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Effect of Pressure and Magnetic Field on the Electrical Resistivity of Cerium Kondo Compounds

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Electrical resistivity of three Ce Kondo compounds having different characteristic temperatures T_K, $CeAl_3$, $CeInCu_2$ and CeNi has been measured at high pressure and high magnetic field. It is found that the electrical resistivity of these compounds shows T^2-dependence at low temperature and the coefficient of the term decreases with increasing pressure. The magnetoresistance of $CeInCu_2$ changes the sign from negative to positive as pressure increases, which indicates a pressure-induced crossover from incoherent to coherent Kondo regime.

KEYWORDS: high pressure, electrical resistance, heavy fermion

§1. Introduction

There have been a lot of investigations about the electronic and magnetic properties of concentrated Kondo (CK) compounds containing Ce or U because these compounds give an important information for studying the role of strong electron correlations in the metallic systems.[1,2] The CK compounds show several anomalous properties, such as a huge value of linear term coefficient γ in the electronic specific heat, a large T^2-term in the electrical resistivity $\rho(T)$ at low temperatures, a $\log T$-term in the $\rho(T)$ at high temperatures and so forth.

It is well known that the electronic states of CK compounds are strongly dependent on the change in pressure or volume and magnetic field since these are electronically unstable.[3] The electronic states of the systems are characterized by the so-called Kondo temperature T_K. The fact mentioned above indicates the large change in the magnitude of T_K by an application of pressure or a change in magnetic field. Usually the heavy fermion (HF) system having a low T_K of the order of several degree Kelvin has a large Grüneisen parameter Γ of the order of 100.[4] T_K of HF compounds increases rapidly with increasing pressure to show a crossover into a new electronic state,[5] which is called the intermediate valence (IV) state having relatively small Γ.[6] Recently we have reported such pressure-induced crossover for several Ce and U compounds[7,8] and found a systematics for the factor of $JN(0)$ and the values of Γ,[9] where J is the s-f exchange interaction and $N(0)$ the density of states at the Fermi level. In order to get a deep insight in the study of electronic structure of CK compounds, it is worthwhile to extend our investigation at high pressure to other CK compounds having different T_K values.

In the present work we made an attempt to measure the electrical resistivity of three CK compounds having different T_K, $CeAl_3$, $CeInCu_2$ and CeNi under high pressure and high magnetic field. The volume (or lattice constants) of the three compounds was also measured at high pressure. The results are discussed by taking into account the volume (or pressure) dependent Grüneisen parameters.[10] We point out that the electronic crossover in CK compounds at high pressure is accompanied by a change in the value of Γ at high pressure.

§2. Experimentals

The specimens in the present work were single crystals of $CeInCu_2$ and CeNi and polycrystal of $CeAl_3$. The details of the preparation were described elsewhere.[11-13] The pressure dependence of lattice constants was measured by means of powder X-ray diffraction technique. Hydrostatic pressure was generated by using a piston-cylinder device. The details of the high pressure apparatus was reported in our previous publication.[14]

§3. Results

The temperature dependence of the electrical resistivity $\rho(T)$ of $CeAl_3$ at various pressures up to 8 GPa and $\rho(T)$ of $LaAl_3$ at ambient pressure are shown in Fig. 1. The $\rho(T)$ of $LaAl_3$ is similar to the ordinary non-magnetic metal; it varies linearly with temperature above 100 K without any anomaly. Whereas, at ambient pressure, ρ of $CeAl_3$ increases logarithmically with decreasing temperature until it reaches a maximum at 35 K and has a shoulder near 6 K. This behavior is due to the Kondo scattering on a thermally populated level splitted by crystalline electric field.[15] With increasing pressure, the peak and the shoulder are merged into one peak, which is shifted towards higher temperatures. The $\rho(T)$ at 8 GPa becomes similar to that of $LaAl_3$. This result is interpreted as a pressure-induced crossover in the electronic state of $CeAl_3$ from low- T_K HF state to high- T_K intermediate valence (IV) state associated with an increase in the hybridization between conduction and $4f$ electrons.

In order to get the temperature-dependent $4f$ magnetic contribution $\rho_{mag}(T)$, the $\rho(T)$ of $LaAl_3$ is assumed to be pressure independent phonon part of $CeAl_3$ and subtracted from the $\rho(T)$ data of $CeAl_3$ at various pressures. Figure 2 illustrates the $\rho_{mag}(T)$ as a function of $\log T$.

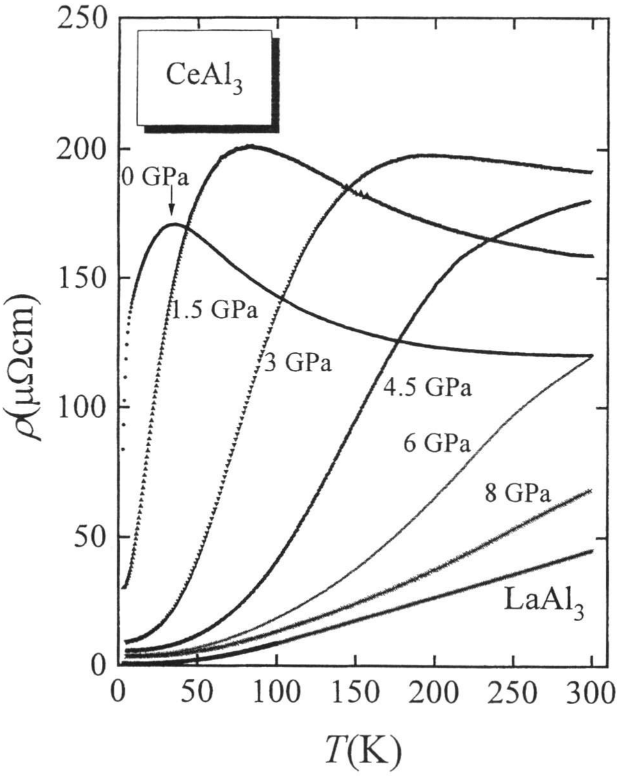

Fig. 1. The electrical resistivity $\rho(T)$ of $CeAl_3$ under high pressure as a function of temperature. $\rho(T)$ of $LaAl_3$ at ambient pressure is also shown for comparison.

