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Effect of pressure on the electrical resistivity of $CePtSi_2$

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Abstract

Electrical resistivity $\rho(T)$ of the concentrated Kondo compound $CePtSi_2$ has been measured as a function of temperature at various pressures up to 80 kbar. Two maxima are observed near 5 K ($=T_1$) and 28 K ($=T_2$) in the $\rho(T)$ curve at ambient pressure. It is found that T_2 increases with increasing pressure but T_1 disappears above 5 kbar and the two maxima are merged into a broad maximum above 20 kbar. The overall behavior of the $\rho(T)$ curves at high pressures indicates that a crossover in the electronic state of $CePtSi_2$ is induced by applying pressure. On the basis of the data in the present work, we examine a semi-quantitative analysis to obtain the value of $|JN(0)|$ at ambient pressure.

1. Introduction

It is well known that the physical properties of concentrated Kondo compounds (CKC) are strongly affected by external forces such as temperature, magnetic field, high pressure and so forth [1]. High pressure is a useful tool to investigate the mechanism of the CK effect because the electronic state changes drastically by applying pressure, *i.e.* the characteristic temperature T_K (the Kondo temperature) is strongly dependent on pressure. Many interesting electronic transitions (or crossovers) induced by pressure have been reported [1–3].

$CePtSi_2$ crystallizes in the $CeNiSn_2$-type orthorhombic structure. Recently, this compound was found to be a magnetically ordered CKC ($T_N=1.5$ K) with a large C/T value of about 1.5 J/mol K^2 at 1.5 K [4,5] and a large enhancement of α/T at low temperature [6], where C and α are the specific heat and the thermal expansion coefficient. T_K of $CePtSi_2$ was estimated to be around 3 K.

In the present work, we made an attempt to measure the electrical resistance of $CePtSi_2$ up to 80 kbar in order to investigate the instability of the electronic state at high pressure. We also observed the pressure dependence of lattice constants of $CePtSi_2$ to clarify the existence of the volume anomaly associated with the pressure-induced crossover. By using these data, we estimate the value of the most important parameter in the Kondo system, $|JN(0)|$ at ambient pressure, where J is the coupling constant and $N(0)$ is the density of state at the Fermi energy.

2. Experimental procedure

Polycrystalline samples of $CePtSi_2$ and $LaPtSi_2$ were prepared by arc-melting stoichiometric amounts of starting materials, Ce(3N), La(3N), Pt(3N) and Si(6N) in an argon atmosphere. The detailed characterization of the samples has been described previously [6]. Electrical resistance measurements were carried out in the temperature range between 2 K and 300 K by using the standard four-probe method. The hydrostatic pressure was generated by using a Cu–Be piston-cylinder device up to 20 kbar and a cubic-anvil press above 20 kbar with an anvil face of 4×4 mm^2. A 1:1 mixture of Fluorinert, FC 70 and 77, was used as a pressure transmitting medium. The pressure was changed only at room temperature to minimize the internal strain in the sample and kept constant within ±1% throughout the measurement. The details of the high pressure system have been reported elsewhere [7,8].

Hydrostatic pressure for X-ray diffraction 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 disk-shaped beryllium pressure vessel 0.5 mm thick [9]. A 4:1 mixture of methanol and ethanol was used as the pressure transmitting medium. The pressure

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was determined using Decker's equation of state for NaCl [10].

3. Results

3.1. Electrical resistivity under high pressure

Figure 1 shows the $\rho(T)$ curves of CePtSi$_2$ at high pressure up to 80 kbar and LaPtSi$_2$ at ambient pressure. A drastic change is seen in $\rho(T)$. At ambient pressure $\rho(T)$ of CePtSi$_2$ shows a characteristic temperature dependence on CKC: $\rho(T)$ increases with decreasing temperature until it shows two maxima at 28.5 K ($=T_2$) and 5.4 K ($=T_1$). ρ of LaPtSi$_2$ shows a smooth change against temperature, which is the same behavior as that of normal metal. This result indicates that the anomalous temperature dependence of CePtSi$_2$ originates from the unstable 4f electrons of Ce.

By applying pressure, the two maxima at T_1 and T_2 in the $\rho(T)$ curve are found to merge into one maximum ($T_{\max}$) (see below). $T_{\max}$ also increases with increasing pressure but the maximum becomes broader than that at ambient pressure. It seems to be difficult to define $T_{\max}$ in the $\rho(T)$ curve at 80 kbar. Similar behavior in the $\rho(T)$ curve at high pressure has been observed in the other Ce compounds, which is interpreted as a pressure-induced crossover from the CK state or heavy fermion (HF) state (low T_K) to the intermediate valence (IV) state (high T_K) [11,12].

