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Pressure-induced valence instability of the heavy-fermion compound $CeInCu_2$

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The temperature dependence of the electrical resistivity $\rho(T)$ of the heavy-fermion compound $CeInCu_2$ has been measured under hydrostatic pressures up to 8 GPa. It is found that the high pressure induces valence instability in $CeInCu_2$ associated with the large enhancement of the Kondo temperature T_K. The resistivity-maximum temperature T_{max} increases with increasing pressure and the coefficient A of the T^2 term in $\rho(T)$ at low temperatures decreases largely with increasing pressure. These results are discussed briefly on the basis of the Yoshimori-Kasai theory.

It is well known that the physical properties of the heavy-fermion (HF) system are strongly dependent on the volume or pressure.[1,2] The most important energy scale for the system is the so-called Kondo temperature, T_K, which may be roughly estimated from the resistivity-maximum temperature, T_{max}. The application of pressure enhances the T_K, mainly due to an increase of the hybridization between f and the conduction band.[3] The second energy scale T_{coh}, which characterizes the low-temperature coherent phase, is also expected to increase with increasing pressure.[4]

There have been several works about the electrical resistivity $\rho(T)$ of HF compounds under high pressure.[5–8] $\rho(T)$ of the HF compounds such as $CeCu_6$ or $CeAl_3$ has a maximum at T_{max}, which is the order of 10 K and shows a log T dependence above T_{max}. Below T_{max}, $\rho(T)$ decreases with decreasing temperature and shows a T^2 dependence at very low temperature below $\sim$1 K, which is a characteristic behavior of the Fermi liquid.[9] It has been reported that the T_{max} and the coefficient A of the T^2 term of $CeCu_2Si_2$ (Ref. 5) and $CeCu_6$ (Refs. 7 and 8) are strongly affected by the application of pressure, implying an increase of T_K, and that the change of the overall behavior of $\rho(T)$ curve at high pressure indicates a pressure-induced crossover from the concentrated Kondo state (CKS) to the intermediate-valence state (IVS).

$CeInCu_2$ is found to be a cubic Heusler-type HF compound having the large value of the specific-heat coefficient $\gamma \sim 1.2$ J / mol K^2 at 1 K.[10,11] Since the various physical properties of noncubic HF compounds such as $CeCu_6$ are largely anisotropic, the quantitative discussion of the results is often very complicated.[12] Physical properties of $CeInCu_2$ show isotropic behaviors due to cubic structure as was observed in the thermal expansion.[13] The $\rho(T)$ of $CeInCu_2$ under pressure was reported by Najib *et al.*[14] up to 1.55 GPa. According to their result, T_{max} increased at high pressure with a rate of $\partial T_{max}/\partial P \simeq 40$ K/GPa. This change in T_{max} is larger than those of other HF compounds, $\partial T_{max}/\partial P = 10-30$ K/GPa.[15,16] In other words, it is expected that we can easily induce the valence instability in $CeInCu_2$ by applying pressure.

In this paper the pressure dependence of $\rho(T)$ of single crystalline $CeInCu_2$ is reported up to 8 GPa in order to examine whether a valence instability is induced or not by pressure. The results about T_{max} and A are discussed in connection with the effect of pressure on T_K or $|JN(0)|$ by using the Yoshimori-Kasai theory.[17]

Single-crystalline $CeInCu_2$ was prepared by the Czochralski pulling method. The details of preparation were reported elsewhere.[10] Electrical resistance was measured by the standard four-probe method in which four Au leads (of 20 μm cross-sectional diameter) were attached on the sample by gold paste. The hydrostatic pressure was generated by a cubic-anvil press having an anvil face of 4×4 mm^2.[18] The pressure-transmitting medium was a mixture of Fluorinert, FC 70 and FC 77.

Figure 1 shows the electrical resistance R (mΩ) of $CeInCu_2$ as a function of pressure at room temperature. R is found to increase with increasing pressure approximately in the linear fashion up to $\sim$2 GPa having a max-

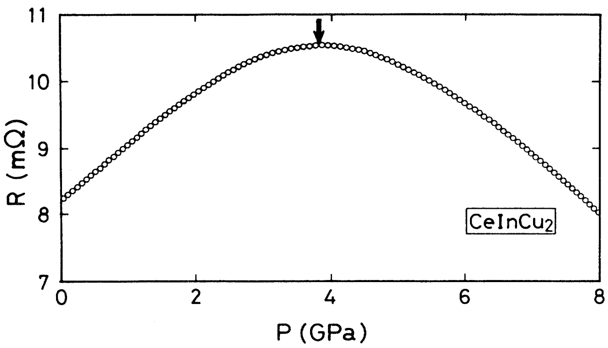

FIG. 1. Pressure dependence of electrical resistance R of $CeInCu_2$ at room temperature. The arrow shows the resistance-maximum pressure.