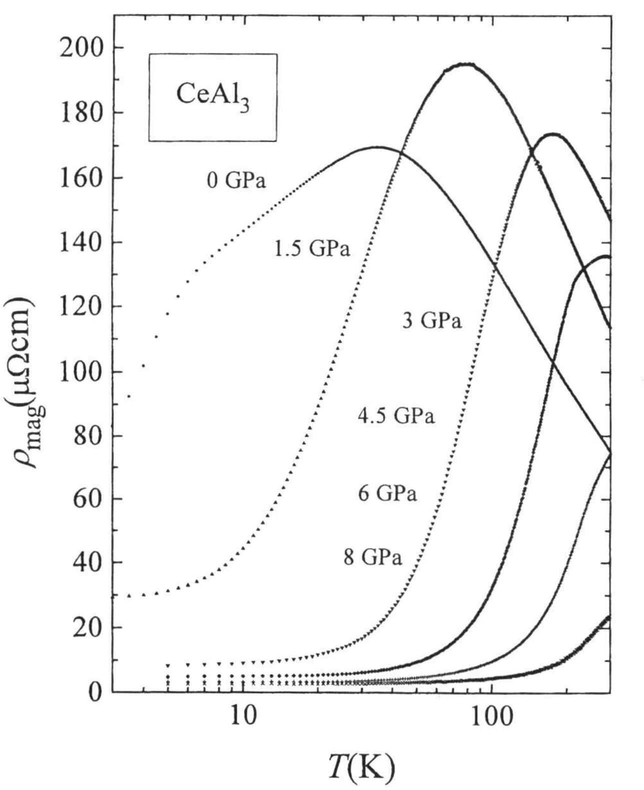

Fig. 2. The magnetic part of electrical resistivity, ρ_{mag}, as a function of $\log T$ at various pressures.

The maximum temperature T_{max} in the $\rho_{mag}(T)$ is shown in Fig. 3. T_{max} is found to increase with increasing pres-

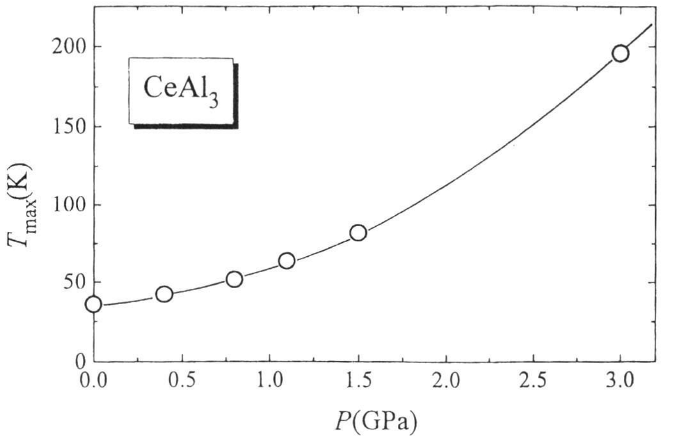

Fig. 3. The pressure dependence of the temperature T_{max} at which ρ_{mag} has a maximum.

sure. Since T_{max} is roughly proportional to the T_K,[16] the pressure dependence of T_K may be inferred from the result in Fig. 3. On the other hand the logarithmic dependence of the ρ_{mag} on temperature is observed in the wide range above the T_{max}. The negative slope becomes steeper at higher pressure reflecting the strong Kondo scattering with large enhancement of T_K at high pressures.

In order to examine the T^2-dependence in the $\rho_{mag}(T)$ at low temperature, ρ_{mag} is plotted as a function of T^2 for $CeAl_3$ in Fig. 4 up to 17 K for $P \leq 1.5$ GPa and to 80 K for $P \geq 3$ GPa. Above 0.8 GPa the T^2-dependence is clearly observed in the temperature range of the present work as shown by straight line. As pressure increases,