The magnetic contribution to ρ due to 4f electrons, ρ_{mag}, is estimated by using the relation, $\rho_{\text{mag}}=\rho(\text{CePtSi}_2)-\rho(\text{LaPtSi}_2)$, assuming that $\rho(\text{LaPtSi}_2)$ indicates the phonon contribution and is independent of pressure. The $\rho_{\text{mag}}(T)$ curve below 20 kbar is shown in Fig. 2 in the low temperature part below 50 K. T_1 and T_2 are shown by arrows. Because of a problem with the experiment at 10 kbar, only the data above 8 K are shown in Fig. 2. T_1 and T_2 increase slowly with pressure having coefficients of $\partial T_1/\partial P=0.12$ K/kbar and $\partial T_2/\partial P=0.10$ K/kbar, respectively. Although the knee-type anomaly in $\rho(T)$ is clearly seen near 5 K below 5 kbar, no trace of that is observed at 15 kbar. This fact indicates that T_2 is merged into T_1 above 10 kbar to give $T_{\max}$.Figure 3 shows T_1, T_2 and $T_{\max}$ as a function of pressure. $T_{\max}$ increases with pressure and its pressure coefficient is larger than $\partial T_1/\partial P$ and $\partial T_2/\partial P$. It is difficult to determine at what pressure T_1 and T_2 merges into

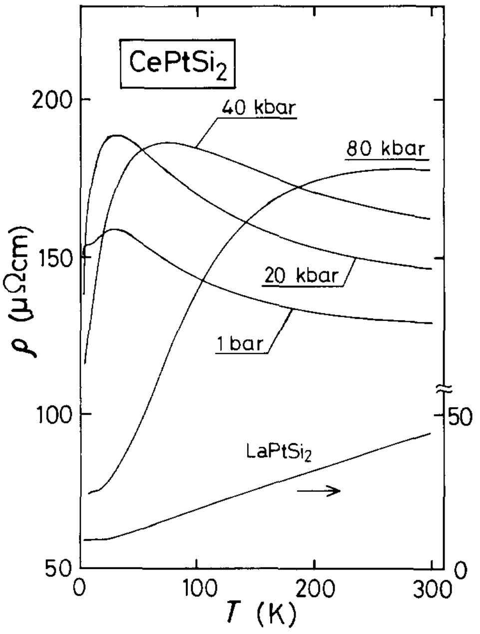

Fig. 1. The $\rho(T)$ curves of CePtSi$_2$ at various pressures up to 80 kbar. The ρ(T) curve of LaPtSi$_2$ is also shown for comparison.

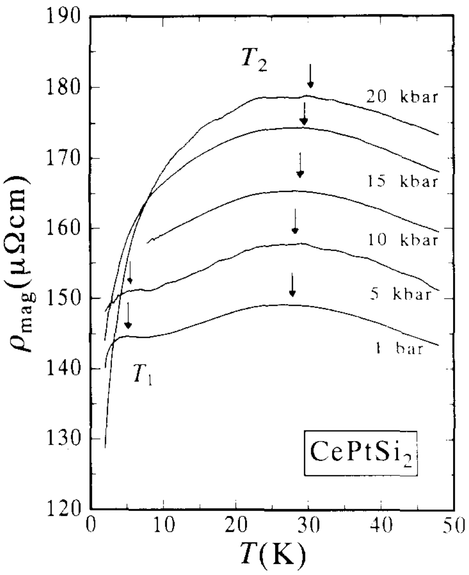

Fig. 2. Magnetic contribution to the electrical resistivity, ρ_{mag}, as a function of temperature at various pressures. T_1 and T_2 are shown by arrows.

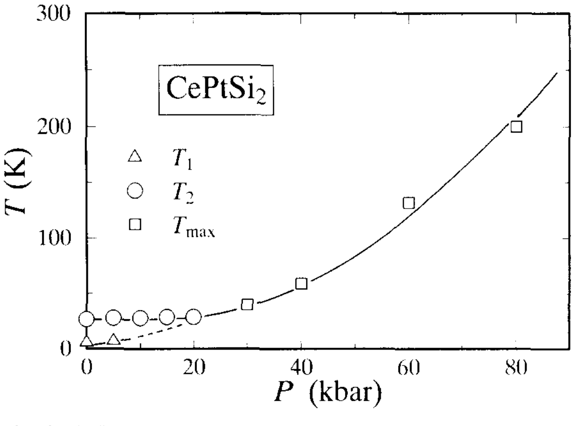

Fig. 3. P–T phase diagram of CePtSi$_2$ determined from the $\rho(T)$ curves at high pressures. The definitions of T_1, T_2 and $T_{\max}$ are described in the text.

