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Pressure effect on the antiferromagnetic compound $Ce_2Ni_3Ge_5$

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In this study, the electrical resistivity and magnetization of a single crystal of $Ce_2Ni_3Ge_5$ heavy fermion compound were performed under pressure. The resistivity and magnetization showed two antiferromagnetic transitions at ambient pressure. On applying pressure, the transitions merged at 1 GPa. At higher pressures, the antiferromagnetic transition temperature decreases, and disappears. It is suggesting that the critical pressure of $Ce_2Ni_3Ge_5$ was 4.1 GPa.
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I. INTRODUCTION

The heavy-fermion material, $Ce_2Ni_3Ge_5$, with an electronic specific heat coefficient of $\gamma \sim 90$ mJ/K^{2}mol Ce, exhibits an orthorhombic structure (space group: I_{bam}) and undergoes two antiferromagnetic (AF) transitions at T_{N1} = 5.0 K and T_{N2} = 4.3 K. The effective magnetic moment μ_{eff} = 2.51μ_B/Ce for $H \parallel$ [100]. This value is comparable to that of free-Ce^{3+} ions (2.54μ_B/Ce).[1] The results of neutron diffraction experiments show that the magnetic structure stabilizes only below T_{N2}. The AF structure shows the moments are oriented and titled 20° from the *a*-axis in the *ab*-plane, with a value of 0.4 μ_B/Ce. The high temperature phase between T_{N1} and T_{N2} is thought to characterize another magnetic phase.[2] In the previous investigation, Nakashima *et al.* measured the electrical resistivity of a polycrystalline sample under pressure and observed pressure-induced superconductivity at T_c = 0.26 K, suggesting that it originates from antiferromagnetic fluctuation.[3] The upper critical field H_{c2} in the superconductivity regime is approximately 0.7 T. Although the superconducting transition temperature T_c = 0.26 K is low, the H_{c2} value is large, indicating a heavy-fermion state. The critical pressure of $Ce_2Ni_3Ge_5$ is 3.9 GPa because the AF transition temperature becomes zero under pressure.

Recently, we were able to perform physical property measurements by cooling down to 10 mK and extend the study on the pressure-induced superconductivity than before.[4] In this study, we measured the electrical resistivity and magnetic susceptibility of single crystal $Ce_2Ni_3Ge_5$ under pressure to obtain the pressure dependences of T_{N1} and T_{N2} at low pressure region and the superconducting properties around the critical pressure.

II. EXPERIMENTAL PROCEDURE

Single crystals of $Ce_2Ni_3Ge_5$ were grown by the Bi-flux method. The details are described in a previous investigation.[1] The magnetic susceptibility was measured by a commercial SQUID

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magnetometer (Quantum Design, MPMS) and a piston-cylinder pressure cell with Daphne oil 7373 as the pressure medium. The applied pressure was determined by the change in the superconducting transition temperature of Pb. The electrical resistivity was measured by a standard AC four-probe method up to 11 GPa. High pressure experiments were performed by using a cubic anvil cell, which is known to generate hydrostatic pressure owing to the multiple-anvil geometry with Fluorinert as the pressure medium. The pressure cell was set to a ^{3}He-^{4}He dilution refrigerator (BlueFors, LD-400) and cooled to 10 mK.

III. RESULT AND DISCUSSION

Figure 1 shows the temperature dependence of the magnetic susceptibility of $Ce_2Ni_3Ge_5$ under several pressures up to 1.12 GPa. The magnetic field was applied parallel to the *c*-axis. At ambient pressure, the susceptibility data show two antiferromagnetic transitions: a second-order transition T_{N1} = 5.0 K and a first-order transition T_{N2} = 4.5 K. With increasing pressure, the T_{N1} and T_{N2} shift to slightly lower and higher temperatures, respectively (indicated by arrows) and eventually merge around 0.97 GPa (inset of Fig. 1). To determine the magnetic transition above 0.97 GPa, the pressure dependence of the magnetic structure must be evaluated.

