sup>C. D. Bredl, F. Steglich, and K. D. Schotte, Z. Phys. B **29**, 327 (1978).  
<sup>13</sup>B. Barbara, M. F. Rossignol, J. X. Boucherle, J. Schweizer, and J. L. Buevoz, J. Appl. Phys. **50**, 2300 (1979).  
<sup>14</sup>B. Barbara, M. F. Rossignol, J. X. Boucherle, and C. Vettier, Phys. Rev. Lett. **45**, 938 (1980).  
<sup>15</sup>S. Tomisawa, S. Wada, M. Ohashi, and G. Oomi, J. Phys.: Condens. Matter **18**, 10413 (2006).  
<sup>16</sup>M. Loewenhaupt, B. D. Rainford, and F. Steglich, Phys. Rev. Lett. **42**, 1709 (1979).  
<sup>17</sup>P. Thalmeier and P. Fulde, Phys. Rev. Lett. **49**, 1588 (1982).  
<sup>18</sup>T. Sawamura, T. Andoh, G. Oomi, and Y. Ōnuki, Rev. High Pressure Sci. Technol. **7**, 614 (1998).  
<sup>19</sup>C. Probst and J. Wittig, J. Magn. Magn. Mater. **9**, 62 (1978).  
<sup>20</sup>F. Honda, S. Kaji, I. Minamitake, M. Ohashi, G. Oomi, T. Eto, and T. Kagayama, J. Phys.: Condens. Matter **14**, 11501 (2002).  
<sup>21</sup>N. Mōri, H. Takahashi, and N. Takeshita, High Press. Res. **24**, 225 (2004).  
<sup>22</sup>D. Jaccard and A. T. Holmes, Physica B **359-361**, 333 (2005).  
<sup>23</sup>R. Schefzyk, W. Lieke, and F. Steglich, Solid State Commun. **54**, 525 (1985).  
<sup>24</sup>N. D. Mathur, F. M. Grosche, S. R. Julian, I. R. Walker, D. M. Freye, R. K. Haselwimmer, and G. G. Lonzarich, Nature (London) **394**, 39 (1998).  
<sup>25</sup>A. Sumiyama, Y. Oda, H. Nagano, Y. Ōnuki, K. Shibusani, and T. Komatsubara, J. Phys. Soc. Jpn. **55**, 1294 (1986).  
<sup>26</sup>N. Kawakami and A. Okiji, J. Phys. Soc. Jpn. **55**, 2114 (1986).  
<sup>27</sup>K. Hanzawa, K. Yamada, and K. Yoshida, J. Magn. Magn. Mater. **47-48**, 357 (1985).  
<sup>28</sup>B. Cornut and B. Coqblin, Phys. Rev. B **5**, 4541 (1972).  
<sup>29</sup>M. Nicolas-Francillon, A. Percheron, J. C. Achard, O. Gorochoy, B. Cornut, D. Jerome, and B. Coqblin, Solid State Commun. **11**, 845 (1972).  
<sup>30</sup>Y. Ōnuki, Y. Furukawa, and T. Komatsubara, J. Phys. Soc. Jpn. **53**, 2734 (1984).  
<sup>31</sup>T. Kagayama, G. Oomi, K. Iki, N. Mori, Y. Ōnuki, and T. Komatsubara, J. Alloys Compd. **213-214**, 387 (1994).  
<sup>32</sup>G. Oomi, T. Kagayama, Y. Ōnuki, H. Mitamura, and T. Goto, in *Proceedings of the Moscow International Symposium on Magnetism (MISM'99)*, Moscow, 1999 (Faculty of Physics, M. V. Lomonosov Moscow State University, Moscow), p. 351.  
<sup>33</sup>A. Yoshimori and H. Kasai, J. Magn. Magn. Mater. **31-34**, 475 (1983).  
<sup>34</sup>B. Barbara, J. Beille, B. Cheiato, J. M. Laurant, M. F. Rossignol, A. Waintal, and S. Zemirli, Phys. Lett. **113A**, 381 (1986).