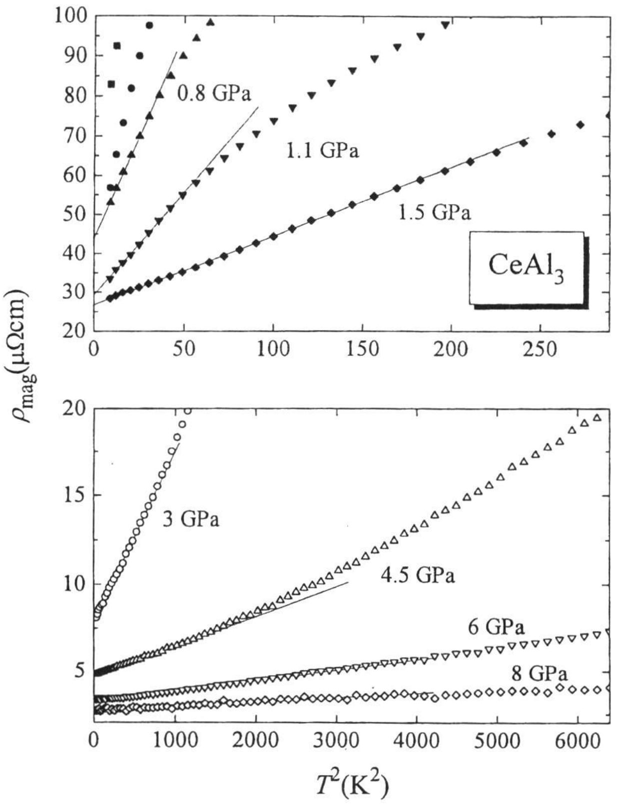

Fig. 4. ρ_{mag} of $CeAl_3$ as a function of T^2.

the slope decreases and the temperature range showing T^2-dependence becomes wider. The coefficient A of T^2-term is shown in Fig. 5 as a function of pressure. The value of A is reduced to 3 orders of magnitude smaller than that at ambient pressure, which was reported previously to be $35\,\mu\Omega cm/K^2$.[17] The rapid decrease in the magnitude of A is explained by the enhancement of T_K by applying pressure, which is consistent with the increase in T_{max}, because the coefficient A is inversely proportional to T_K^2.[16]

The systematics mentioned above are also found in other CK compounds. Figure 6 shows the temperature dependent ρ of the cubic HF compound $CeInCu_2$.[7] The $\rho(T)$ of $CeInCu_2$ has a well known peak around 26 K which moves up with pressure.

Figure 7 shows pressure dependence of T_{max} for $CeInCu_2$ together with the data of the measurement at higher temperature up to 1,000 K.[10] It appears that T_{max} increases exponentially with pressure.

The T^2-dependence of the ρ_{mag} subtracted by the residual resistivity ρ_0 at low temperature, $\rho_{mag} - \rho_0$, is shown in Fig. 8 for $CeInCu_2$. It is noted that the coherency in Kondo lattice of $CeInCu_2$ is partially broken, owing to the disorder between Ce and In-site in Heusler-type crystal structure, which gives rise to a large value of ρ_0. Therefore the value of the coefficient A of T^2-term is reduced to 2 orders smaller than that of $CeAl_3$ in spite of comparable value of the specific heat coefficient γ.[17] Figure 9 shows the pressure dependence of the value of A for $CeInCu_2$. The large decrease is also observed in

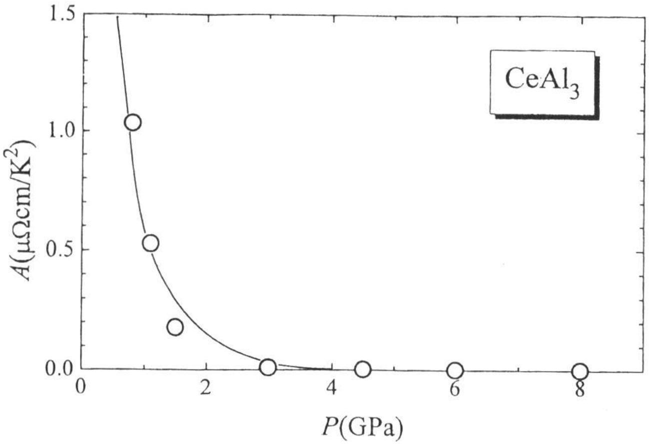

Fig. 5. The coefficient A of T^2-term as a function of pressure.

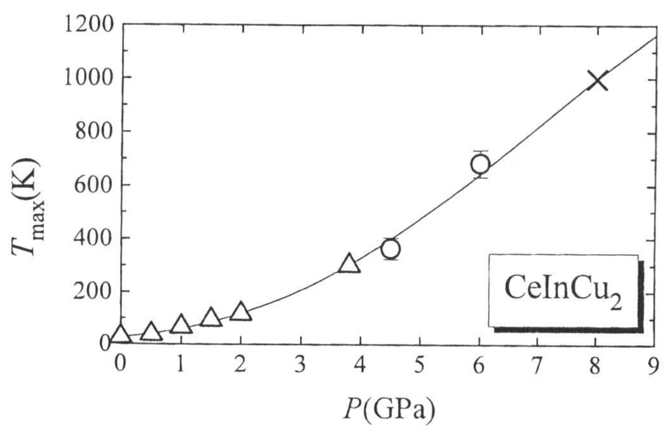

Fig. 7. The pressure dependence of the temperature T_{max} at which ρ_{mag} has a maximum.

