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Magnetic-to-nonmagnetic transition in the ferromagnetic heavy fermion compound $CeRu_2Ge_2$ at high pressures

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A zero-temperature magnetic-to-nonmagnetic transition has been investigated in the heavy-fermion ferromagnet $CeRu_2Ge_2$ from electrical-resistance measurements at pressures up to 15 GPa and temperatures down to 60 mK. We found that the magnetic-to-nonmagnetic phase boundary exists at around $P_c \sim 10$ GPa. The resistivity shows the quadratic temperature variation expected for the conventional Fermi liquid at low temperatures near P_c. In the present pressure range, neither the metamagnetic nor the superconducting transition was observed at least down to 60 mK. [S0163-1829(98)01210-7]

An application of pressure can induce the zero-temperature magnetic-to-nonmagnetic transition in heavy-fermion systems. Deviation from the canonical Fermi liquid behavior near the phase boundary is reported in the measurements of the temperature dependence of the resistivity and the specific heat.[1–4] Most of superconductivity (SC) in heavy-fermion systems is discovered with the ubiquitous signatures of the heavy Fermi liquid state. By contrast, it is reported that the pressure-induced superconducting transition discovered in $CePd_2Si_2$ occurs from an *anomalous* normal state which has a quasilinear temperature variation in resistivity.[3] Unlike the quadratic form expected for a conventional Fermi liquid found in other heavy-fermion superconductors, it bears a resemblance to the form observed in some of the high-T_c cuprate superconductors.

It has been established that non-Fermi-liquid behavior is relevant to the closeness of an antiferromagnetic (AF) quantum critical point (QCP) in $CeCu_{6-x}Au_x$,[1] $CeCu_2Si_2$,[2] and $CeNi_2Ge_2$.[2] A quantitative accordance is obtained between the experiment in $Ce_{1-x}La_xRu_2Si_2$ (Ref. 5) and the calculation based on the self-consistent renormalization (SCR) spin fluctuation theory.[6] On the other hand, in $CeRh_2Si_2$ where the AF QCP is accessed by application of pressure or the substitution of Rh for Ru in $CeRh_{2-x}Ru_xSi_2$ with $x=1$, a comparison of specific heat and resistivity measurements emphasized the importance of disorder in producing a non-Fermi-liquid behavior.[7] In order to obtain further insights into the non-Fermi-liquid effects near the QCP and its relevance to the onset of the SC transition, we need further systematic studies in the heavy-fermion magnets.

The physical properties of a series of CeM_2X_2 with the tetragonal $ThCr_2Si_2$ structure differ in the number of d electrons in the transition metal constituent M and their unit cell volume. It is reported that most of the compounds with a larger unit cell volume than $V_c \sim 170 Å^3$ are magnetically ordered. In the heavy-fermion helimagnet $CeCu_2Ge_2(177.7 Å^3)$, an application of pressure induced SC at $T_c \sim 0.6$ K in pressures exceeding 7.6 GPa.[8,9] It is striking that the physical properties of $CeCu_2Ge_2$ at 7.6 GPa are quite analogous to those in $CeCu_2Si_2$ at ambient pressure.[10] Due to the same unit cell volume and d electron numbers, both exhibited SC with almost the same $T_c = 0.7$ K and an identical temperature dependence in the normal-state resistivity. This result suggests that the substitution of all Ge for Si acts as a chemical pressure for the compounds with the same d metal (Cu) constituent.

$CeRu_2Ge_2(183 Å^3)$ is ferromagnetically ordered at temperatures below 8 K.[11] Though the unit cell volume of $CeRu_2Si_2(171.9\ Å^3)$ is larger than V_c, it remains in the paramagnetic state down to 20 mK without showing magnetic long-range order or SC. However, $CeRu_2Si_2$ shows the metamagnetic transition under the magnetic field of $H_c \sim 8$ T.[12] The high-pressure study of $CeRu_2Si_2$ has not given any sign of SC up to 10.5 GPa.[13] The effect of chemical pressure in $CeRu_2(Si_{1-x}Ge_x)_2$ was extensively investigated by heat capacity, resistivity and magnetization

