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A change of electronic state tuned by pressure: pressure-induced superconductivity of the antiferromagnet $Ce_2Ni_3Ge_5$

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Abstract
We measured the electrical resistivity of an antiferromagnet $Ce_2Ni_3Ge_5$ with the orthorhombic crystal structure under pressure. The Néel temperature $T_N = 5.2$ K decreases with increasing pressure P and becomes zero at a critical $P_c \simeq 3.9$ GPa. The A and ρ_0 values of the low-temperature electrical resistivity $\rho = \rho_0 + AT^2$ in the Fermi liquid relation increase steeply above 3 GPa. A value of $A = 10.7\ \mu\Omega$ cm K^{-2} at 3.9 GPa is comparable to $A = 10\ \mu\Omega$ cm K^{-2} in a heavy fermion superconductor $CeCu_2Si_2$. The heavy fermion state was found to be formed around P_c, in which pressure region superconductivity was found below 0.26 K.

The cerium and uranium compounds form heavy fermions at low temperatures, which is a consequence of competition between the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction and the Kondo effect. High pressure is useful in tuning the electronic states in these compounds. When pressure is applied to the magnetically ordered cerium and uranium compounds, the ordering temperature T_{mag} decreases and becomes zero ($T_{mag} \to 0$) at a critical pressure $P = P_c$ [1].

The recent discovery of superconductivity in a non-Fermi-liquid compound $CeNi_2Ge_2$ [2] motivated us to study the Ce–Ni–Ge intermetallic compounds. There exist ten intermetallic compounds in the Ce–Ni–Ge ternary system. We investigated the electronic states of $CeNiGe_2$ [3], and $CeNiGe_3$ [4] by applying pressure. In an antiferromagnet $CeNiGe_3$ with

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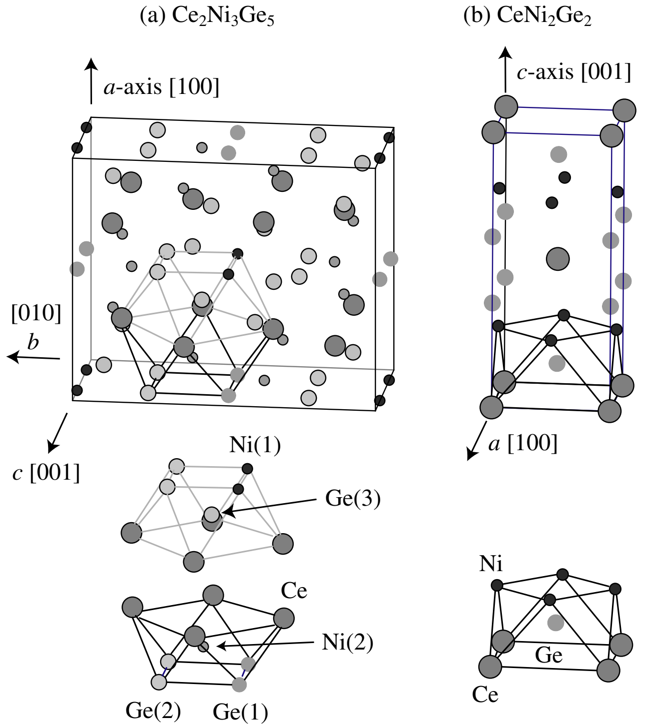

Figure 1. Crystal structures of $Ce_2Ni_3Ge_5$ (a) and $CeNi_2Ge_2$ (b).
(This figure is in colour only in the electronic version)

the orthorhombic crystal structure, we found superconductivity below 0.48 K in the pressure region from 4 to 10 GPa [4]. We continued in studying the pressure-induced superconductivity for one of the Ce–Ni–Ge compounds, namely $Ce_2Ni_3Ge_5$.