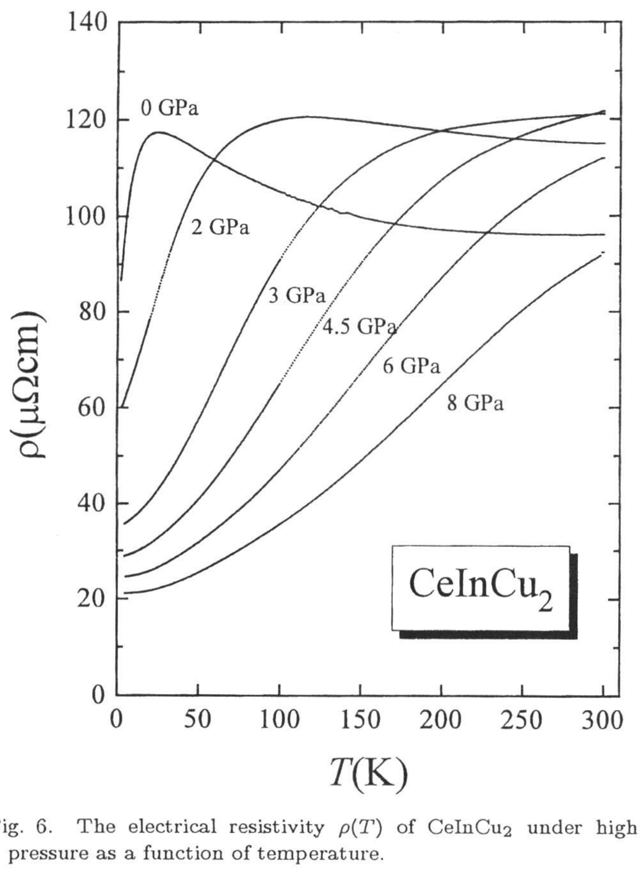

Fig. 6. The electrical resistivity $\rho(T)$ of $CeInCu_2$ under high pressure as a function of temperature.

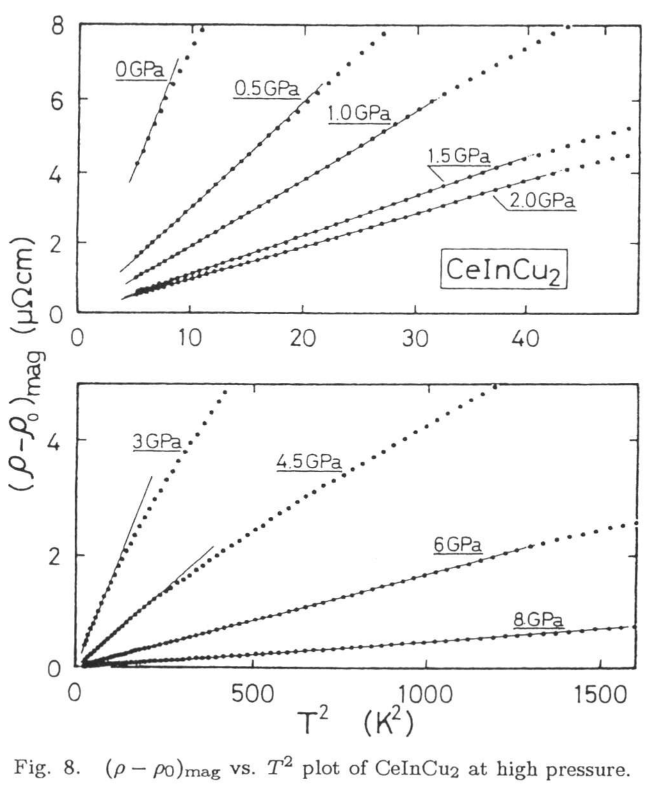

Fig. 8. $(\rho - \rho_0)_{mag}$ vs. T^2 plot of $CeInCu_2$ at high pressure.

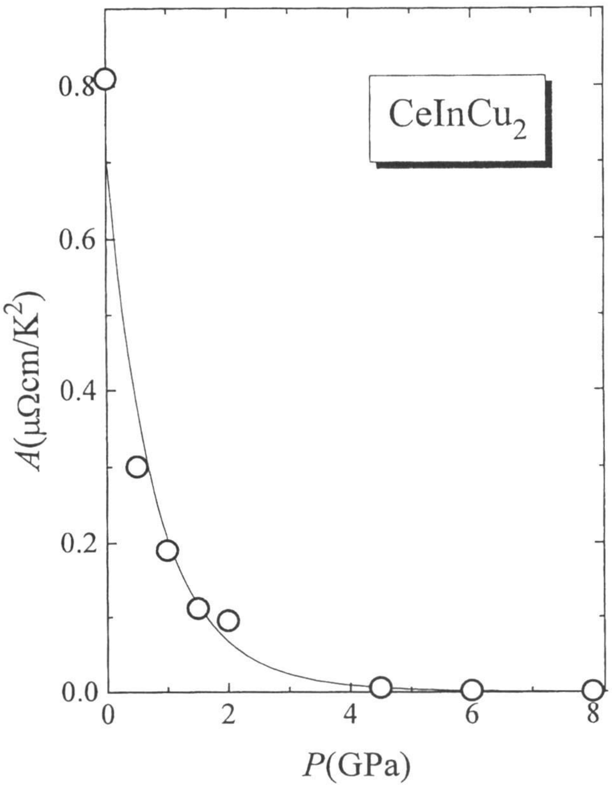

Fig. 9. Pressure dependence of A of $CeInCu_2$.

the pressure dependence of the magnitude of A and consistent with the result of T_{max} (or T_K) which was shown in Fig. 7.

CeNi is a typical IV compound having a large T_K of about 100 K.[18] Consequently T^2-term in the electrical resistance is expected to be observed in relatively wide temperature range compared with the low T_K HF compounds such as $CeAl_3$ or $CeInCu_2$. Figure 10 shows the magnetic part of electrical resistivity ρ_{mag} of CeNi subtracted by the value of ρ_0, as a function of T^2. T^2-dependence in the $\rho(T)$ curve of CeNi is seen in the usual experimental temperature range: $\rho(T)$ at ambient pressure shows T^2-dependence up to 30 K. The T^2-term coefficient A is shown in Fig. 11 as a function of pressure.[19] The behavior against pressure is qualitatively the same as that in low-T_K HF compounds mentioned above but the pressure response is less prominent than that of $CeAl_3$.

