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LETTER TO THE EDITOR

Pressure study of an antiferromagnet, $CeMg_2Cu_9$

H Nakawaki[1], Y Inada[1,4], R Asai[1], M Yamada[1], T Okubo[1], S Ikeda[1], A Thamizhavel[1], T C Kobayashi[2], R Settai[1], E Yamamoto[3] and Y Ōnuki[1,3]

[1] Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan
[2] Research Centre for Materials Science at Extreme Conditions, Osaka University, Toyonaka, Osaka 560-8531, Japan
[3] Advanced Science Research Centre, Japan Atomic Energy Research Institute, Tokai, Ibaraki 319-1195, Japan

E-mail: inada@phys.sci.osaka-u.ac.jp

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Abstract

We have studied the effect of pressure on the electrical resistivity of the antiferromagnet $CeMg_2Cu_9$ which crystallizes in the hexagonal structure. The structure is built up of alternating $MgCu_2$ Laves-type and $CeCu_5$-type layers along the [0001] direction. The Néel temperature $T_N = 2.7$ K at ambient pressure decreases with increasing pressure p and disappears at a critical pressure $p_c \simeq 2.5$ GPa. Correspondingly, the residual resistivity ρ_0 and the coefficient A in a Fermi-liquid relation $\rho = \rho_0 + AT^2$ are found to have maximum values around p_c.

In cerium compounds, the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction and the Kondo effect compete with each other [1, 2]. The former enhances long-range magnetic ordering, in which the 4f electrons with magnetic moments are treated as localized electrons and the indirect f–f interaction plays a predominant role via the conduction electrons. On the other hand, the latter quenches the magnetic moments of the localized 4f electrons via spin polarization of conduction electrons, consequently producing the singlet state with a binding energy $k_B T_K$, where T_K is called the Kondo temperature. Competition between the RKKY interaction and the Kondo effect was discussed by Doniach [3], as a function of $|J_{cf}|D(\varepsilon_F)$, where $|J_{cf}|$ is the magnitude of the magnetic exchange interaction and $D(\varepsilon_F)$ is the electronic density of states at the Fermi energy ε_F.

Most cerium compounds order magnetically, when the RKKY interaction overcomes the Kondo effect at low temperatures. The magnetic order is formed by localized 4f moments. On the other hand, some cerium compounds such as $CeCu_6$ and $CeRu_2Si_2$ show no long-range magnetic ordering, because the Kondo effect overcomes the RKKY interaction. Characteristic properties of these compounds are called heavy-fermion properties, with a large electronic

[4] Author to whom any correspondence should be addressed.

specific heat coefficient γ: $\gamma \simeq 10^4/T_K$ (mJ K^{-2} mol^{-1}) [1,2]. In fact, the γ-value and the Kondo temperature are 1600 mJ K^{-2} mol^{-1} and 5 K in $CeCu_6$, and 350 mJ K^{-2} mol^{-1} and 20 K in $CeRu_2Si_2$. The heavy-fermion state in these compounds can be understood on the basis of Fermi-liquid behaviour. The electrical resistivity ρ follows a well-known relation: $\rho = \rho_0 + AT^2$. The $\sqrt{A}$ value is extremely large; this is correlated with an enhanced Pauli susceptibility $\chi \simeq \chi_0$ ($T \to 0$) and a large γ-value ($=C/T$: $T \to 0$).

Recently a new aspect of cerium and uranium compounds with magnetic ordering has been discovered. When pressure p is applied to these compounds with antiferromagnetic ordering such as $CeIn_3$ and $CePd_2Si_2$ [4], the Néel temperature T_N decreases, and a quantum critical point corresponding to the extrapolation $T_N \to 0$ is reached at $p = p_c$. Here, $|J_{cf}|D(\varepsilon_F)$ in the Doniach model can be replaced by pressure. Surprisingly, superconductivity appears around p_c. A non-Fermi-liquid nature of these compounds is observed. Similar pressure-induced superconductivity was reported for other compounds such as $CeCu_2Ge_2$ [5], $CeRhIn_5$ [6] and UGe_2 [7]. The crossover from the magnetically ordered state to the non-magnetic state under pressure, crossing the quantum critical point, is the most interesting issue for the strongly correlated f-electron systems.

