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LETTER TO THE EDITOR

Non-Fermi-liquid behaviour around the magnetic quantum critical point in UGa_3

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Abstract

We studied the electronic state of UGa_3 by measuring the electrical resistivity under pressure. When pressure is applied to UGa_3, the Néel temperature $T_N = 67$ K decreases steeply and becomes zero around 2.6 GPa. Correspondingly, the coefficient A in the electrical resistivity $\rho = \rho_0 + AT^2$, which is characteristic for Fermi liquid, increases steeply with increasing pressure. Around a magnetic quantum critical point of 2.6 GPa, a non-Fermi-liquid behaviour is found, showing a linear T-dependence of the resistivity, although the Fermi-liquid behaviour recovers above 4 GPa and the A-value decreases with increasing pressure. Furthermore, a resistivity drop, which might be related to superconductivity, was found below 0.3 K at 1.5 GPa, where the residual resistivity ρ_0 has a maximum.

The 4f electrons of rare-earth compounds exhibit a variety of characteristics including spin and valence fluctuations, spin and charge orderings, heavy fermions, Kondo-insulator behaviour and anisotropic superconductivity. In the cerium compounds, the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction and the Kondo effect compete with each other [1]. The former interaction enhances the long-range magnetic order, where the 4f electrons with magnetic moments are treated as localized electrons and the indirect f–f interaction is mediated by the conduction electrons with spins. On the other hand, the latter effect quenches the magnetic moments of the localized 4f electrons by the spin polarization of the conduction electrons, consequently producing the singlet state with the 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 [2] in terms of the function $|J_{cf}|D(\varepsilon_F)$, where $|J_{cf}|$ is the magnitude of the magnetic

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exchange interaction and $D(\varepsilon_F)$ is the electronic density of states at the Fermi energy ε_F. Most of the cerium compounds order magnetically, because the RKKY interaction overcomes the Kondo effect at low temperatures. On the other hand, some cerium compounds such as $CeCu_6$ and $CeRu_2Si_2$ show no long-range magnetic order [1].

Recently a new aspect of the cerium and uranium compounds with antiferromagnetic ordering has been revealed. When pressure p is applied to compounds such as $CeIn_3$ and $CePd_2Si_2$ [3], the Néel temperature T_N decreases and the magnetic 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 the pressure. Surprisingly, superconductivity and/or non-Fermi-liquid character appear around p_c. Correspondingly, a heavy-fermion state is formed as p approaches p_c. Similar pressure-induced superconductivity was also reported for $CeCu_2Ge_2$ [4], $CeRhIn_5$ [5] and UGe_2 [6, 7].

In this work we studied the electronic state near the magnetic quantum critical region in the antiferromagnet UGa_3 with the cubic structure. UGa_3 was once studied from the viewpoint of 5f itinerant antiferromagnetism [8–11]. Its Néel temperature $T_N = 67$K is relatively high. Its characteristic properties are a small ordered moment of 0.75 or 0.95 μ_s/U and a relatively large electronic specific heat coefficient $\gamma = 50$ mJ K^{-2} mol^{-1}. Furthermore, the magnetic susceptibility in the paramagnetic region does not follow the Curie–Weiss law, being dominated by a Pauli-like contribution. From a magnetoresistance experiment, UGa_3 is found to be a compensated metal with equal numbers of carriers of electrons and holes. Carriers with relatively large cyclotron masses of 2.3–9.4 m_0 were detected in a recent de Haas–van Alphen experiment [12].

In the present study, pressure up to 8.5 GPa was applied to the sample of UGa_3. The magnetic ordering disappeared at 2.6 GPa. In this pressure region we found the non-Fermi-liquid behaviour, not following a T^2-dependence of the electrical resistivity but indicating a linear T-dependence of the resistivity.

Single crystals of UGa_3 were grown by the so-called self-flux method. Ga was used as a flux. The residual resistivity ratio ρ_{RT}/ρ_0 was about 80. See reference [12] for details. Pressure up to 8.5 GPa was applied by a cubic anvil apparatus in the temperature region from 2 K to 300 K [13] and by a Bridgman anvil apparatus at lower temperatures down to about 100 mK [14]. We used two different samples for two corresponding measuring systems.