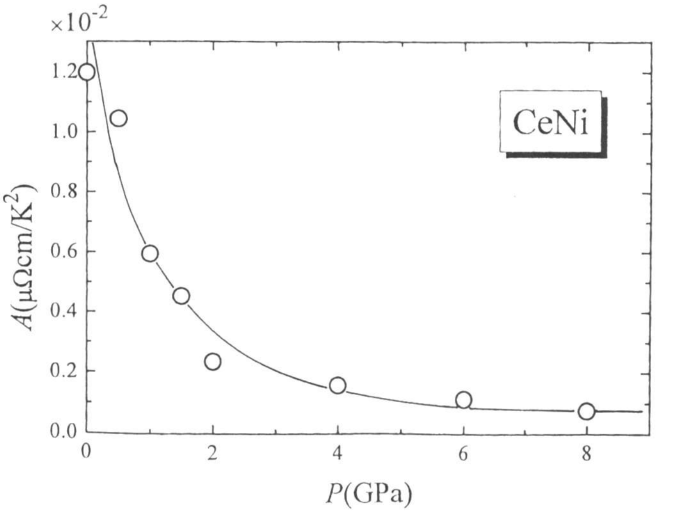

Fig. 11. Pressure dependence of A of CeNi.

Next we show the field dependence of the magnetoresistance (MR) of $CeInCu_2$, $\Delta\rho/\rho = [\rho(H)-\rho(0)]/\rho(0)$, at 4.2 K, which is plotted at various pressures up to 2 GPa in Fig. 12.[20] The value of $\Delta\rho/\rho$ below 1 GPa is negative, but the sign changes between 0.5 and 1.0 GPa. Above 1 GPa, $\Delta\rho/\rho$ increases with increasing H, i.e., the positive MR is observed, but the effect of pressure on $\Delta\rho/\rho$ above 1 GPa is relatively small. Similar pressure behavior of MR at high pressure was reported previously for UBe_{13}.[21]

For several HF compounds such as $CeCu_6$,[22] $CeAl_3$[23] or $CeCu_2Si_2$,[24] the sign of MR changes from negative to positive with decreasing temperature below 1 K. The positive MR is explained to imply that the system enters to so-called coherent state. On the contrary, MR of $CeInCu_2$ remains negative and continues to increase in magnitude down to 0.4 K.[20] The coherent state may be broken by the existence of 4f-site disorder in $CeInCu_2$. A possible interpretation of the change in the sign of MR at high pressure is a crossover from a localized f-electron state to an itinerant or coherent one, i.e., the effect of

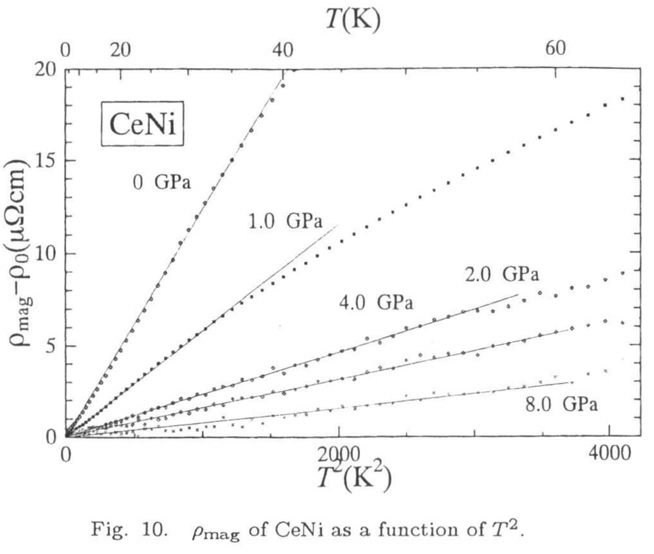

Fig. 10. ρ_{mag} of CeNi as a function of T^2.

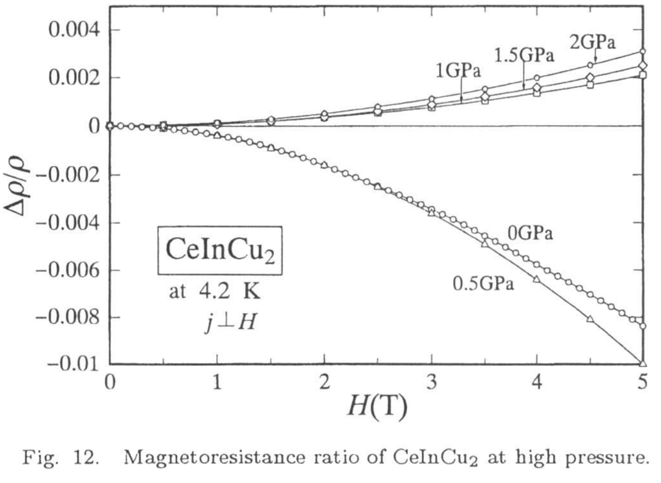

Fig. 12. Magnetoresistance ratio of $CeInCu_2$ at high pressure.

disorder may be less significant at high pressure. Therefore the present result implies that the coherence, which lies below at least 0.4 K at ambient pressure, is induced at 4.2 K by applying pressure of 1 GPa.

