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Intermediate valence state of CeNi single crystal at high pressure

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Abstract

Electrical resistivity $\rho(T)$ of single crystalline CeNi has been measured at high pressure and low temperature in order to clarify the stability of the intermediate valence state of CeNi at high pressure. T^2-dependent electrical resistivity is found at low temperature and its coefficient A decreases with increasing pressure. We estimated the value of $|JN(0)|$ at ambient pressure for CeNi to be about 0.6.

1. Introduction

Ce compounds show a wide variety of electrical and magnetic properties due to the hybridization between unstable 4f electrons and the conduction band. Some of them are characterized as heavy fermions (HF) and others as intermediate valence (IV) compounds depending on the magnitude of hybridization [1]. It is well known that the anomalous properties of Ce compounds are strongly dependent on a change in the volume or the pressure. The large change in the electronic properties in these compounds by application of pressure is well understood by taking account of the volume dependence of the characteristic temperature, the so-called Kondo temperature, T_K.

CeNi crystallizes in the CrB-type orthorhombic structure and is a typical IV compound with $T_K \approx 100$ K [2]. The magnetic susceptibility χ shows Curie–Weiss behavior well above T_K and enhanced Pauli paramagnetism below T_K [3]. Furthermore, the electrical resistivity $\rho(T)$ has a T^2-term at low temperature showing the Fermi liquid property.

In order to settle the electronic state of CeNi at high pressure, we made an attempt to measure the $\rho(T)$ of single crystalline CeNi up to 8 GPa. The coefficients of the T^2 -term A are obtained as a function of pressure and the results are discussed in the light of the phenomenological theory presented recently by Kagayama *et al.* [4].

2. Experimental details

A single crystal was grown by the Bridgman method. A rectangular specimen was cut from the single crystal using a wire saw. Since the sample was a thin sheet (a–c plane), the resistivity of CeNi was measured along the a- and c-axes and at high pressures only along the c-axis. High pressure was generated by means of a piston and cylinder device and a 1:1 mixture of transformer oil and kerosene as a pressure transmitting medium. The load was kept constant within ±5% by controlling the oil pressure of the hydraulic press. The details of the present high pressure apparatus have been described in detail elsewhere [5,6].

3. Results and discussion

Figure 1 shows the $\rho(T)$ of single crystalline CeNi in the current direction along a-(ρ_a) and c-axis (ρ_c) at ambient pressure. The $\rho_a(T)$ curve of a LaNi single crystal is also shown for comparison. ρ_a and ρ_c of CeNi

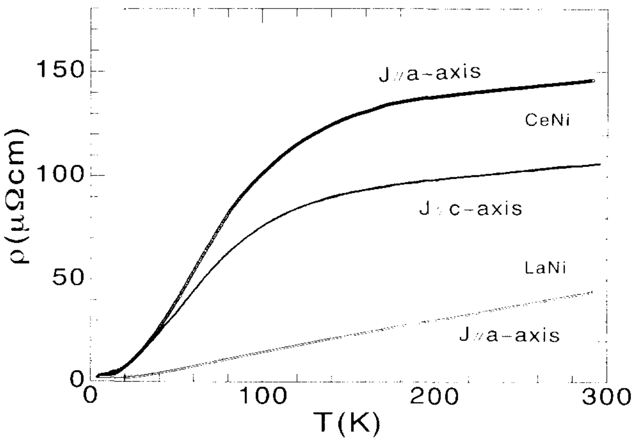

Fig. 1. Electrical resistivity $\rho(T)$ of single crystalline CeN and LaNi at ambient pressure.

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are found to increase rapidly with increasing temperature below *ca.* 100 K followed by a gradual increase above 150 K. This behavior suggests the T_K of CeNi to be around 120 K. On the other hand, the $\rho_a(T)$ curve of LaNi shows a smooth increase like normal metals. $\rho_a(T)$ and $\rho_c(T)$ of CeNi are described well as $\rho_i(T)=\rho_{i,0}+A_iT^2$ ($i=a$ or c) at low temperatures, where $\rho_{i,0}$ is the residual resistivity along the i-axis. The coefficients A are estimated to be 1.41×10^{-2} μΩ-cm K^{-2} ($T<45$ K) and 1.23×10 μΩ-cm K^{-}2 ($T<37$ K) for $J//a$-axis and $J//c$-axis, respectively. These values are about 10 times larger than those of normal metals [7].

The $\rho_c(T)$ curves of the CeNi single crystal are shown in Fig. 2 as a function of temperature at various pressures. The value of ρ_c at ambient pressure was reproduced within an experimental error after releasing the pressure. A drastic change in the temperature dependence of ρ_c is seen in Fig. 2; by an application of pressure the rapid increase in $\rho_c(T)$ at ambient pressure becomes less distinct and the $\rho_c(T)$ curve tends to be similar to that of LaNi as seen from $\rho_c(T)$ at 6 GPa. This type of behavior against pressure in the $\rho(T)$ curve has been observed in other Ce compounds such as $CeCu_6$ [8], $CeInCu_2$ [9] and $CeCu_2Si_2$ [10]. The drastic change in $\rho(T)$ is considered as the crossover in the electronic state of the Ce compound from low T_K (concentrated Kondo state) to high T_K (intermediate valence state) by applying pressure.

