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AC Magnetic Susceptibility Studies of Single Crystalline CeNiGe$_2$ under High Pressure

Masashi OHASHI[1]*, Gendo OOMI[2] and Isamu SATOH[3]

^{1}*Division of Civil and Enviromental Engineering, Kanazawa University, Kakuma-machi, Kanazawa 920-1192, Japan*
^{2}*Department of Physics, Kyushu University, 4-2-1 Ropponmatsu, Fukuoka 812-8581, Japan*
^{3}*IMR, Tohoku University, 2-1-1 Katahira, Sendai 980-8577, Japan*

The magnetic susceptibility of CeNiGe$_2$ single crystal has been measured under high pressure up to 2.0 GPa. Although two anomalies were observed at T_{N1} = 4.0 K and at T_{N2} = 3.0 K at ambient pressure, respectively, T_{N2} disappeared above 0.13 GPa while T_{N1} still remains near 2.0 K up to 2.0 GPa. On the other hand, the peak height of the susceptibility curve decreases with increasing pressure, which may suggest that the correlation between conduction and f-electron is strengthened by pressure. Kondo temperature T_K is estimated from the maximum of $\chi_c - T$ curve. T_K is remarkably enhanced by applying pressure.

KEYWORDS: Antiferromagnetic ordering, heavy fermion compound, AC magnetic susceptibility, Heartshorn bridge, quantum critical behavior

1. Introduction

Ce-based compounds have been well-known to show many interesting electronic properties such as magnetic ordering, superconductivity, and so forth[1]. These anomalous properties are considered to be due to electronic and magnetic instability of $4f$ electron state since several kinds of interactions such as RKKY interaction, the Kondo effect and crystalline electric field are competing each other in these compounds. A superconducting transition has been found in several compounds under hydrostatic pressure near a quantum critical point just at the border of magnetism where $T_N \rightarrow 0$, notably in CeCu$_2$Ge$_2$,[2] CePd$_2$Si$_2$[3] and CeRh$_2$Si$_2$.[4] All these systems show antiferromagnetic ordering at ambient pressure. Up to presents, we also have investigated the electronic properties of several Ce compounds under pressure by measuring the electrical resistance, thermal expansion, magnetic susceptibility, X-ray diffraction and so on.[5–9]

CeNiGe$_2$ crystallizes in orthorhombic CeNiSi$_2$-type layered structure (Cmcm),[10] which is constructed from deformed fragments of the CeGa$_2$Al$_2$ and α-ThSi$_2$ structures. The long b-axis brings about the flat Brillouin zone, which might produce a cylindrical Fermi surface along the b-axis. It is well-known that CeNiGe$_2$ is an antiferromagnetic Kondo system that orders magnetically at T_{N1} = 3.9 K and undergoes a spin structure rearrengement at T_{N2} = 3.2 K. It is expected to have competition between the RKKY interaction and Kondo effect because of its a quasi-two dimensional electronic state. In the present work, we report the effects of external hydrostatic pressure on the magnetic susceptibility of CeNiGe$_2$ single crystals. The results will be discussed in connection with the quantum critical behavior.

2. Experimental

Although these compounds do not melt congruently, the phase of the the orthorhombic CeNiGe$_2$ type structure can be grown by the Czochralsky method from Ge-rich sample as the initial one.[11,12] The pulling parameter was kept constant during the growth (pulling rate: 10 mm/h; seed rotation speed: 10 rpm; crucible-rotation speed: 5 rpm). A single crystal ingot was 3-5 mm in diameter. The samples of CeNiGe$_2$ were annealed for 2 weeks in a quartz tube under the vacuum of 1×10^{-6} Torr at 800 °C.

The samples were checked by conventional x-ray powder diffraction experiments using Cu-K$_\alpha$ radiation. The refined lattice parameters were a= 4.252 Å, b=16.78 Å and c= 4.202 Å, which are in good agreement with the previous values.[13–15] The single crystalline state was confirmed using back-scattering Laue technique and the four circle neutron diffractometer.[16] The DC magnetic susceptibility was measured at ambient pressure using a Quantum Design MPMS-5 superconducting quantum interference device magnetometer.