T_{max}. Such behavior was also observed in the heavy fermion compound $CeCu_2Si_2$ [13].

3.2. *X-Ray diffraction under high pressure*

Since no new diffraction lines were observed, the orthorhombic $CeNiSi_2$-type structure is stable at least up to 60 kbar at room temperature. The lattice constants were obtained by using eight reflections at each pressure. These are found to decrease smoothly with pressure, having no discontinuous change within experimental error. The ratio V/V_0 is plotted as a function of pressure in Fig. 4, where V and V_0 are the volume at high and ambient pressure, respectively. The solid line shows the result of least-squares fit to the first-order Murnaghan's equation of state,

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

where B_0 and B_0' are the zero-pressure values of the isothermal bulk modulus and its pressure derivative, respectively. These are estimated to be 1190 kbar and 3.0. B_0 of $CePtSi_2$ is larger than that of the typical CK or heavy fermion (HF) compound $CeInCu_2$ ($B_0 = 901$ kbar) but B_0' is smaller ($B_0' = 3.9$ for $CeInCu_2$) [9].

4. Discussion

4.1. *Pressure-induced crossover from the CK state to the IV state*

As is seen from the overall behavior of $\rho(T)$ curves in Fig. 1, we expect a drastic change in the electronic state of CKC $CePtSi_2$ at high pressure. We reported previously the crossover from the HF to the IV state induced by pressure for HF compounds $CeInCu_2$ [2] and $CeCu_6$ [14]. In these materials, the value of $\partial T_{max}/\partial P$ is about (3–4) K/kbar, which is an order of magnitude larger than those of $\partial T_i/\partial P$ ($i=1,2$). But the average value of $\partial T_{max}/\partial P$ between 40 kbar and 80 kbar is about 3.5 K/kbar, which is comparable with those of HF compounds. It should be noted that similar P–T phase diagrams as shown in Fig. 3 are reported for other HF compounds, $CeCu_2Si_2$ [13] and $CeCu_2Ge_2$ [15], in which two maxima were observed in the $\rho(T)$ curves merging into a broad peak at high pressure. This complicated behavior of T_1, T_2 and T_{max} is considered to be due to the interplay of the Kondo effect and crystalline electric field as pointed out by Yamada *et al.* [16].

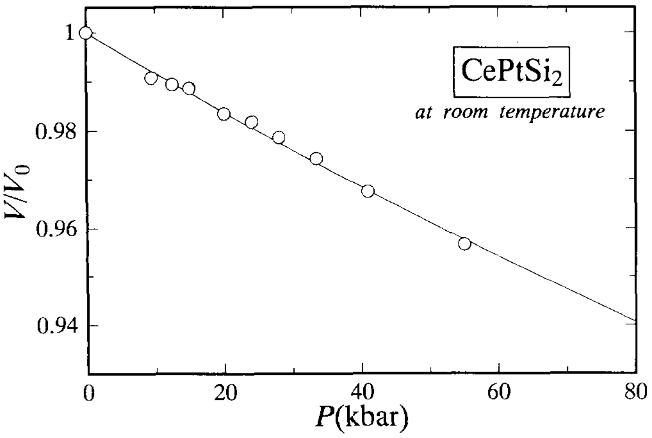

Fig. 4. The pressure dependence on volume of $CePtSi_2$ at room temperature. The solid curve is determined by the least-squares fit to the first-order Murnaghan's equation.

The Kondo temperature T_K is described as

$$T_K \propto \exp\left[-\frac{1}{|JN(0)|}\right] \tag{2}$$

where J is the coupling constant and $N(0)$ is the density of state at the Fermi energy. Since T_{max} is proportional to T_K [17], an increase in T_{max} with pressure indicates an increase in $|JN(0)|$. In other words, $|JN(0)|$ is one of the most fundamental parameters because T_K is mainly dominated by its change under the application of pressure. The large change in the magnitude of $|JN(0)|$ gives rise to the drastic change in T_{max} or T_K. Here we assume the following volume dependence of $|JN(0)|$ [18]:

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

where $|JN(0)|_0$ is the value of $|JN(0)|$ at ambient pressure and q is a dimensionless constant. q may be defined as a Grüneisen parameter of $|JN(0)|$,

$$q=-\left.\frac{\partial \ln|JN(0)|}{\partial \ln V}\right|_{V=V_0} \tag{4}$$

and usually has a value between 6 and 8 [2,12,18]. From eqns. (2) and (4), the Grüneisen parameter for T_K is written

$$\left.\frac{\partial \ln T_k}{\partial \ln V}\right|_{V=V_0}=\frac{q}{|JN(0)|_0} \tag{5}$$

Then we obtain the following relations:

$$\ln\frac{T_{max}(P)}{T_{max}(0)}=\frac{q}{|JN(0)|_0}\,\frac{V_0-V}{V_0} \tag{6}$$

By using the values of $T_{max}(P)$ in Fig. 3 and the pressure dependence of volume in Fig. 4, the values of left-hand side $\ln[T_{max}(P)/T_{max}(0)]$ are plotted in Fig. 5 as a function of the fractional volume change $(V_0-V)/V_0$. From Fig. 5, we obtained $q/|JN(0)|_0 = 47.3$ and then $|JN(0)|_0 = 0.13$ by assuming $q=6$. The magnitude of $|JN(0)|_0$ is comparable with that of HF (~0.1) [12] but

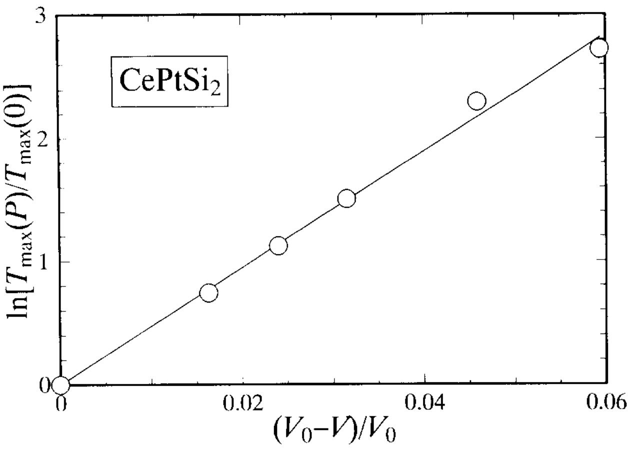

Fig. 5. The value of $\ln[T_{max}(P)/T_{max}(0)]$ as a function of the fractional volume $(V_0 - V)/V_0$.

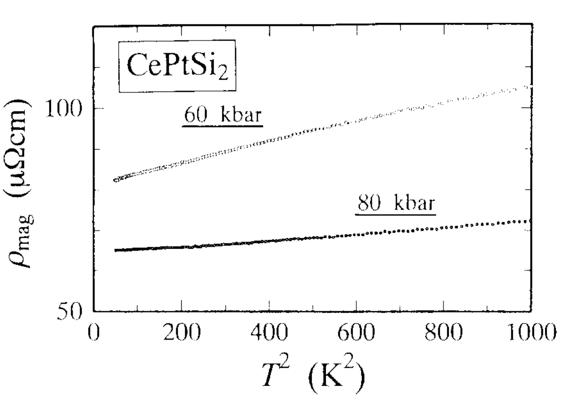

Fig. 6. ρ_{mag} as a function of T^2 at 60 kbar and 80 kbar.

smaller than that of IVC such as α-Ce and CeNi (~0.5) [19].

4.2. Fermi liquid behavior at low temperature and high pressure above 30 kbar

It is shown that the strong T^2 dependence in $\rho(T)$, which is a characteristic of Fermi liquids, is observed in the HF compounds in the low temperature range $T < T_K$ [2,12]. In $CePtSi_2$, however, such a term is masked by the existence of antiferromagnetic ordering ($T_N \approx 2$ K) and a crystalline electric field (T_1). These effects may be removed by the application of pressure as shown by Figs. 1 and 2. In order to examine the T^2 term at high pressure, the values of ρ_{mag} are plotted as a function of T^2. A selection of the plot is presented in Fig. 6. The T^2 term is seen clearly in the $\rho(T)$ curves at 60 and 80 kbar. The coefficients A ($\times 10^3$) of the T^2 term are estimated to be 25 and 7.7 μΩ-cm K^2 at 60 and 80 kbar, respectively. These values are comparable with those of $CeCu_6$ [12]. Thus, the electronic state of $CePtSi_2$ at high pressure is described by the Fermi liquid similar to other Ce-based heavy fermions.

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