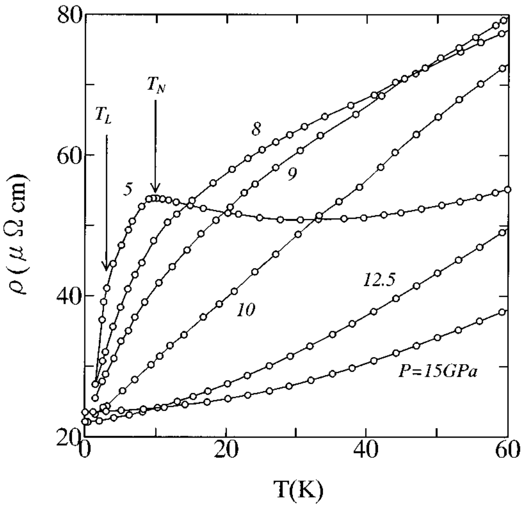

FIG. 1. Temperature dependence of the resistivity of $CeRu_2Ge_2$ in the pressure range of 5–15 GPa.

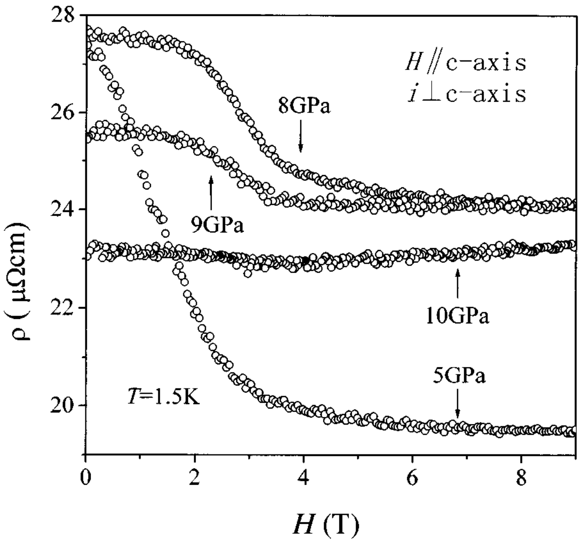

FIG. 2. Magnetoresistance under high pressure with the magnetic field parallel to the c axis for $CeRu_2Ge_2$.

measurements.[11,14] It is reported that ferromagnetic ordering persists for $x > 0.8$, whereas AF ordering appears with a maximum value of $T_N = 10$ K for $x = 0.6$ as Si content, namely, the chemical pressure, increases. An unidentified magnetic anomaly appeared at temperatures well below T_N. It is not clear how extensive the disorder affects the complicated magnetic properties in these compounds.

Like, the $CeCu_2X_2$ series (X = Si and Ge), application of pressure on $CeRu_2Ge_2$ allows us to access the evolution of the electronic state in $CeRu_2Si_2$ at ambient pressure and under negative pressure where its unit cell volume would be expanded. Furthermore, the QCP affected by ferromagnetic spin fluctuations is possible to be compared with the AF QCP in other Ce-based compounds mentioned above. Motivated by these, we carried out high-pressure measurements on single crystals of $CeRu_2Ge_2$ in order to investigate the nature near the QCP of the system with a ferromagnetic ground state and search for the possibility of pressure-induced SC without introducing any disorder by the chemical substitution.

Since the unit cell volume ($V = 183$ Å) of $CeRu_2Ge_2$ is relatively larger than the compounds mentioned above, a study by the conventional piston-and-cylinder cells is not available, inducing a zero-temperature magnetic-to-nonmagnetic transition. Alternatively, a clump-type diamond-anvil cell (DAC) was employed. Details of experimental procedures are given elsewhere.[15] Al_2O_3 fine powder was used for electrical insulation between the metal gasket and electrodes. A small single crystal with a dimension of $100 \times 70 \times 10$ μm^3, which is smaller than the pressure surface of the diamond anvil with 750 μm in diameter, was used in order to reduce the pressure distribution. The pressure value is determined by the ruby fluorescence method at both room temperature and 77 K. The thermal variation of pressure in the cooling process from room temperature to 77 K is less than 1 GPa. The resistivity measurement was performed by the four-probe ac method with a measuring current of $I = 100$ μA.