Hydrostatic pressure was generated by using a hybrid-type piston-cylinder device which is made of Ni-Co-Cr-Mo (MP35N) alloy for inner cell.[17] A 1:1 mixture of Fruorinert FC 70 and FC 77 was used as a pressure transmitting medium. The AC magnetic susceptibility was measured by using Heartshorn bridge method along the easy magnetization direction. The bridge circuit was prepared by using the inductive and the resistive component as shown in Figure 1. The primary and the secondary coils were wound on the sample directly and put into the pressure cell. The numbers of the turns were 100 for the primary coil by using 0.05 mm Cu wire and 200 + 200 for the secondary coil by using 0.02 mm Cu wire. In order to measure the pressure at low temperature, the conventional superconductor such as Pb is also put into the pressure cell.

A modulation frequency of 500 Hz was generated to the primary circuit. A peak-to-peak amplitude of the modulation field was ∼0.1 Oe. Both the inductive M and the resistive component R are roughly balanced at 4.2 K before measurement. The inductive balancer is fixed

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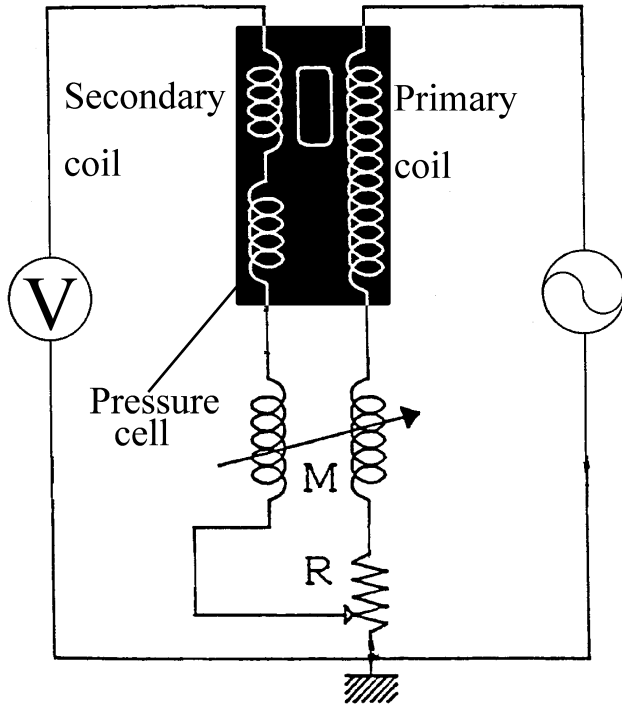*E-mail address: ohashi@t.kanazawa-u.ac.jp

Fig. 1. The bridge circuit for AC susceptibility measurement under high pressure.

Fig. 2. DC magnetic susceptibilities of CeNiGe$_2$ at ambient pressure.

Fig. 3. AC magnetic susceptibilities of CeNiGe$_2$ at various pressures. The arrows mark the characteristic temperatures, T_{N1}, T_{N2} and T_{max} (see text).

during the measurement. The output voltage of the secondary circuit V was directly connected to a lock-in amplifier and the inductive component was observed.

3. Results

Figure 2 shows the temperature dependence of DC magnetic susceptibilities of CeNiGe$_2$ in an applied magnetic field of 1 kOe along a-, b- and c-axis, respectively. It is found that $\chi(T)$ along c-axis is much larger than that for a- and b- axes, qualifying that the easy magnetic direction is parallel to c-axis, which is in agreement with that of Okada et al.[14] All the results of $\chi(T)$ along a-, b- and c-axes exhibit two anomalies near $T_{N1} \sim 3.7$ K and $T_{N2} \sim 3.0$ K, respectively. These results are in good agreement with the previous results.[13–15]

Figure 3 shows AC magnetic susceptibilities along the easy c-axis of CeNiGe$_2$ at various pressures. At ambient pressure, χ_c shows a sudden decrease at 4.0 K ($=T_{N1}$) and an anomaly at 3.0 K ($=T_{N2}$). T_{N1} obtained by AC susceptibility is found to be larger than that by DC one. It comes from the fact that the antiferromagnetic ordering at T_{N1} is sensitive to an applied magnetic field since DC magnetic susceptibility is measured at 1 kOe. Indeed, it is consistent with the fact that T_{N1} decreases as increasing magnetic field.[13–15]

T_{N2} disappears above 0.13 GPa. Further experiments is needed to clarify whether T_{N2} is merged into T_{N1} or decreases below 1.5 K at 0.13 GPa. On the other hand, T_{N1} still remains but an anomaly of $\chi_c(T)$ curve becomes less clear near T_{N1} as increasing pressure. Instead, the broad maximum of $\chi_c(T)$ curve is observed above T_{N1} under pressure.