Almost the same result is obtained for $CeAl_3$, which is indicated in Fig. 13. The MR at 1.5 GPa is almost zero and then it is expected to change to positive at higher pressure than 1.5 GPa.

The temperature dependence of MR was calculated for Ce-based heavy fermion system by Kawakami and Okiji[25] as a function of T/T_K on the basis of the periodic Anderson model. According to their result, the positive MR appears at low temperature $T/T_K \leq 0.2$ due to the gap-structure of the Kondo resonance a little above the Fermi level. Their calculation succeeded in explaining qualitatively many characteristic properties for the MR of $CeAl_3$ and $CeCu_6$. At ambient pressure the coherent state is induced only by lowering temperature. On the other hand, it is also induced by high pressure because T_K is enhanced largely by an application of pressure to give the small value of T/T_K. Since the T_K of $CeInCu_2$ at 1 GPa, as mentioned above, is several times as large as that at ambient pressure, the reduction of T/T_K at $T = 4.2$ K by applying pressure of 1 GPa may be sufficient to induce a positive MR or coherent state.

As we mentioned above, the pressure sensitivity of the T_K has a wide variety depending on materials. Furthermore, these three compounds are crystallized in different structures and elastic properties are different. Investigation of the crystal structure at high pressure, thus, is important to compare the pressure dependence of the T_K for each material.

Figure 14 shows the result of X-ray diffraction experiment at high pressure and at room temperature for $CeAl_3$. The fractional change of lattice constants a and c along with the unit cell volume $V = \sqrt{3}a^2c/2$ is shown as a function of pressure.[26] The diffraction pattern at high pressure reveals that the Ni_3Sn-type hexagonal structure is stable at room temperature at least up to 17 GPa. A discontinuous change in the lattice constants, like γ-α transition in Ce metal, is not observed within an experimental error. Thus the crossover from CK to IV state in

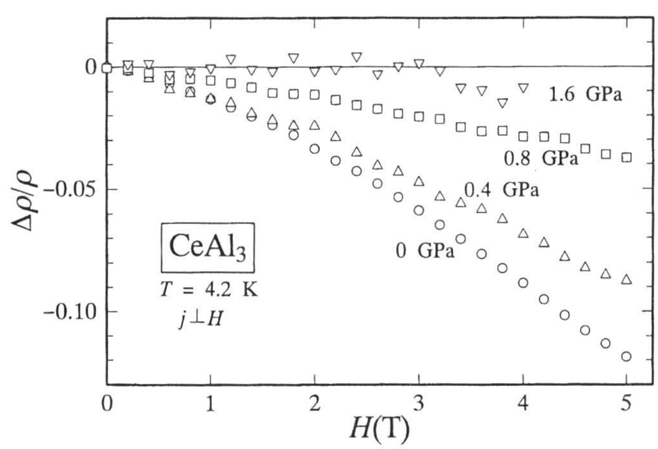

Fig. 13. Magnetoresistance ratio of $CeAl_3$ as a function of pressure.

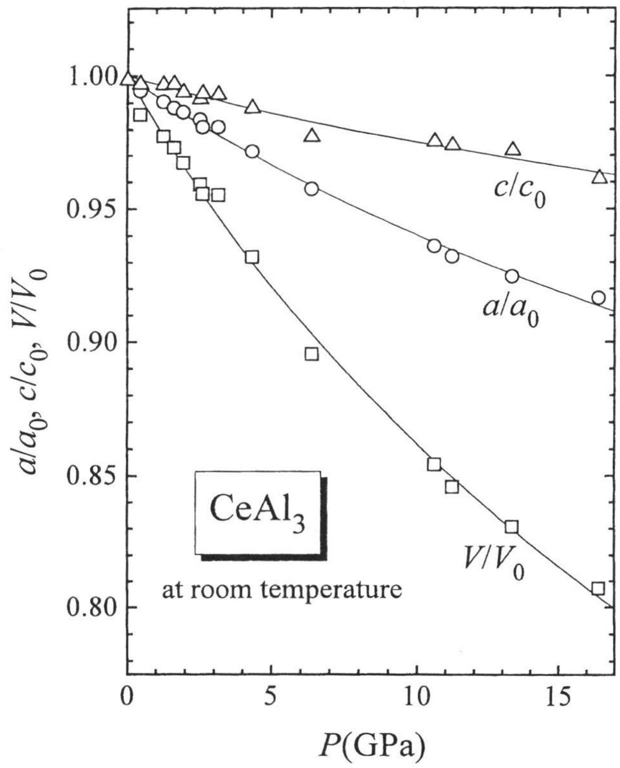

Fig. 14. Volume and lattice constants of $CeAl_3$ as a function of pressure.

$CeAl_3$ observed in the electrical resistivity occurs gradually without volume anomaly. To estimate the bulk modulus, we attempted a least-squares fit of the data of V to the following first-order Murnaghan's equation of state:

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

where B_0 is the bulk modulus at ambient pressure, $-\partial P/\partial \ln V|_{P=0}$, and B_0' is its pressure derivative, $\partial B/\partial P|_{P=0} - 1$. We obtained $V_0 = 171.2\,\text{Å}^3$, $B_0 = 53.6$ GPa and $B_0' = 2.97$. The solid line for V/V_0 in Fig. 14 shows the result of fitting. The values of B_0 for $CeInCu_2$ and CeNi were obtained to be 97 GPa and 25 GPa at room temperature, respectively.[19,27]

§4. Discussion

In order to discuss the volume dependence of the T_K in detail, we analyzed the present result mentioned in §3 as follows.