Figure 1 shows the typical temperature dependence of resistivity $\rho(T)$ of a $CeRu_2Ge_2$ single crystal obtained in the course of increasing pressure. In the low-pressure region less than 6 GPa, the behavior of $\rho(T)$ revealing a steep drop below 10 K is associated with the onset of AF ordering as reported already in experiments at pressures smaller than 2 GPa (Ref. 16) and experiments in $CeRu_2(Si_{1-x}Ge_x)_2$.[14] These experiments point out that the ferromagnetic transition T_C is easily suppressed by applying pressure or by the substitution of Ge for Si. At around 5 GPa, another magnetic anomaly appears at $T_L \sim 3$K, which is similar to that observed in $CeRu_2(Si_{1-x}Ge_x)_2$ with $x = 0.2$, 0.4, and 0.6. This result suggests that the disorder does not affect the complicated magnetic properties in these compounds. In the intermediate range from 7 to 9 GPa, $\rho(T)$ decreases gradually with an upward curvature. At around 10 GPa, $\rho(T)$ has a quasilinear dependence up to 50 K. In the high-pressure region exceeding 10 GPa, $\rho(T)$ approaches the quadratic temperature variation expected for a conventional Fermi liquid.

It should be noted that $\rho(T)$ at 9 GPa is similar to that in $CeRu_2Si_2$ at ambient pressure, but the magnetoresistance shown in Fig. 2 decreases at 8 and 9 GPa with increasing magnetic field, bearing some resemblance to the behavior at 5 GPa at which the magnetic ordering persists. A similar behavior to the negative magnetoresistance was reported in the antiferromagnetic $CeRu_2(Si_{1-x}Ge_x)_2$ with $x = 0.4$,0.6 as well.[14] On the other hand, the magnetoresistance in $CeRu_2Si_2$ has maximum around the metamagneticlike transition at $H_c \sim$ 8 T.[12] Therefore, $CeRu_2Ge_2$ under a pressure of 8 and 9 GPa is expected to possess a magnetically ordered ground state. The disappearance of negative magnetoresistance at 10 GPa signals the closeness of the QCP, where no maximum associated with the metamagnetic transition is observed.

To see the evolution of the power law behavior in $\rho(T)$, $\Delta\rho(T) = \rho(T) - \rho(0)$ is presented as a full logarithmic plot in Fig. 3 in the case of a pressure of 10, 12.5, and 15 GPa. At 10 GPa near the QCP, $\rho(T)$ has a quasilinear form at high temperatures but $\Delta\rho(T)$ shows the Fermi liquid behavior of the T^2 law at temperatures below 2 K. At 12.5 GPa, the temperature region where $\Delta\rho(T)$ follows the T^2 law increases slightly while the coefficient of the T^2 term increases

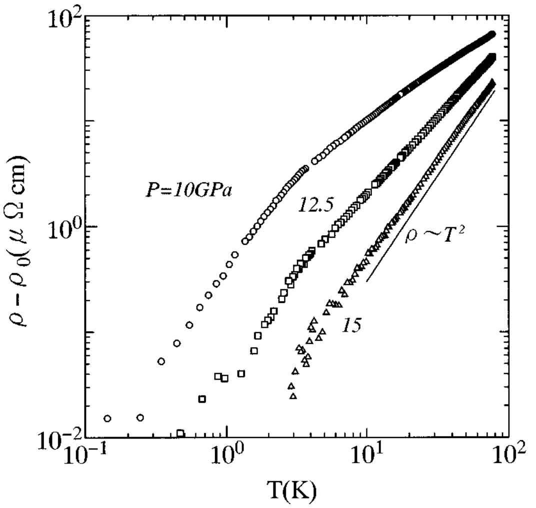

FIG. 3. Power law behavior of $\Delta\rho(T)$ under a pressure of 10, 12.5, and 15 GPa.

drastically. At the highest pressure of 15 GPa, a Fermi liquid state is clearly induced with evidence of a quadratic variation in the wide temperature range up to 50 K. In the high-temperature region above 3 K, $\Delta\rho(T)$ at 10, 12.5, and 15 GPa show the power law behavior, where the values of the power are estimated as 1, 1.5, and 2, respectively. In the present experiment, we cannot observe non-Fermi-liquid behavior at lowest temperature region near the QCP.