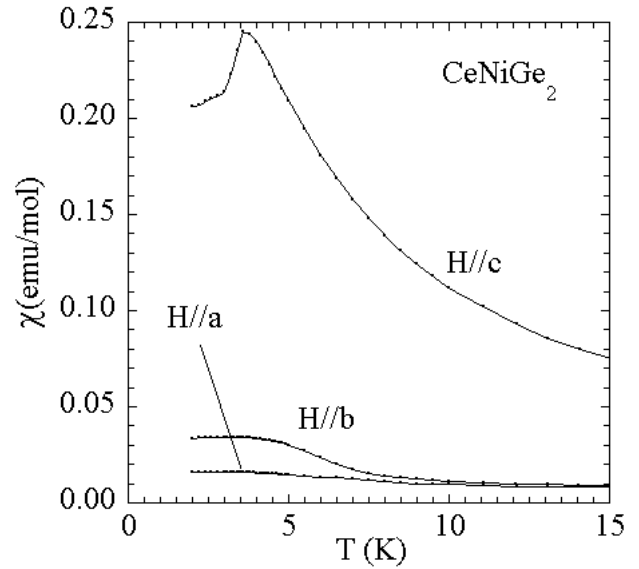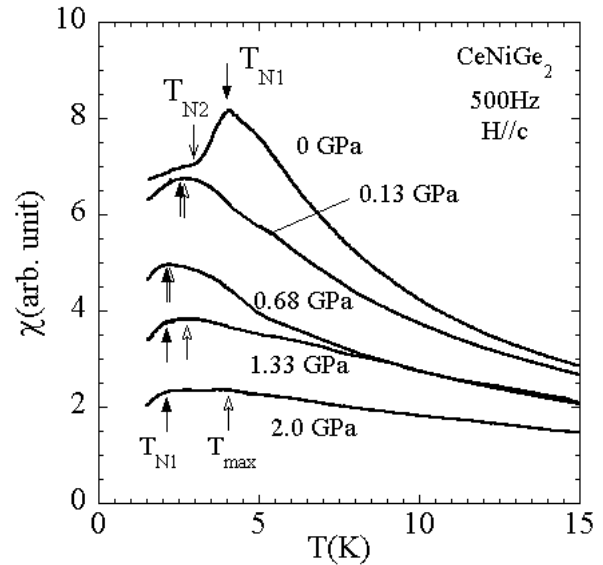Here the temperature derivative of AC susceptibility,