Figure 1 shows the temperature dependence of $d\rho/dT$ under pressure. In the inset we show the temperature dependence of the electrical resistivity ρ at ambient pressure, namely 0 GPa. A small hump in the resistivity at $T_N = 67$ K in the inset of figure 1 corresponds to the Néel temperature. As discussed in our recent paper [12], other transitions are found at $T_1 = 40$ K and $T_2 = 8$ K at ambient pressure, which are not observed under pressure, most probably disappearing. The Néel temperature is found to decrease steeply with increasing pressure, as shown by arrows in figure 1. At 3 GPa, the signature for antiferromagnetic ordering is not detected.

Figure 2 shows a T_N versus pressure phase diagram. The critical pressure p_c is about 2.6 GPa, where T_N becomes zero. The solid line in figure 2 is a guide:

$$T_N(p) = T_N(p = 0)\left(1 - \left(\frac{p}{p_c}\right)^n\right)$$

with $T_N(p = 0) = 65.4$ K, $p_c = 2.6$ GPa and $n = 0.45$.

The electrical resistivity at ambient pressure in UGa_3 is known to follow a T^2-dependence below about 20 K [12]. That is, the electrical resistivity ρ follows a Fermi-liquid dependence of $\rho = \rho_0 + AT^2$. The value of $\sqrt{A}$ correlates with a Pauli susceptibility $\chi \simeq \chi_0$ and a constant γ-value. This Fermi-liquid behaviour is found at 2.0 GPa below about 15 K, as

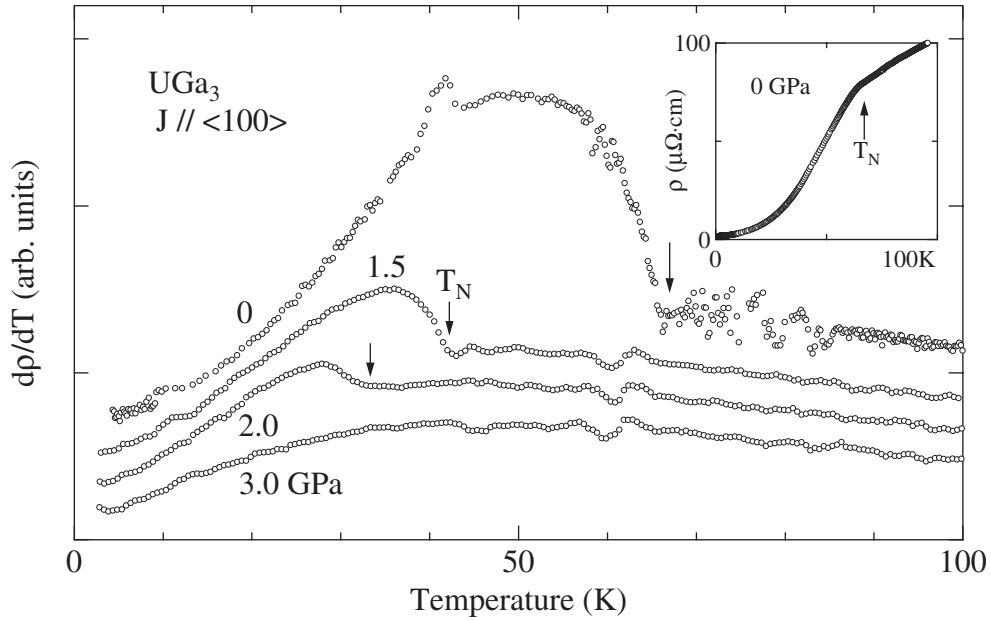

Figure 1. Temperature dependences of the electrical resistivity ρ shown as $\mathrm{d}\rho/\mathrm{d}T$ under pressure in UGa_3. The inset indicates the temperature dependence of the electrical resistivity at ambient pressure.

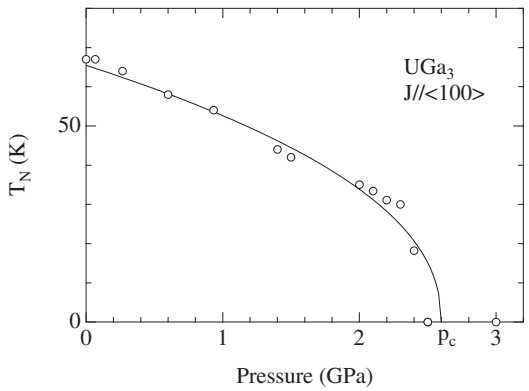

Figure 2. The T_N versus pressure phase diagram of UGa_3.

shown in figure 3. At 2.5 and 3.0 GPa, the T^2-dependence of the electrical resistivity is unclear because the slope of the ρ versus T^2 curve changes with decreasing temperature. With further increasing pressure, the T^2-dependence fully recovers and is followed over a wide temperature range, as seen at 5.0 and 6.0 GPa in figure 3.