The purpose of present work is to study a similar pressure effect on another compound, $CeMg_2Cu_9$. This compound is similar to $CeMg_2Ni_9$, with a hexagonal structure which is built up of alternating $MgNi_2$ Laves-type and $CeNi_5$-type layers along the [0001] direction [8, 9]. The lattice parameter c (=23.846 Å) is extremely large compared to the a-value of 4.8706 Å in $CeMg_2Ni_9$. $CeMg_2Cu_9$ is also hexagonal and it was recently clarified that it is of $CeNi_3$ type [10]. $CeMg_2Cu_9$ orders antiferromagnetically below $T_N = 2.7$ K, whereas $CeMg_2Ni_9$ is a non-magnetic compound. The large lattice parameter c in $CeMg_2Ni_9$ or $CeMg_2Cu_9$ corresponds to a flat Brillouin zone, which is expected to bring about a two-dimensional electronic state as in $CeRhIn_5$ [6]. We have studied the effect of pressure by measuring the electrical resistivity under hydrostatic pressure p up to about 3 GPa in the temperature range from room temperature to 90 mK. The Néel temperature T_N is found to disappear at $p_c \simeq 2.5$ GPa.

Single crystals of $CeMg_2Cu_9$ were grown by the slow-cooling method. Highly pure materials of 3N (99.9% pure) Ce, 5N Mg, and 5N Cu with a starting composition of $CeMg_{2.05}Cu_9$ were sealed in a Mo crucible, which was heated up to 1400 °C and cooled down slowly under an Ar atmosphere in an electric furnace; it took ten days in total to complete the growth. The direction of the crystal with the hexagonal structure was determined by the x-ray Laue method.

The electrical resistivity was measured by the four-probe DC method in an indenter pressure cell, with Daphne oil (7373) as a pressure-transmitting medium, which was cooled down to 90 mK in a dilution refrigerator. The pressure value was determined from the superconducting transition temperature of lead.

Figure 1 shows the temperature dependence of the electrical resistivity ρ, magnetic susceptibility χ, and specific heat C in the form of C/T. The current I was applied along the (0001) plane or the c-plane. The resistivity shows a broad maximum around 50 K and decreases sharply below $T_N = 2.7$ K. Correspondingly, the magnetic susceptibility decreases slightly below T_N, and the specific heat shows a λ-like peak at T_N. The Néel temperature $T_N = 2.7$ K and the electronic specific heat coefficient $\gamma = 160$ mJ K^{-2} mol^{-1}, which is estimated by extrapolation to $T = 0$ K in the ordered state, are the same as recent results [10]. The effective magnetic moment $\mu_{eff} = 2.43$ μ_B/Ce for $H \parallel [10\bar{1}0]$ in the Curie–Weiss law is close to 2.54 μ_B of Ce^{3+}, and the paramagnetic Curie temperature $\theta_p = -14$ K is negative. The values of μ_{eff} and θ_p are 2.24 μ_B/Ce and -8.3 K for $H \parallel [11\bar{2}0]$, and 2.31 μ_B/Ce and -11 K for $H \parallel [0001]$. The magnetization for three field directions indicates a metamagnetic transition around 40 kOe. The inset in figure 1(b) shows the magnetization curve for $H \parallel [10\bar{1}0]$.