$Ce_2Ni_3Ge_5$ crystallizes in the $U_2Co_3Si_5$-type structure (*Ibam* space group), as shown in figure 1. According to the report after Durivault *et al* [5], it is noted that the orthorhombic structure of $Ce_2Ni_3Ge_5$ derives from the tetragonal ($ThCr_2Si_2$-type) structure of $CeNi_2Ge_2$. $CeNi_2Ge_2$ is based on a stacking of layers consisting of Ce_4Ni_4 antiprisms with the Ge atoms, as shown in figure 1(b), while the orthorhombic structure of $Ce_2Ni_3Ge_5$ in figure 1(a) results from a stacking of two distorted $Ce_4Ge(1)_2Ge(2)_2$ and $Ce_4Ni(1)_2Ge(2)_2$ antiprisms with the Ni(2) and Ge(3) atoms, respectively. The substitution of Ge(1) and Ge(2) atoms for Ni induces a deformation of the antiprisms. Another structural particularity is that the Ce atoms, which are located in planes perpendicular to the [001] direction (*c*-axis) in $CeNi_2Ge_2$, form wavy planes perpendicular to the [100] direction (*a*-axis) in $Ce_2Ni_3Ge_5$. Furthermore, the average interatomic distances $d_{\mathrm{Ce-Ni}} = 3.305$ Å and $d_{\mathrm{Ce-Ge}} = 3.176$ Å in $Ce_2Ni_3Ge_5$ are slightly comparable to 3.221 Å and 3.183 Å in $CeNi_2Ge_2$, respectively, suggesting the similar hybridization effect between the 4f electron in the Ce atom and the 4p electrons in the Ge atom for two compounds.

$Ce_2Ni_3Ge_5$ is an antiferromagnet with a Néel temperature $T_{\mathrm{N1}} = 4.8$ K and undergoes a successive trasition $T_{\mathrm{N2}} = 4.2$ K [5]. From the neutron diffraction experiment, only one transition is evidenced below T_{N1}, where the magnetic moments with an ordered moment of 0.4 μ_{B}/Ce are oriented along the [100] direction (*a*-axis), and the magnetic structure consists

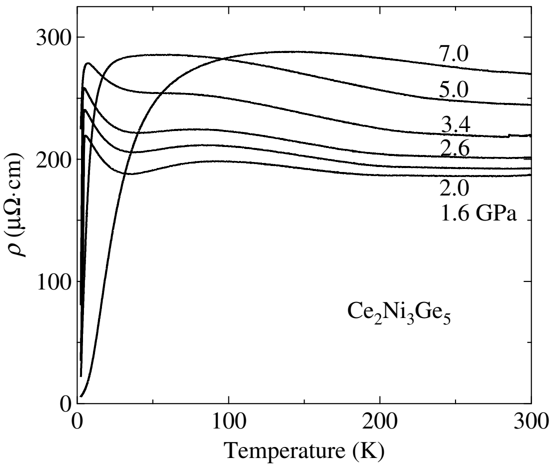

Figure 2. Temperature dependence of the electrical resistivity under pressure in $Ce_2Ni_3Ge_5$, which was obtained by using the cubic anvil cell.

of a stacking of ferromagnetic (010) planes with the (↑↑↓↓) sequence along the [010] direction (*b*-axis) [5]. We measured the electrical resistivity of $Ce_2Ni_3Ge_5$ under pressure and observed superconductivity around 3.9 GPa where the Néel temperature becomes zero.

Polycrystal samples of $Ce_2Ni_3Ge_5$ were prepared by arc-melting stoichiometric amounts of the elements under argon atmosphere. A pellet was wrapped in Ta foil, sealed in an evacuated quartz tube and annealed at 800 °C for one week, as in the recent paper [6]. The sample was checked by means of the x-ray powder diffraction and microprobe analyses. The data were analysed with the Rietveld profile method: $a = 9.815$ Å, $b = 11.843$ Å and $c = 5.962$ Å, which are in good agreement with the previous values [5].

We used two methods for pressure experiments on the electrical resistivity of $Ce_2Ni_3Ge_5$. One was carried out by using a cubic anvil cell at high pressures up to 8 GPa in the temperature range from 2 to 300 K [7] and the other by using an indenter cell in the pressure range from ambient pressure to about 4 GPa in the temperature range from 40 mK to 300 K [4].