Figure 2(a) shows the temperature dependence of the electrical resistivity below 4 GPa. The current was applied parallel to the *c*-axis. The peaks around 100 and 5 K, which characterize the Kondo compound, are observed at ambient pressure. The electrical resistivity decreased rapidly below 5.2 K, indicating the AF transition. The AF transition temperatures were T_{N1} = 5.2 K and T_{N2} = 4.3 K. T_{N2} disappeared at 1.5 GPa and T_{N1} continued to decrease under increased pressures (inset of Fig. 2(a)), finally disappearing above 4.25 GPa (Fig. 2(b)). The two-peak structure merged into a single-peak at 7 GPa at 70 K. At higher pressure, the single resistivity peak at 7 GPa shifted to higher temperatures. The pressure dependence of the AF transition temperature is shown in Fig. 3(a). Our data are compared with the previous data on a polycrystalline sample and the conventional equation of $T_{N1}(P) = T_{N1}(P = 0)(1 - P/P_c)^n$, where $T_{N1}(0) = 5.2$K, $P_c(0) = 4.1$ GPa and $n = 0.2$. The critical pressure P_c, at which the AF transition temperature becomes zero, was estimated to be around $P_c \sim$ 4.1 GPa. When another value n was used, the fitting was not good. Thus, it suggests that $n = 0.2$ is suitable value. In the previous study, they fit by same value $n = 0.2$ and concluded that these pressure region in the antiferromagnetically spin-fluctuation state,[3] where the present superconductivity is realized. These results consistent with our present study. However, the superconducting transition at

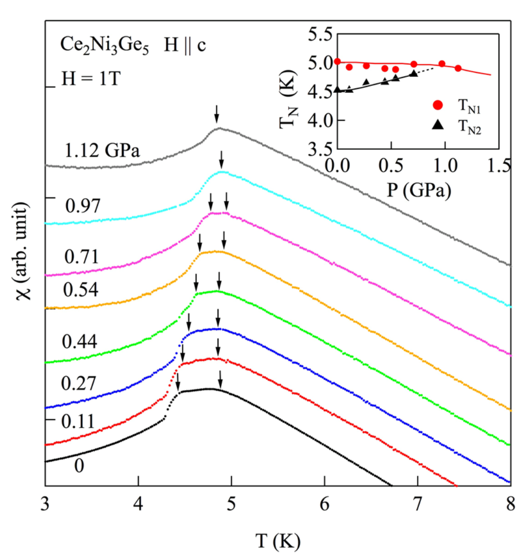

FIG. 1. Temperature dependences of magnetic susceptibility of $Ce_2Ni_3Ge_5$ under various pressures in a magnetic field H = 1T. Arrows indicate the antiferromagnetic transitions. Inset shows the T_{N1} and T_{N2} versus pressure phase diagram.

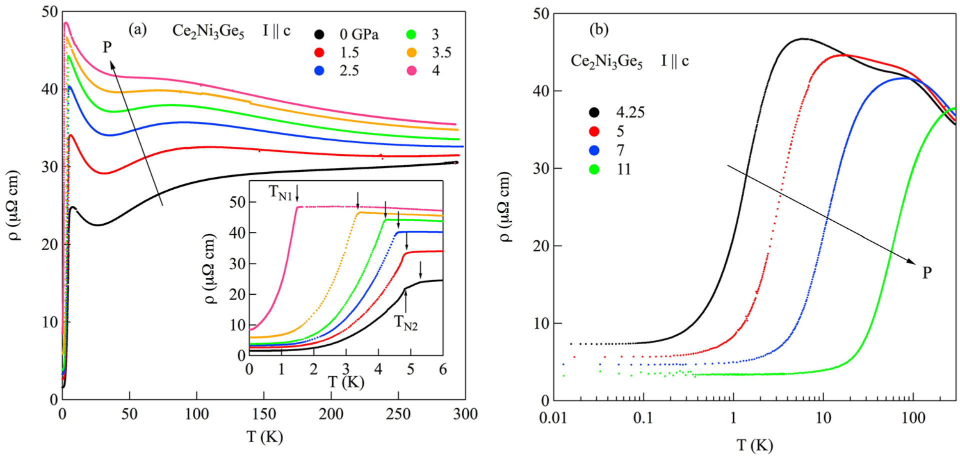

FIG. 2. (a) Temperature dependences of electrical resistivity of $Ce_2Ni_3Ge_5$ below 4 GPa. Inset: Magnified view at low temperatures. (b) Temperature dependences of electrical resistivity of $Ce_2Ni_3Ge_5$ above 4.25 GPa.

the lowest temperature (closed triangles in Fig. 3(a)) could not be observed in the present study. The causes of this deficiently will be discussed later.