imum around 3.8 GPa. Above that R begins to decrease with increasing pressure. The pressure coefficients of R, $R^{-1}\partial R/\partial P$, are 0.10 GPa^{-1} at ambient pressure and -0.09 GPa^{-1} around 7 GPa. The absolute values are nearly the same. This type of behavior against pressure was observed in the other HF compounds.[5,7] The maximum in the R-P curve is due to the Kondo effect, as will be mentioned later.

The temperature dependence of resistivity $\rho(T)$ at various pressures up to 8 GPa is shown in Fig. 2. The values of ρ at high pressure were corrected by taking into account the change of geometrical factor $1/S$ of the specimen at high pressure: $1/S$ increased by about 2% at 8 GPa, which was obtained by the previous data of the thermal expansion[13] and the compressibility.[19] At ambient pressure, ρ increases gradually with decreasing temperature, reaches a maximum around 27 K and then decreases by further cooling.

The temperature of the resistivity maximum, T_{max}, is shown by an arrow in Fig. 2. It is found that T_{max} increases with increasing pressure and the maximum in $\rho(T)$ curves tends to be smeared at high pressure. In the temperature range of the present work, the $\rho(T)$ above 4.5 GPa increases monotonously with temperature. Such a variation in the $\rho-T$ curve by applying pressure has also been observed for other HF compounds.[5,7]

This behavior may be explained in the following way.

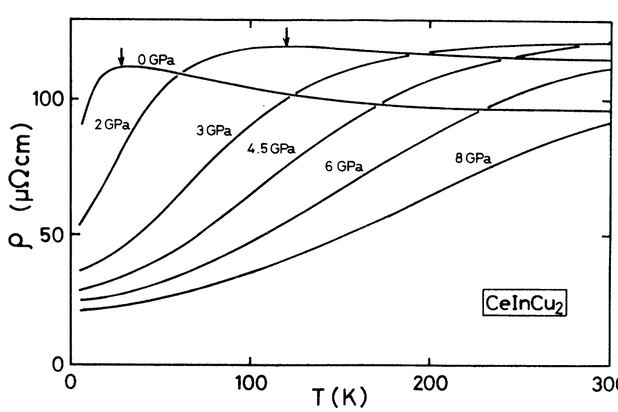

FIG. 2. Electrical resistivity ρ of $CeInCu_2$ under high pressure as a function of temperature T (K). The arrows indicate the resistivity-maximum temperature T_{max}.

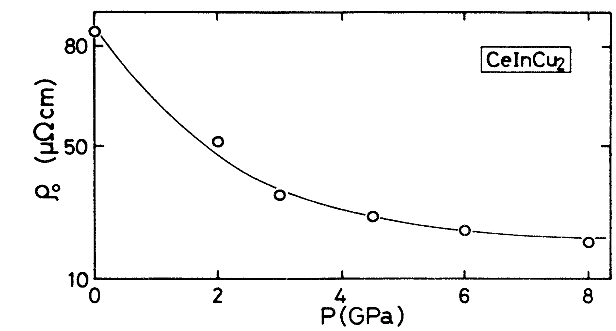

FIG. 3. Pressure dependence of the residual resistivity ρ_0.

The pseudobinary system $Ce(In_{1-x}Sn_x)_3$ is well known to show a crossover from CKS ($x=0$) to IVS ($x=1$).[20] The $\rho(T)$ at $x=0$ ($CeIn_3$) shows a well-defined maximum around 50 K, which is characteristic of CKS. The maximum becomes less prominent with increasing x. At $x=1$ ($CeSn_3$), $\rho(T)$ increases only monotonously with temperature ($T<300$ K), which is very similar to the $\rho(T)$ of $CeInCu_2$ at 8 GPa. Taking these facts into consideration, the change in the overall behavior in the $\rho(T)$ of $CeInCu_2$ observed in Fig. 2 implies a crossover from CKS at low pressure ($\lesssim 3$ GPa) to IVS at high pressure ($\gtrsim 4.5$ GPa). The maximum of the R-P curve in Fig. 1 is considered to be due to a shift of T_{max} to higher temperatures by applying pressure: T_{max} may be around 300 K at 3.8 GPa.