Figure 2 shows the temperature dependence of the electrical resistivity under various different pressures ranging from 1.6 to 7.0 GPa, which was measured by using the cubic anvil cell. We note that the present sample indicates slightly larger transition temperatures T_N ($=T_{N1}$) $= 5.2$ K and $T_{N2} = 4.5$ K at ambient pressure than the previous values of $T_N = 4.8$ K and $T_{N2} = 4.2$ K, as mentioned above. The electrical resistivity at 1.6 GPa possesses two peaks around 100 and 5 K, and decreases steeply below T_N ($=T_{N1}$) $= 4.8$ K. A two-peak structure is characteristic in the Kondo compound [4], but is changed into a single peak above 5.0 GPa. A broad peak is seen at 130 K at 7.0 GPa. These characteristic features are similar to those of a pressure-induced superconductor $CeNiGe_3$ [4]. From the present electrical resistivity measurement, we estimated that the Néel temperature becomes zero above 5.0 GPa.

To observe a precise change of the resistivity at low temperatures, we measured the resistivity in the temperature range from 0.6 to 300 K under pressures up to 3.9 GPa by using an indenter cell, as shown in figure 3. The low-temperature resistivity in the indenter cell is slightly different from that in the cubic cell. An upturn of the resistivity below 10 K is rather weak in the indenter cell, compared to that in the cubic cell, although a resistivity drop due to the antiferromagnetic ordering is approximately the same between two measurements. The magnetic transition $T_{N2} = 4.5$ K at ambient pressure disappears under

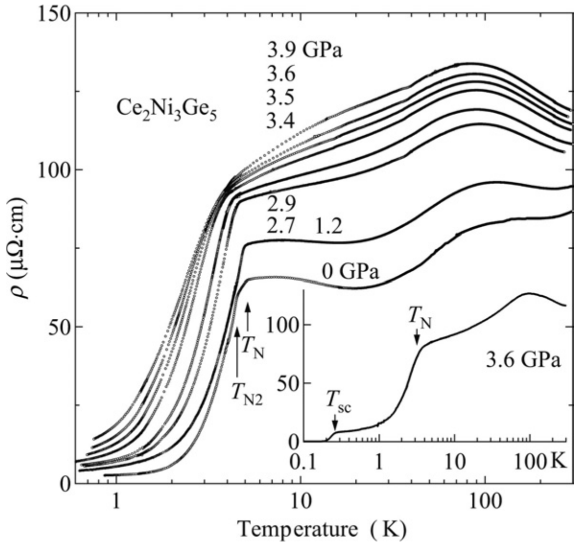

Figure 3. Logarithmic scale of temperature dependence of the electrical resistivity under pressures in $Ce_2Ni_3Ge_5$, which was obtained by using the indenter cell. The inset shows the resistivity result at 3.6 GPa in the temperature region from 300 K to 40 mK, indicating superconductivity below 0.26 K.

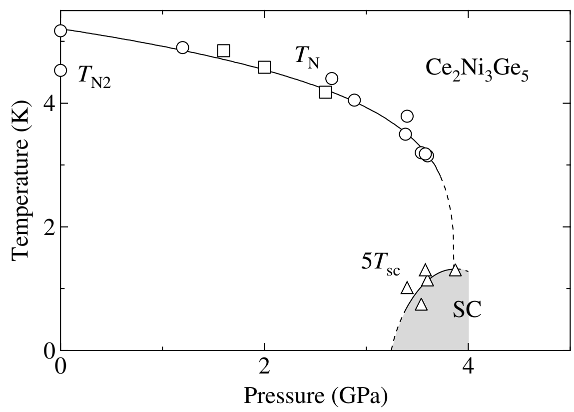

Figure 4. Pressure dependence of a Néel temperature T_N in $Ce_2Ni_3Ge_5$. The data shown by squares and circles were obtained by the cubic anvil and indenter cells, respectively. The superconducting transition temperature T_{sc} shown by triangles is enlarged five times in the temperature scale.

pressure, as shown in figure 3. The inset shows the low-temperature resistivity at 3.6 GPa, indicating superconductivity below $T_{sc} = 0.26$ K, together with the antiferromagnetic ordering at $T_N = 3.2$ K, which will be described later.