We estimate the magnetic contribution to the electrical resistivity, ρ_m, of CeNi as follows. The electrical resistivity may by written

$$\rho(T)=\rho_0+\rho_{ph}(T)+\rho_m(T) \tag{1}$$

where ρ_0 is the residual resistivity and ρ_{ph} the contribution from phonon scattering. In the present work, we assume that $\rho_0+\rho_{ph}$ is approximated by $\rho_c(T)$ of LaNi, ρ_c(LaNi). Then we have the following equation:

$$\rho_m(T)=\rho_c(\mathrm{CeNi})-\rho_c(\mathrm{LaNi}) \tag{2}$$

Figure 3 shows ρ_m in the current direction along the c-axis as a function of T at various pressures. At ambient pressure, $\rho_m(T)$ shows a broad maximum around $T_m=160$ K as is shown by an arrow in Fig. 3. T_m increases to 220 K at 5 kbar. Above 10 kbar, no maximum is observed in the $\rho_m(T)$ curves. $\rho_m(0)$ seems to show a maximum around 30 kbar. We reported the same behavior of T_m against pressure for other Ce compounds [8,9]. Since T_m is roughly proportional to T_K [11], an increase in the magnitude of T_K at high pressure is due to the increase in $JN(0)$ [4,9], where J is the exchange interaction between f and conduction electrons and $N(0)$ is the density of state at the Fermi level. The estimation of $JN(0)$ at ambient pressure is shown later.

In order to examine the T^2 dependence in the $\rho_m(T)$ curve at low temperature, we plotted $\rho_m(T)$ as a function of T^2 in Fig. 4 at various pressures. The temperature range having T^2 dependence becomes wider by applying pressure. At 60 kbar, it is observed below 70 K. The coefficients A are shown in Fig. 5 as a function of pressure. A is found to decrease rapidly with increasing pressure; the value of A at 80 kbar is about 9.3×10^{-6} μΩ-cm K^{-2}, which is smaller than that at ambient pressure by an order of magnitude. But the

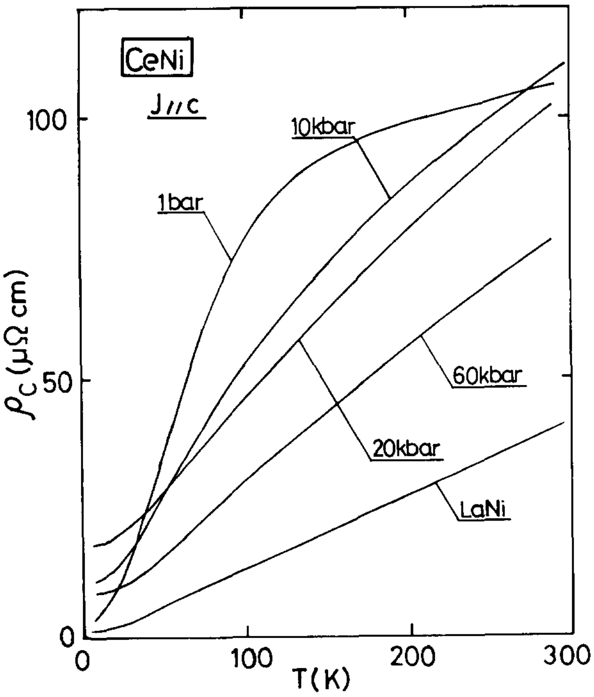

Fig. 2. Temperature dependent electrical resistivity $\rho_c(T)$ of CeNi and LaNi single crystals at various pressures.

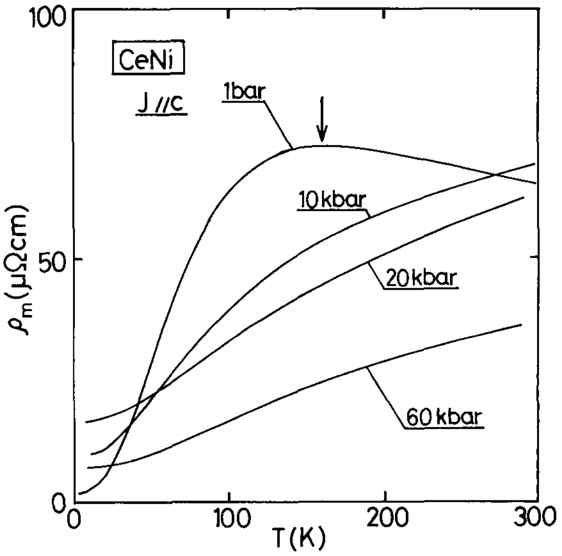

Fig. 3. Magnetic contribution to $\rho_c(T)$, $\rho_m(T)$, at various pressures. A broad maximum is shown by the arrow.