The Grüneisen parameter of the T_K is defined as,

$$\Gamma \equiv -\frac{\partial \ln T_K}{\partial \ln V} . \qquad (2)$$

If we assume that the T_{max} is proportional to the T_K and the coefficient A of T^2-term is inversely proportional to T_K^2, i.e., $T_{max} \propto T_K$ and $A \propto 1/T_K^2$, we have,

$$\Gamma = -\frac{\partial \ln T_{max}}{\partial \ln V} = \frac{1}{2}\frac{\partial \ln A}{\partial \ln V} . \qquad (3)$$

Equation (3) can be rewritten as follows,

$$\ln\left[\frac{T_{\max}(P)}{T_{\max}(0)}\right] = -\frac{1}{2}\ln\left[\frac{A(P)}{A(0)}\right] = -\Gamma\ln\left(\frac{V}{V_0}\right) \quad . \quad (4)$$

In Fig. 15 we plot the logarithm of the relative change of the A against that of volume for $CeAl_3$, in which the pressure is included as an implicit variable. The relative change of volume, $-\ln(V/V_0)$ is evaluated by using eq. (1). The ordinate in this plot indicates the relative change in the T_K and hence the slope corresponds to the Γ (see eq. (4)). From tangent at the origin of this plot, the value of Γ is estimated to be 97 at ambient pressure. As is seen in Fig. 15, the Γ is dependent on the volume change. The slope (Γ) becomes smaller as the change in volume is larger (higher pressure). Since the system shows a crossover from low-T_K to high-T_K state induced by pressure, it is expected that the Γ decreases at high pressure. This result agrees with the fact that the values of Γ for IV materials having large T_K are smaller than those of low-T_K HF materials.

Fig. 16 shows the same plot as Fig. 15 for the $A(P)$ data of CeNi up to 8 GPa as was shown in Fig. 11. The Γ value is estimated to be about 11 at ambient pressure. It is noted that the A of CeNi behaves against V in the same way as $CeAl_3$; the slope in the $-\ln[A(P)/A(0)]/2$ vs $-\ln(V/V_0)$ plot becomes smaller at high pressure. The decrease in the Γ occurs even in the intermediate valence compound indicating the more itinerant state at high pressure.

Next we show the case in which the same analysis is valid to the data of T_{max}. Fig. 17 illustrates the relative change of the T_{max} against V in logarithmic scale for $CeInCu_2$. Though the pressure range in the experiment of the electric resistance (up to 8 GPa) is the same as $CeAl_3$ and CeNi, the range in the $-\ln(V/V_0)$ are largely different because of the difference in the bulk moduli. If we adjust the origin of these plot, i.e., the values of T_K and volume at ambient pressure, it is found that all observed points for the three CK materials fall on a "universal" curve.[28] This fact suggests that the present treatment for the values of A and T_{max} is a good tool to analyze quantitatively the pressure dependent $\rho(T)$ curve and further should be applied to the other HF and IV compounds. The extension of the present method to other Kondo materials is now in progress and will be reported in the near future.

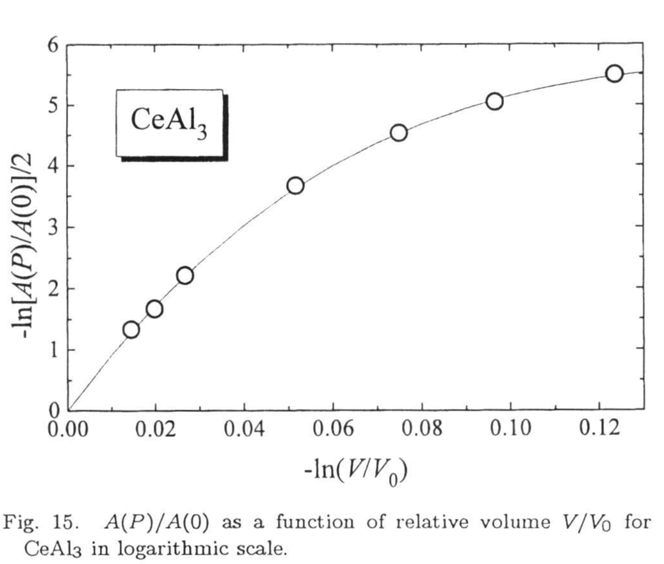

Fig. 15. $A(P)/A(0)$ as a function of relative volume V/V_0 for $CeAl_3$ in logarithmic scale.

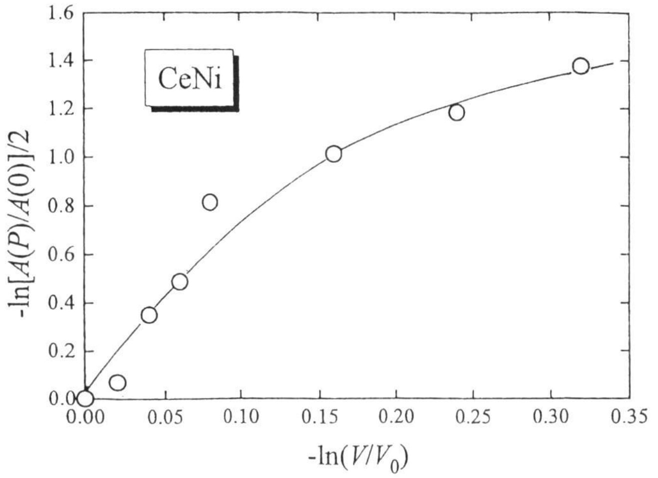

Fig. 16. Logarithmic plot of $A(P)/A(0)$ vs. V/V_0 for CeNi.