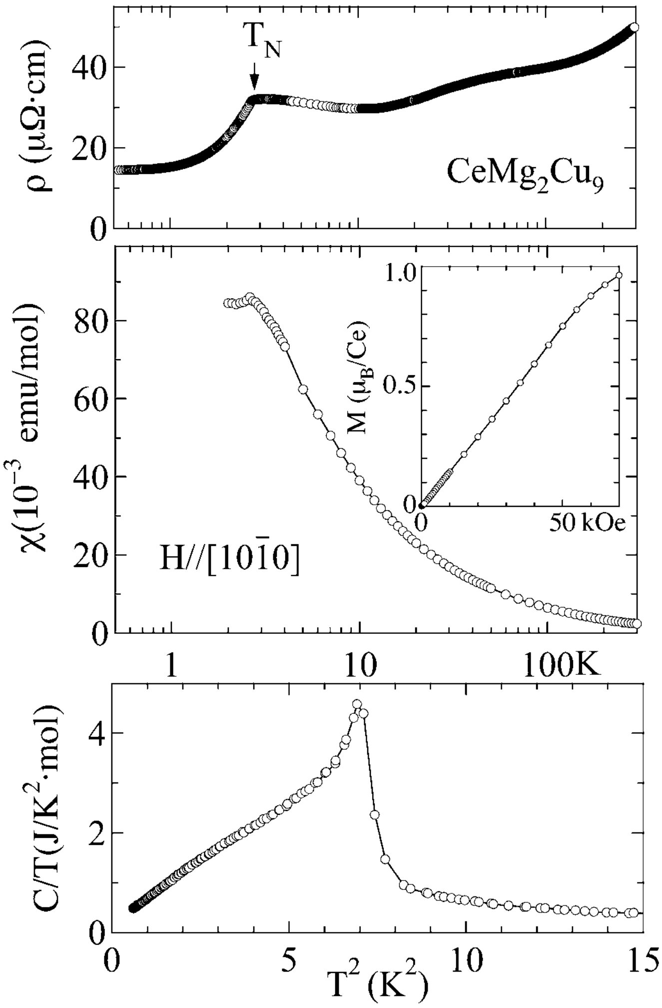

Figure 1. The temperature dependence of (*a*) the electrical resistivity, (*b*) the magnetic susceptibility, and (*c*) the specific heat C in the form of C/T in $CeMg_2Cu_9$. The inset in (*b*) shows the magnetization curve for $H \parallel [10\bar{1}0]$.

The value of the magnetization for this direction at 70 kOe is slightly larger than those for other directions; therefore this direction is most probably the easy axis of antiferromagnetism.

The resistivity maximum around 50 K is due to a combination of the Kondo effect and the crystalline-electric-field effect. This compound is similar to the usual Kondo antiferromagnets, such as $CeCu_2$, with $T_N = 3.5$ K and $\gamma = 82$ mJ K^{-2} mol [11]. We focus our interest on the pressure effect around the critical pressure p_c where the Néel temperature becomes zero, although the magnetic structure and the two-dimensional electronic state, based on the unique crystal structure, are also of interest.

Figure 2 shows the temperature dependence of the electrical resistivity under different applied pressures. The resistivity maximum around 50 K at ambient pressure, mentioned above, shifts to a lower temperature with increasing pressure up to 2.38 GPa, while the maximum shifts to a higher temperature with further increasing pressure. This is closely related to a steep decrease of the Néel temperature due to pressure, as shown in figures 3 and 4. The Néel

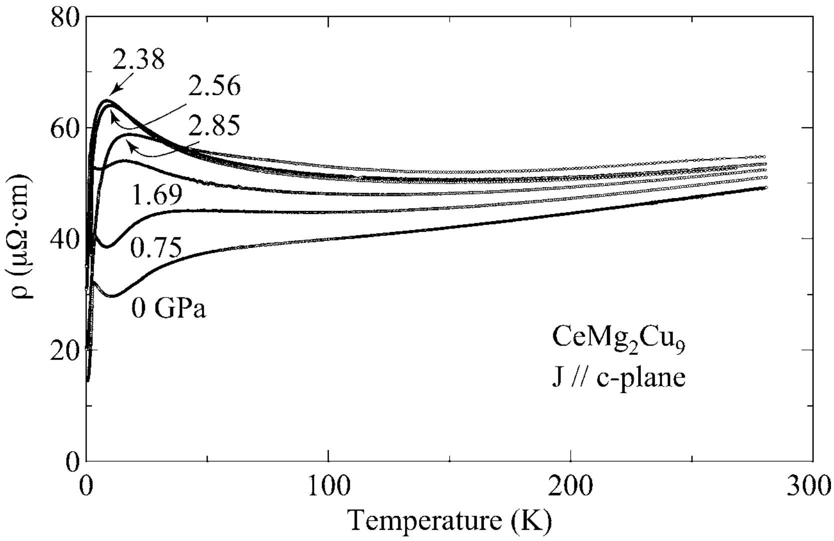

Figure 2. The temperature dependence of the electrical resistivity under pressure in $CeMg_2Cu_9$.

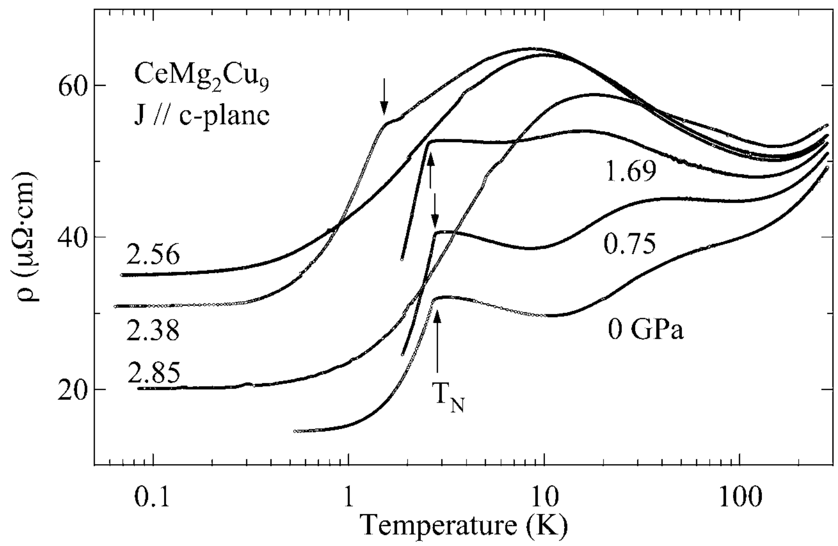

Figure 3. A logarithmic plot of the temperature dependence of the electrical resistivity under pressure in $CeMg_2Cu_9$.