The residual resistivity ρ_0 is shown in Fig. 3 as a function of pressure. ρ_0 decreases with increasing pressure. The decreasing rate below ~ 3 GPa is larger than that above ~ 4.5 GPa. The larger decrease in ρ_0 may correspond to a large decrease in effective mass due to the crossover from CKS to IVS by applying pressure.

In order to examine the T^2 dependence in the $\rho(T)$, $\rho(T)$-ρ_0 is plotted as a function of T^2 in Fig. 4. As the pressure increases, the temperature range of T^2 dependence becomes wider compared with that at ambient pressure, i.e., T_{coh} may increase with pressure. These behaviors were observed in other HF compounds including U-compounds.[5,21-23] The value of A is shown in Fig. 5 as a function of pressure. It is seen that A decreases

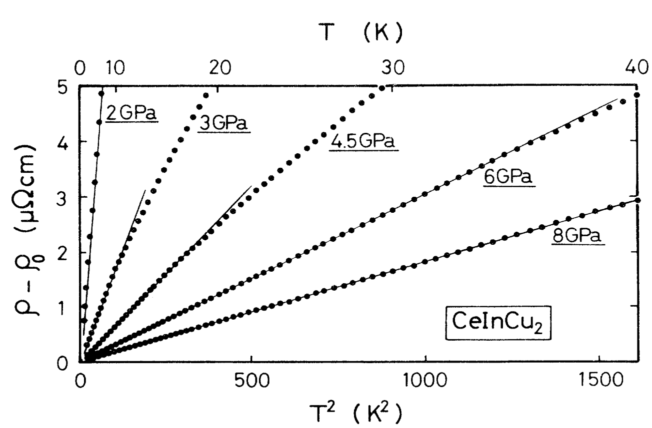

FIG. 4. ρ-ρ_0 vs T^2 below 40 K. The solid lines show the T^2 dependence.

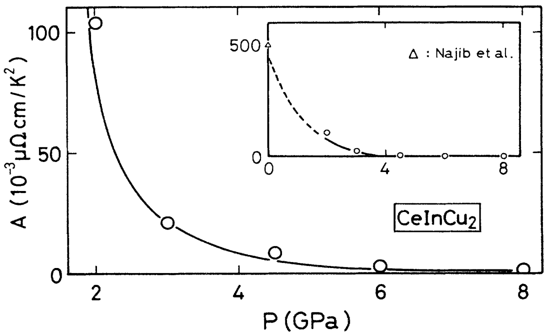

FIG. 5. The coefficient A of the T^2 term as a function of pressure P. The inset illustrates the values of A in the expanded range. The A open triangle value at 0 GPa obtained by Najib *et al.* (Ref. 14) is shown by Δ.

largely with increasing pressure below ~3 GPa and the decreasing rate becomes small at high pressure.

According to the theory of Yoshimori and Kasai,[17] A is proportional to T_K^{-2} and T_K is approximately proportional to T_{max}, i.e.,

$$T_{max} \propto \frac{1}{\sqrt{A}} . \tag{1}$$

In order to check this relation, we plotted the value of $1/\sqrt{A}$ as a function of T_{max} in Fig. 6. $1/\sqrt{A}$ is found to increase linearly with T_{max} as is shown by a solid line in Fig. 6. The large decrease in A in the low-pressure range in Fig. 5 is due to the large increase of T_K by applying pressure. This fact indicates that the present results are well understood in the framework of the Yoshimori-Kasai theory.

Next we discuss the pressure dependence of T_{max}. T_{max} is described as

$$T_{max} \propto T_K \propto \exp\left[-\frac{1}{|JN(0)|}\right] \quad (J<0) , \tag{2}$$

where J is the exchange interaction between the conduction electron and localized $4f$ spin and $N(0)$ is the density of states at the Fermi level. The increase of T_{max} with pressure indicates the increase of $|JN(0)|$.

Next we try to estimate the magnitude of $|JN(0)|$. In the compressible Kondo model,[24] the volume dependence of $|JN(0)|$ is assumed as follows:

$$|JN(0)| = |JN(0)|_0 \exp\left[-q\frac{V-V_0}{V_0}\right] , \tag{3}$$

where $|JN(0)|_0$ is the value of $|JN(0)|$ at ambient pressure, q is a numerical constant usually taken to be between 6 and 8, V and V_0 are the volume at pressure P and at ambient pressure, respectively. If the change in volume is small, we would have the following expression:

$$\frac{1}{|JN(0)|} = \frac{1}{|JN(0)|_0} \exp\left[q\frac{V-V_0}{V_0}\right]$$
$$\simeq \frac{1}{|JN(0)|_0}(1-\kappa_0 q P) , \tag{4}$$

where κ_0 is the compressibility. Then, from Eqs. (2) and (4), we have

$$\frac{T_{max}(P)}{T_{max}(0)} \simeq \exp\left[\frac{\kappa_0 q P}{|JN(0)|_0}\right] . \tag{5}$$

This equation shows that the sensitivity of T_K or T_{max} against pressure depends on the factor $\kappa_0 q/|JN(0)|_0$.