Figure 4 shows the pressure dependence of the Néel temperature, where the data obtained by the cubic anvil and indenter cells are shown by squares and circles, respectively. A critical pressure P_c, where the Néel temperature T_N $(=T_{N1})$ becomes zero, is estimated at $P_c \simeq 3.9$ GPa. A solid line is a guideline based on a conventional equation of $T_N(P) = T_N(P = 0)(1 - P/P_c)^n$, where $T_N(0) = 5.2$ K, $P_c(0) = 3.9$ GPa and $n = 0.2$. The superconducting region, which is shown as a shaded region, will be described later.

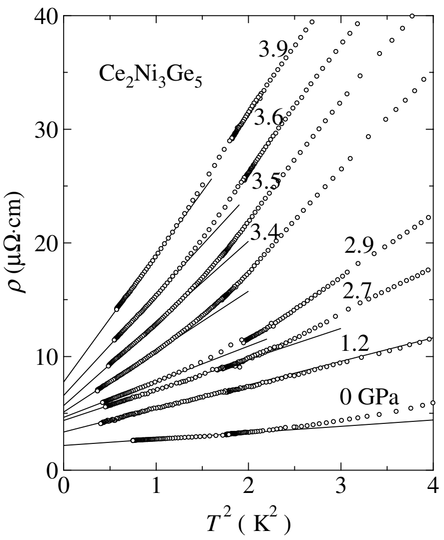

Figure 5. T^2-dependence of the electrical resistivity of $Ce_2Ni_3Ge_5$, obtained by using the indenter cell.

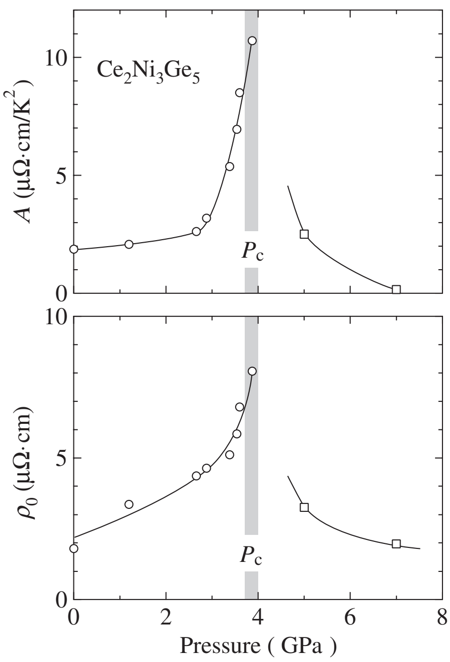

Figure 6. Pressure dependence of A and ρ_0 values in $Ce_2Ni_3Ge_5$. A critical pressure is $P_c \simeq 3.9$ GPa. The data shown by circles and squares were obtained by using the indenter and cubic anvil cells, respectively.

The low-temperature resistivity approximately follows the Fermi liquid relation of $\rho = \rho_0 + AT^2$. Figure 5 shows the T^2-dependence of the resistivity in the pressure range from 0 to 3.9 GPa. A slope of the straight line becomes larger with increasing pressure. Figure 6

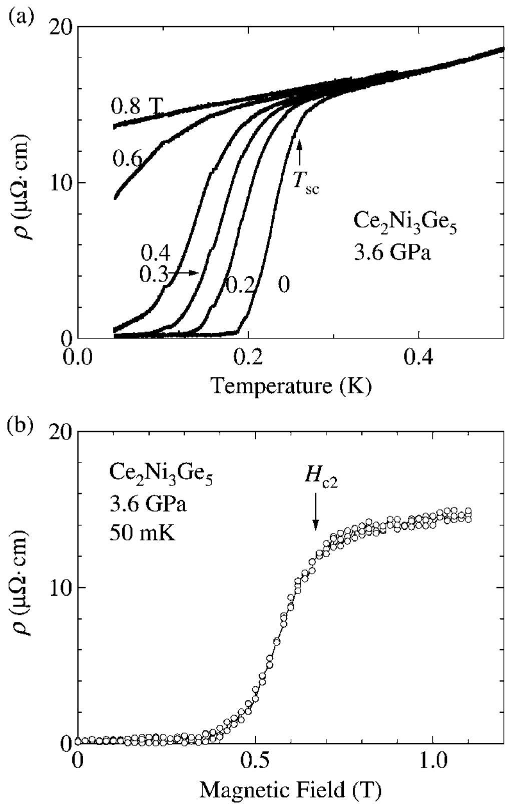

Figure 7. Temperature dependence of the electrical resistivity under several magnetic fields (a) and the field dependence of the resistivity (b) at 3.6 GPa in $Ce_2Ni_3Ge_5$. Arrows indicate the onset of superconductivity.