Though we carried out the measurements down to low temperatures to search the pressure-induced SC, no sign of SC has been obtained in the present experimental conditions of temperature down to 60 mK and pressure up to 15 GPa. In order to check the reliability of our experiments under such extreme multiple conditions, we reexamined the transport property in $\mathrm{CeCu_2Ge_2}$ up to 20 GPa and down to 100 mK. Successful reconfirmation of pressure-induced SC in the range from 7.6 to 20 GPa for $\mathrm{CeCu_2Ge_2}$ makes its absence in $\mathrm{CeRu_2Ge_2}$ quite reliable.[9] However, it is not completely excluded that the poor hydrostatic condition in such a high-pressure experiment would be responsible for a possible absence of SC.

In Fig. 4, the evolution of the ground state in $\mathrm{CeRu_2Ge_2}$ is summarized as a function of pressure. The ferromagnetic state is easily suppressed in the low-pressure regime less than 2 GPa as is already reported in the literature,[16] whereas the Néel temperature T_N is rather enhanced with an increase of pressure. At around 5 GPa, another magnetic transition appears. Although the temperature variations of the resistivity at 8 and 9 GPa are similar to that in $\mathrm{CeRu_2Si_2}$ at ambient pressure, their magnetic field variations are rather analogous to those for the AF-ordered state. In this region, we cannot define both T_N and T_L. Above 10 GPa, a quadratic temperature variation expected for the conventional Fermi liquid is observed in the resistivity. We indicate the region where $\rho(T)$ follows the T^2 dependence in Fig. 4.

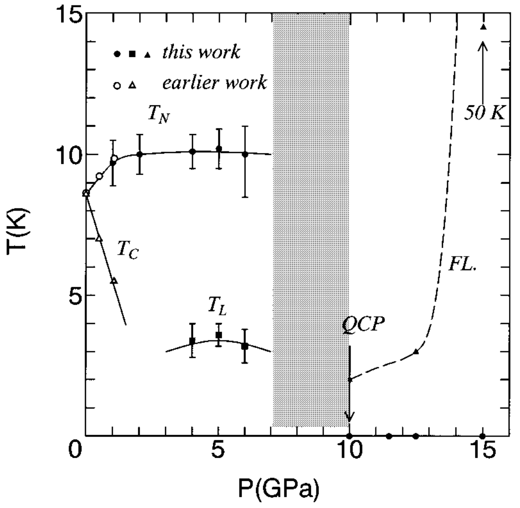

FIG. 4. Phase diagram in $\mathrm{CeRu_2Ge_2}$ under high pressure. T_N and T_C are the antiferromagnetic and ferromagnetic transition temperatures, respectively. T_L is another magnetic transition. At 8 and 9 GPa, T_N and T_L cannot be defined.

In conclusion, the resistivity measurements under pressure have thus clarified the evolution of the ground state in a single crystal of $\mathrm{CeRu_2Ge_2}$ from the ferromagnetic to antiferromagnetic one and through the QCP to the heavy-fermion state. Near the QCP, we observed a quadratic temperature variation expected for a conventional Fermi liquid below 2 K. Through the resistivity measurements of the single crystal of $\mathrm{CeRu_2Ge_2}$ under pressures up to 15 GPa, superconductivity has not been found at temperatures down to $T = 60$ mK near criticality. The magnetoresistance has not provided a signature of the anomaly associated with the metamagnetic transition observed in $\mathrm{CeRu_2Si_2}$.

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