temperature $T_N = 2.7$ K at ambient pressure decreases with increasing pressure: $T_N = 1.5$ K at $p = 2.38$ GPa and becomes zero at 2.56 GPa. The critical pressure p_c is about 2.5 GPa. With further increasing pressure, the resistivity maximum increases from 10 K at 2.56 GPa to 20 K at 2.85 GPa. This is because the Kondo temperature increases with increasing pressure. The temperature dependence of the electrical resistivity under 2.85 GPa is similar to that for a typical heavy-fermion compound $CeCu_6$ [12].

The low-temperature resistivity follows the Fermi-liquid relation of $\rho = \rho_0 + AT^2$, as shown in figure 5. The resistivity for each pressure in figure 5 is shifted on the vertical scale. Figure 6 shows the pressure dependence of the coefficient A and the residual

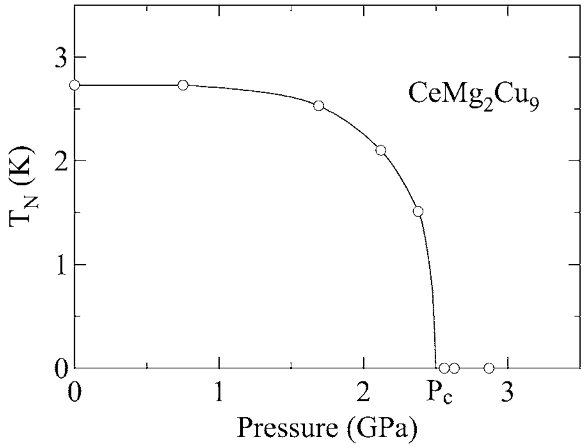

Figure 4. The pressure dependence of the Néel temperature in $CeMg_2Cu_9$.

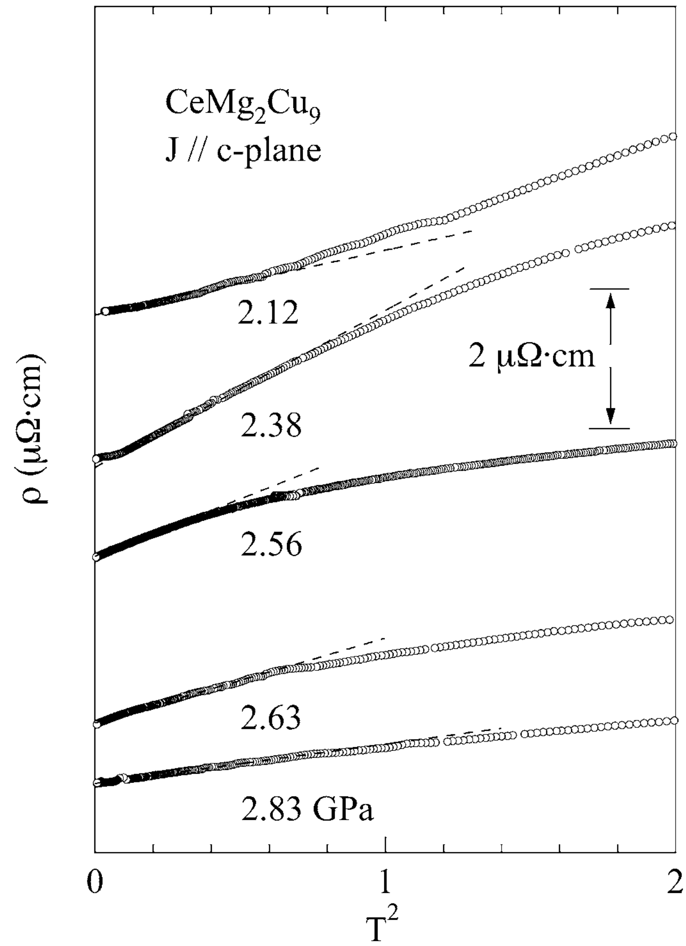

Figure 5. The T^2-dependence of the electrical resistivity under pressure in $CeMg_2Cu_9$. Broken lines are guides to the eyes.