Figure 7 shows $\ln[T_{max}(P)/T_{max}(0)]$ as a function of pressure P (GPa). $\ln[T_{max}(P)/T_{max}(0)]$ has approximately linear dependence on P. From this result, the value of $\kappa_0 q/|JN(0)|_0$ is about 0.76 GPa^{-1}. By substituting the observed value of κ_0, 10.3×10^{-3} GPa^{-1} (Ref. 19) and assuming $q=6$, which is the same as the case of $CeCu_6$,[25] $|JN(0)|_0$ is estimated to be 8.1×10^{-2}. $|JN(0)|_0$ was reported to be 12×10^{-2} for $CeInCu_2$ (Ref. 11) and 9.1×10^{-2} for $CeCu_6$.[25] Considering the facts, $|JN(0)|_0$ of HF materials may be of the order of 0.1. To confirm this point, one needs to accumulate the data of $|JN(0)|_0$ in the wide range of Kondo compounds.

Finally we briefly discuss the effect of crystalline electric field (CEF) on the $\rho(T)$ at high pressure. There have been few data about the effect of pressure on the CEF of the HF compounds. According to the measurement of

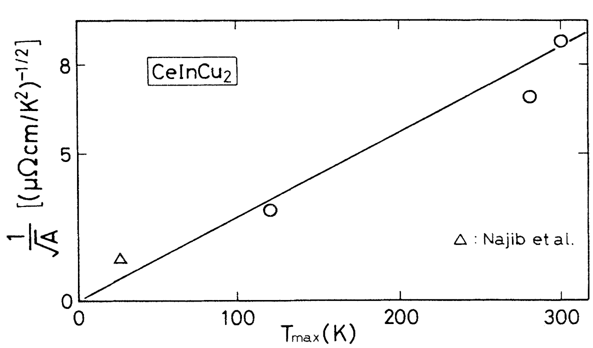

FIG. 6. $1/\sqrt{A}$ plotted against T_{max}. The solid line indicates a linear dependence. Δ shows the data obtained by Najib *et al.* (Ref. 14).

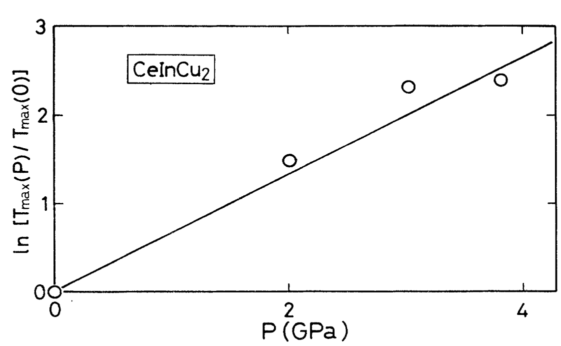

FIG. 7. Values of $\ln[T_{max}(P)/T_{max}(0)]$ as a function of pressure.

the thermal expansion coefficient of $CeInCu_2$,[13] the pressure coefficient of CEF splitting energy Δ, $\partial \ln\Delta/\partial P$, is about 0.3 GPa^{-1}. On the other hand, $\partial \ln T_K/\partial P \simeq \partial \ln T_{max}/\partial P$ is 0.76 GPa^{-1} as is obtained from Eq. (5). Using the values of Δ and T_K at ambient pressure,[10] $\partial\Delta/\partial P$ and $\partial T_K/\partial P$ are estimated to be $\sim$20 and 4 K/GPa, respectively. From this rough estimation, it is suggested that the temperature range in which $\rho(T)$ is dominated by the Kondo effect is largely different at high pressure from that by CEF splitting. The effect of CEF on $\rho(T)$ at low temperatures is expected to decrease by applying pressure. Thus, in the present work, we assumed that the effect of CEF on $\rho(T)$ is negligibly small in the present high-pressure range ($P \gtrsim 2$ GPa) as far as the low-temperature $\rho(T)$ is concerned. However, in order to do a more detailed discussion of the CEF effect on $\rho(T)$, we need more data about $\rho(T)$, heat capacity, and thermal expansion coefficients below 2 GPa.

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