shows the pressure dependence of A and ρ_0 values. Both the A and ρ_0 values increase steeply above 3 GPa. Here, a value of $A = 10.7\ \mu\Omega$ cm K^{-2} at 3.9 GPa is comparable to $A = 10\ \mu\Omega$ cm K^{-2} in a heavy fermion superconductor $CeCu_2Si_2$ [8]. These results indicate that $Ce_2Ni_3Ge_5$ forms a heavy fermion state around P_c. Moreover, a huge ρ_0-value around P_c is closely related to superconductivity, as discussed by Miyake and Maebashi theoretically on the basis of the experimental result of pressure-induced superconductivity in an antiferromagnet $CeCu_2Ge_2$ [9, 10].

We thus measured the electrical resistivity at lower temperatures down to 40 mK. The insets of figures 3 and 7(a) show the temperature dependence of the resistivity at 3.6 GPa. The electrical resistivity starts to drop below 0.26 K and approaches zero below 0.18 K. With increasing magnetic field, the onset of the resistivity drop shifts to a lower temperature and the resistivity drop is not observed at 0.8 T, as shown in figure 7(a). We define here the superconducting transition temperature T_{sc} as the onset temperature of the resistivity drop, as shown by an arrow in figure 7(a). Figure 7(b) shows the field dependence of the electrical resistivity at 50 mK. The zero resistivity starts to increase steeply above 0.4 T and approximately saturates above 0.8 T. A magnetic field of 0.67 T, shown by an arrow in figure 7(b), is defined as the upper critical field H_{c2}, where the superconducting state is changed into the normal state.

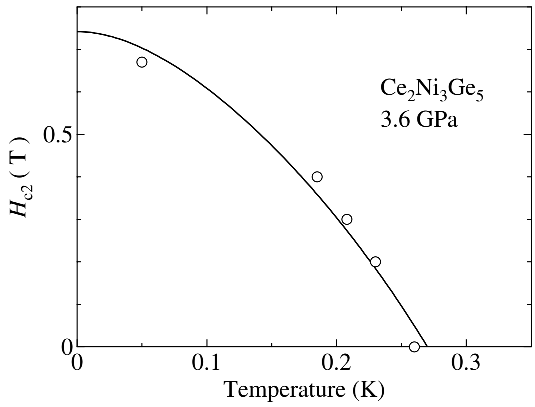

Figure 8. Temperature dependence of the upper critical field H_{c2} at 3.6 GPa in $Ce_2Ni_3Ge_5$. The solid line is a guideline based on the WHH theory.

The present results indicate that the resistivity drop is due to superconductivity. Figure 8 shows the superconducting phase diagram in magnetic fields at 3.6 GPa. A solid line is a guideline based on the WHH theory [11]. The upper critical field in superconductivity is roughly estimated as $H_{c2}(0) \simeq 0.7$ T. Although the superconducting transition temperature $T_{sc} = 0.26$ K is extremely low, a $H_{c2}(0)$ value is extremely large, reflecting a heavy fermion state. The coherence length ξ is estimated as 210 Å from a relation of $H_{c2} = \phi_0/2\pi\xi^2$ (ϕ_0: quantum fluxoid).

Finally we note that superconductivity was observed in the pressure region from 3.4 GPa to $P_c \simeq 3.9$ GPa, as shown in figure 3, which is most likely extended up to about 4.5 GPa. These pressure regions are in the antiferromagnetically ordered state and/or in the antiferromagnetically spin-fluctuated state, where the present superconductivity is realized.

In the present experiment we studied a change of the electronic state as a function of pressure in an antiferromagnet $Ce_2Ni_3Ge_5$ by measuring the electrical resistivity. The Kondo state with magnetic ordering in $Ce_2Ni_3Ge_5$ changes into the non-magnetic state above a critical $P_c \simeq 3.9$ GPa. The heavy fermion state is found to be formed around P_c, in which pressure region superconductivity was observed below 0.26 K.

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