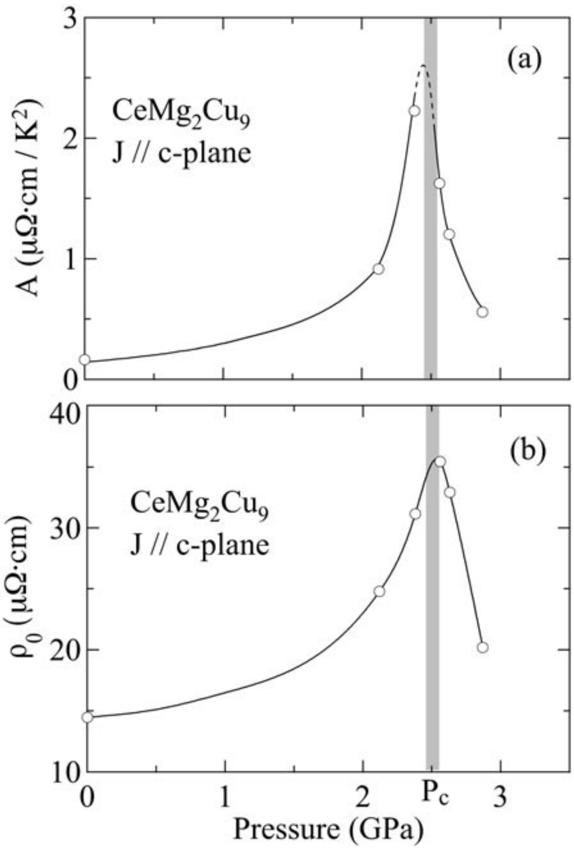

Figure 6. Pressure dependences of (*a*) A and (*b*) ρ_0 for $CeMg_2Cu_9$.

resistivity ρ_0. The Néel temperature becomes zero at $p_c \simeq 2.5$ GPa, as mentioned above. Correspondingly, the A- and ρ_0-values become maximal around $p_c \simeq 2.5$ GPa. The A-value of 2.5 $\mu\Omega$ cm K^{-2} at $p_c \simeq 2.5$ GPa is large, as compared to $A = 50\ \mu\Omega$ cm K^{-2} for $CeCu_6$ with $\gamma = 1600$ mJ K^{-2} mol^{-1} and $A = 1\ \mu\Omega$ cm K^{-2} for UPt_3 with $\gamma = 420$ mJ K^{-2} mol^{-1}, following the Kadowaki–Woods plot [13].

Around the critical pressure, non-Fermi-liquid behaviour and/or superconductivity were observed in $CeCu_2Ge_2$ [5] and $CeRhIn_5$ [6, 14], as described above. Superconductivity often correlates with non-Fermi-liquid nature, as for $CeCu_2Ge_2$ and $CeRhIn_5$. The T-linear dependence of the electrical resistivity in $\rho = \rho_0 + A'T$ is characteristic of these compounds. The residual resistivity ρ_0 also correlates with the superconducting transition temperature T_c, as discussed theoretically [15, 16].

On the other hand, the present electrical resistivity in $CeMg_2Cu_9$ exhibits a Fermi-liquid nature. It is not clear whether this is closely related to the non-appearance of superconductivity. We note that superconductivity is found in the Fermi-liquid state for $CeRh_2Si_2$ [17, 18] and UGe_2 [19]. It is also noted that the present large residual resistivity of $\rho_0 = 14\ \mu\Omega$ cm at ambient pressure might enhance pair breaking in the superconductivity. In fact, we confirmed that the sample of $CeRh_2Si_2$ with residual resistivity ratio (RRR) less than 30 showed no superconductivity, whereas superconductivity was clearly observed in the sample with RRR = 110 [18].

There is, however, another example where superconductivity is found in $CeCu_2Ge_2$ with a large residual resistivity at ambient pressure [5]—almost the same value as for $CeMg_2Cu_9$. The appearance of superconductivity is thus a very complicated phenomenon in the strongly correlated f-electron systems. Future study of superconductivity in $CeMg_2Cu_9$ using a much higher-quality sample would be beneficial.

We now summarize our experimental results. The antiferromagnet $CeMg_2Cu_9$ was investigated by means of electrical resistivity measurements under hydrostatic pressure. The Néel temperature decreases with increasing pressure, and the critical pressure is determined as $p_c \simeq 2.5$ GPa. Both the A- and ρ_0-values of $\rho = \rho_0 + AT^2$ become maximal around p_c. It is remarkable that the antiferromagnet $CeMg_2Cu_9$ is changed into a typical heavy-fermion compound around p_c.

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