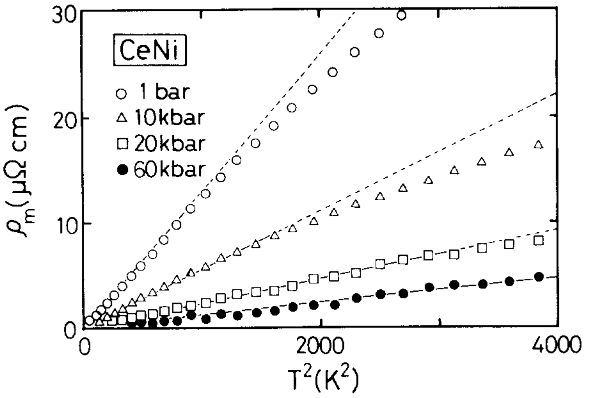

Fig. 4. ρ_m versus T plot of single crystalline CeNi at various pressures.

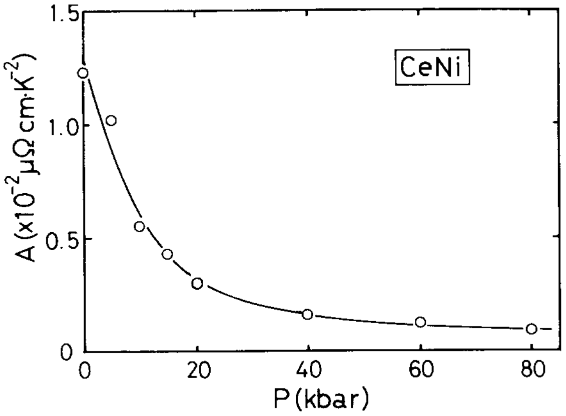

Fig. 5. Pressure dependence of T^2-coefficients A of CeNi.

decreasing rate of A becomes extremely small above 40 kbar. Such rapid decrease in the value of A by applying pressure was observed also in the other Ce compounds (the so-called heavy fermion compounds) such as $CeCu_6$, $CeInCu_2$ and so forth [4]. It is shown theoretically that A is proportional to $T_K{}^{-2}$ [11]. The rapid decrease in A below 20 kbar indicates a rapid increase in the value of T_K [9]. We presented previously a phenomenological model to interpret the pressure dependence of T_m and A in connection with T_K or $JN(0)$ by taking account of the volume change at high pressure [4]. We derived the following relation using the above model:

$$-\frac{1}{2}\ln\frac{A(P)}{A(0)} = \frac{q}{|JN(0)|_0}\,\frac{V_0-V}{V_0} \qquad (3)$$

where V and V_0 are the volume at high and at ambient pressure, respectively. Equation (3) is confirmed to be valid in the heavy fermion compounds, $CeCu_6$ and $CeInCu_2$ [4]. In the present work, we attempt to fit our data to eqn.(3). In Fig. 6, the values of $(1/2)\ln[A(P)/A(0)]$ are plotted as a function of the volume change $(V_0-V)/V_0$. Here we use the compressibility of CeNi of $4\times10^{-3}\,kbar^{-1}$. A linear relationship is found between these quantities. The value of $q/|JN(0)|_0$ is estimated to be 9.35 and then $|JN(0)|_0$ is 0.64 assuming $q=6$ [12]. This magnitude of $|JN(0)|_0$ is larger than those of the heavy fermion compounds $CeInCu_2$ and $CeCu_6$, $|JN(0)|_0\approx0.1$, but nearly the same as that of the intermediate valent α-Ce, ≈0.5 [13]. Thus, the large value of $|JN(0)|_0$ around 0.5 may be a characteristic of IV compounds.

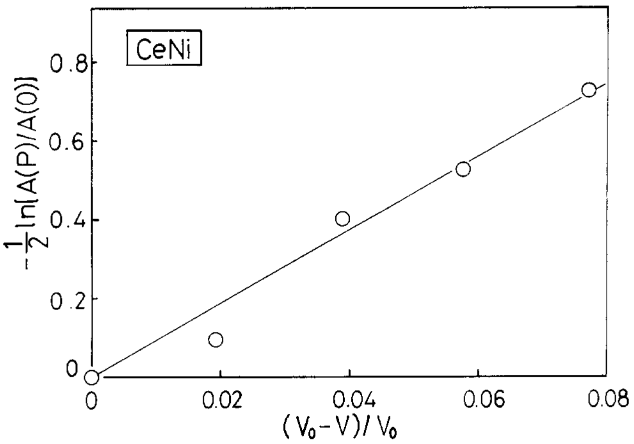

Fig. 6. The values of $-(1/2)\ln[A(P)/A(0)]$ as a function of volume change $(V_0-V)/V$.

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