To determine the critical pressure of single crystal $Ce_2Ni_3Ge_5$, we estimated the A-coefficient and residual resistivity ρ_0 from the electrical resistivity measurements. The pressure dependences of both variables are plotted in Fig. 3(b). The low temperature resistivity approximately follows the Fermi liquid relation $\rho = \rho_0 + AT^2$. Both values increased with increasing pressure and were maximized around 4 GPa. We obtained $A = 19.5\ \mu\Omega\,\mathrm{cm/K^2}$ and $\rho_0 = 7.3\ \mu\Omega\,\mathrm{cm}$ at 4 GPa, indicating the heavy-fermion state. However, the resistivity was non-zero pressure over the entire region (Fig. 3(a)). The A-coefficient is larger than the previous result. This may be the difference of the fitting range at low temperature. We consider that non-Fermi liquid region is very narrow. It is suggesting a first-order transition from AF to paramagnetism, but this judgment is tentative just by our resistivity measurements. The further study is needed, and the work is in progress now.

The residual resistivity ratio was about 20 for the single crystal and 40 for the polycrystalline sample, suggesting that $Ce_2Ni_3Ge_5$ is sensitive to sample quality. Therefore, more high quality samples are required for investigating the superconducting properties of $Ce_2Ni_3Ge_5$.

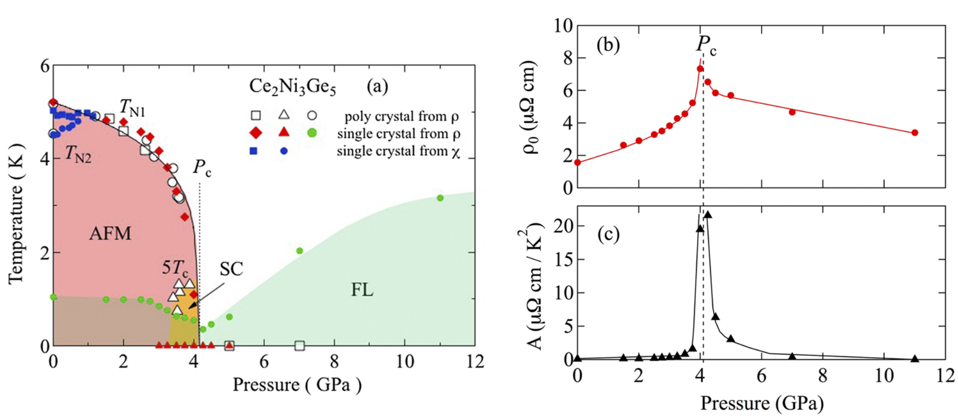

FIG. 3. (a) Temperature-pressure phase diagram of $Ce_2Ni_3Ge_5$. Open and closed symbols plot the previously reported data concerning a polycrystalline sample and the present data concerning the single crystal, respectively. Closed red diamonds, closed red triangle, and closed green circle symbols were obtained from the electrical resistivity data, and the blue circles and squares were obtained from the magnetic susceptibility. AFM, FL, and SC indicate antiferromagnetism, Fermi liquid, and superconductivity, respectively. The dashed line indicates a critical pressure in the present study. The solid line is in the plot of the conventional equation $T_{N1}(P) = T_{N1}(P=0)(1 - P/P_c)^n$. (b), (c) Pressure dependences of the residual resistivity ρ_0 and A-coefficient. The solid lines are a visual guide. The dashed line indicates the critical pressure P_c of $Ce_2Ni_3Ge_5$.

IV. SUMMARY

In Summary, we investigated the magnetic susceptibility and electrical resistivity of single crystals of $Ce_2Ni_3Ge_5$ under various pressures. In the low pressure region, T_{N1} and T_{N2} slowly converge until they merge around 1 GPa. The AF transition temperature disappeared above 4.25 GPa. The coefficient A and residual resistivity ρ_0 are found to be maximum at 4 GPa, suggesting that the critical pressure of $Ce_2Ni_3Ge_5$ is 4 GPa. These results are consistent with the previously observed pressure effects on polycrystalline samples of $Ce_2Ni_3Ge_5$. Nevertheless, the sample quality was insufficient for observing the superconducting transition. More experiments need to be performed to understand the superconducting properties of $Ce_2Ni_3Ge_5$.

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