$d\chi_c/dT$, is shown in figure 4 as a function of temperature. T_{N1} and T_{N2} are defined as the temperature where $d\chi_c/dT$ shows a kink while T_{max} is estimated as that where $d\chi_c/dT = 0$. At ambient pressure, $d\chi_c/dT$ shows sharp anomalies at T_{N1} and T_{N2}, and T_{N1} is almost equal to T_{max} ($= 4.0$ K). As increasing pressure, some uncertainly as to the value of T_{max} arises because the magnitude of $d\chi_c/dT$ is suppressed and becomes zero in a wide temperature region. Indeed, at 2.0 GPa, T_{max} is estimated to be 4.0 ± 1.0 K. On the other hand, T_{N1} is easily estimated and is found to be suppressed by applying pressure.
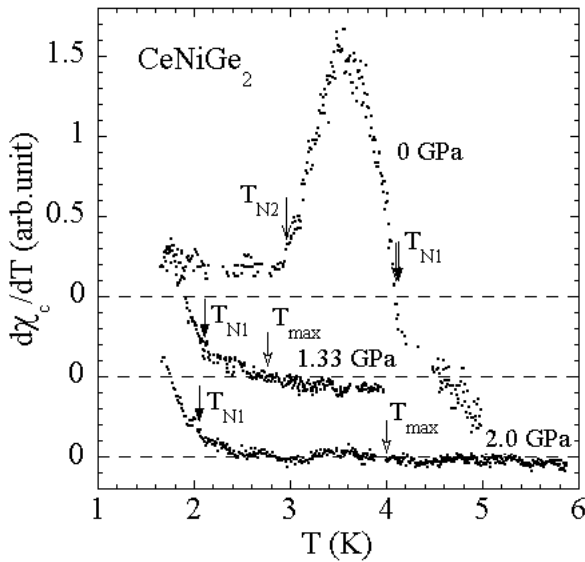
Fig. 4. Temperature derivative $d\chi_c/dT$ of CeNiGe$_2$ as a function of temperature at various pressures.
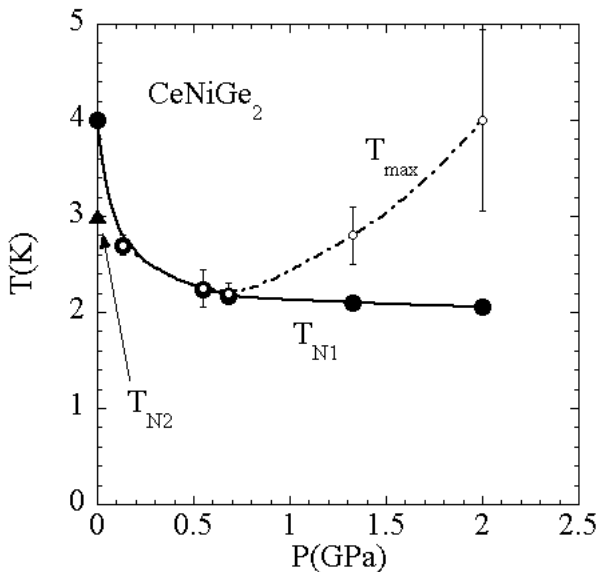

Fig. 5. T_{N1}, T_{N2} and T_{max} as a function of pressure.

4. Discussion

Figure 5 shows T_{N1}, T_{N2} and T_{max} of CeNiGe$_2$ as a function of pressure. It is found that T_{N1} decreases but tends to saturate as increasing pressure. The rate of $\partial T_{N1}/\partial P$ is estimated to be -0.2 K/GPa above 1.0 GPa. This behavior is contrary to the case of CeAl$_2$,[18] CeRh$_2$Si$_2$[19] et al., in which antiferromagnetic ordering temperature decreases as increasing pressure and quantum critical behavior is observed at the pressure near $T_N \rightarrow 0K$.

T_{max} is almost same as T_{N1} up to 0.68 GPa, indicating that T_{max} is related not only to the Kondo effect but also to the antiferromagnetic ordering. Here we note that the maximum value of the susceptibility, $\chi_c(T_{max})$, decreases as increasing pressure. This behavior is similar to the case

of CeRh$_2$Si$_2$.[20] It is explained to be due to the increase of the c-f hybridization, or to the decrease of the magnetic anisotropy. In other words, it is reasonable to assume that the Kondo temperature T_K increases even if T_{max} is suppressed by applying pressure.

Above 1 GPa, T_{max} increases as increasing pressure and a rate of $\partial T_{max}/\partial P$ tends to be enhanced by applying pressure. Assuming that T_{max} indicates the Kondo temperature T_K in the pressure region, T_{max} is described as

$$T_{max} \propto \exp\left(-\frac{1}{|JN(0)|}\right)$$

where J is the exchange interaction between the conduction electron and the localized $4f$ spin and $N(0)$ the density of states at the Fermi level. The increase of T_{max} with pressure indicates the increase of $|JN(0)|$. In the case of CeNiGe$_2$, it suggests that the large change in the magnitude of $|JN(0)|$ gives rise to the large change in T_{max} as increasing pressure. In other words, the electronic state becomes unstable as increasing pressure.

5. Summary

The AC magnetic susceptibility of CeNiGe$_2$ single crystal was measured under high pressure. We described T_{N1}, T_{N2} and T_K of CeNiGe$_2$ as a function of pressure up to 2.0 GPa. T_{N2} disappears at 0.13 GPa but T_{N1} still remains up 2 GPa and tends to saturate. Further measurements above 2.0 GPa and below 1.5 K are in progress in order to search for a quantum critical behavior although it is not known whether T_{N1} and T_{N2} are suppressed to 0 K or not. At $P > 1$ GPa, T_K increases as increasing pressure, and the magnitude of $\partial T_K/\partial P$ is enhanced by applying pressure.

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