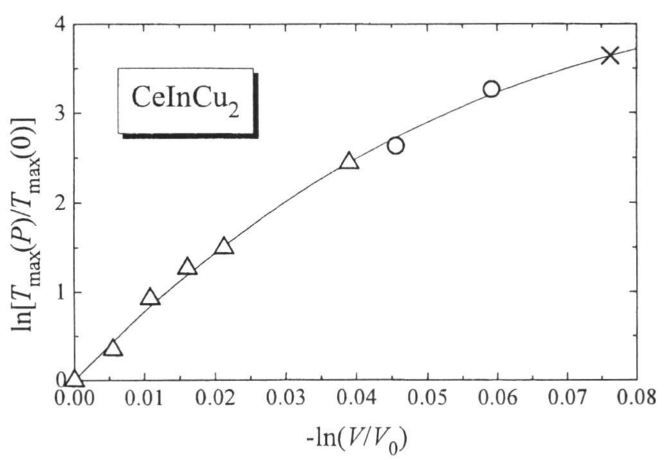

Fig. 17. $T_{max}(P)/T_{max}(0)$ as a function of relative volume V/V_0 for $CeInCu_2$ in logarithmic scale.

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1) G.R. Stewart: Rev. Mod. Phys. **56** (1984) 755.
2) N.B. Brandt and V.V. Moshchalkov: Adv. in Phys. **33** (1984) 373.
3) J.D. Thompson and J.M. Lawrence: *Handbook on the Physics and Chemistry of Rare Earths* ed. K.A. Gschneidner, Jr. *et al.* (Elsevior, Amsterdam, 1994) Vol.19, p.383.
4) A. de Visser, J.J.M. Franse, A. Lacerda, P. Haen and J. Flouquet: Physica B **163** (1990) 49.

5) J.M. Mignot and J. Wittig: *Valence Instabilities*, ed. P. Wachter and H. Boppart (North-Holland, 1982) p.203.
6) J. Flouquet, J.C. Lasjaunias, J. Peyrard and M. Ribault: J. Appl. Phys. **53** (1982) 2127.
7) T. Kagayama, G. Oomi, H. Takahashi, N. Mōri, Y. Ōnuki and T. Komatsubara: Phys. Rev. B **44** (1991) 7690.
8) T. Kagayama, G. Oomi, K. Iki, N. Mōri, Y. Ōnuki and T. Komatsubara: J. Alloys and Compounds **213&214** (1994) 387.
9) T. Kagayama and G. Oomi: *Transport and Thermal Properties of f-electron Systems*, ed. G. Oomi, H. Fujii and T. Fujita (Plenum, NewYork, 1993) p.155.
10) T. Kagayama, G. Oomi, E. Ito, Y. Ōnuki and T. Komatsubara: J. Phys. Soc. Jpn. **63** (1994) 3927.
11) Y. Ōnuki, Y. Shimizu and T. Komatsubara: J. Phys. Soc. Jpn. **54** (1985) 304.
12) Y. Uwatoko, G. Oomi and J. Sakurai: Physica B **186-188** (1993) 560.
13) T. Kagayama, K. Munakata and G. Oomi: J. Alloys and Compounds **192** (1993) 239.
14) G. Oomi, T. Kagayama and Y. Uwatoko: Jpn. J. Appl. Phys. **32** (1993) 349.
15) B. Cornut and B. Coqblin: Phys. Rev. B **5** (1972) 4541.
16) A. Yoshimori and H. Kasai: J. Magn. Magn. Mater. **31-34** (1983) 475.
17) K. Andres, J.E. Graebner and H.R. Ott: Phys. Rev. Lett. **35** (1975) 1779.
18) G. Creuzet and D. Gignoux: Phys. Rev. B **33** (1986) 515.
19) G. Oomi and N. Mōri: J. Alloys and Compounds **207&208** (1994) 275.
20) T. Kagayama, G. Oomi, R. Yagi, Y. Iye, Y. Ōnuki and T. Komatsubara: J. Phys. Soc. Jpn. **61** (1992) 2632.
21) M.C. Aronson, J.D. Thompson, J.L. Smith, Z. Fisk and M.W. McElfresh: Phys. Rev. Lett. **63** (1989) 2311.
22) A. Sumiyama, Y. Oda, H. Nagano, Y. Ōnuki, K. Shibutani and T. Komatsubara: J. Phys. Soc. Jpn. **55** (1986) 1294.
23) G. Remenyi, A. Briggs, J. Flouquet, O. Laborde and F. Lapierre: J. Magn. Magn. Mater. **31-34** (1983) 417.
24) Y. Ōnuki, T. Hirai, T. Komatsubara, S. Takayanagi, A. Sumiyama, A. Furukawa, Y. Oda and H. Nagano: J. Magn. Magn. Mater. **52** (1985) 338.
25) N. Kawakami and A. Okiji: J. Phys. Soc. Jpn. **55** (1986) 2114.
26) T. Kagayama and G. Oomi: J. Magn. Magn. Mater. **140-144** (1995) 1227.
27) T. Kagayama and G. Oomi: Jpn. J. Appl. Phys. **32** (1993) 318.
28) T. Kagayama: Dr. Thesis (Kumamoto Univ. 1995) (in Japanese).