Figure 4 shows the pressure dependence of the coefficient A and the residual resistivity ρ_0. The A-value has a maximum around $p_c = 2.6$ GPa, although the residual resistivity does not follow the same pressure dependence, having a maximum at about 1.5 GPa. The reason for this is not clear.

We measured the electrical resistivity at lower temperatures under pressure ranging from 1.5 to 3.1 GPa, using a dilution refrigerator. Figure 5 shows the temperature dependence of

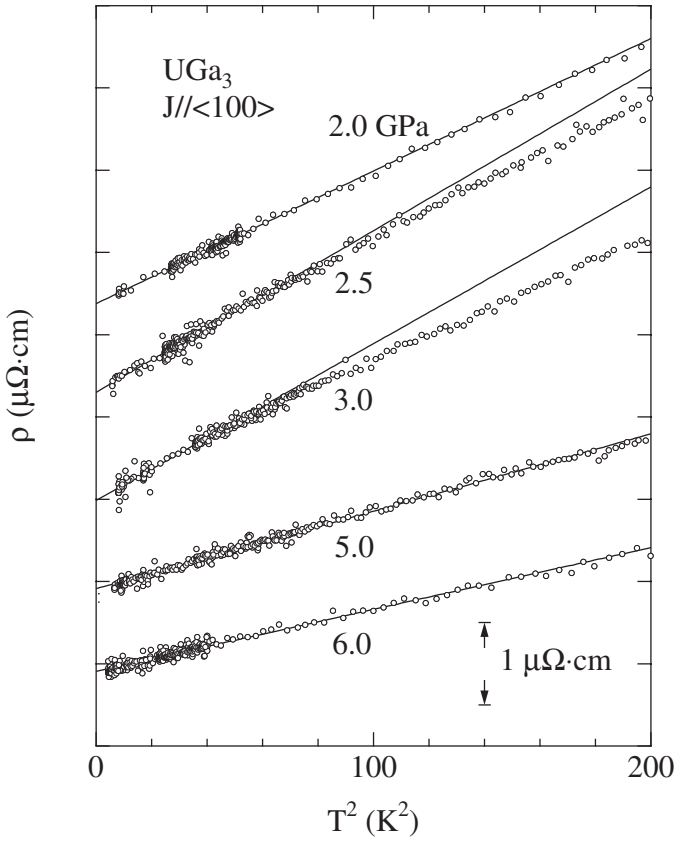

Figure 3. The T^2-dependence of the electrical resistivity under pressure in UGa_3.

the resistivity under pressure. A linear T-dependence is found: $\rho = \rho_0 + A'T$ below 1.6 K at 2.5 GPa and below 2.0 K at 2.8 GPa, which are compared in the figure to the resistivity data under 1.8 and 3.1 GPa. It is clear that the Fermi-liquid behaviour ceases to be present around $p_c = 2.6$ GPa.

Finally, we measured the transverse magnetoresistance

$$\Delta\rho/\rho = \{\rho(H) - \rho(H = 0)\}/\rho(H = 0)$$

at 4.2 K, where the current and magnetic field were applied along the [100] and [001] directions, respectively (see figure 6). The magnetoresistance increases as a function of $\Delta\rho/\rho \sim H^n$ ($n > 1$). This is consistent with the previous result indicating a compensated metal with closed Fermi surfaces [12]. The value of $\Delta\rho/\rho$ decreases with increasing pressure. This means that the mobilities of electrons and holes, μ_e and μ_h, decrease with increasing pressure because $\Delta\rho/\rho \simeq \mu_e\mu_h H^2$ at high fields. In other words, the cyclotron effective mass, which is inversely proportional to the mobility, increases with increasing pressure. The decrease of $\Delta\rho/\rho$ thus corresponds to the increase of the A-value in figure 4(a).

We will discuss the non-Fermi-liquid behaviour in UGa_3 around the magnetic quantum critical point—that is, the electrical resistivity following a T^n-dependence ($n \simeq 1$) at low temperatures around $p_c = 2.6$ GPa. The non-Fermi-liquid behaviour has been discussed in terms of a diverging linear coefficient of the specific heat C for temperatures $T \to 0$:

$$\gamma = C/T = -\ln(T/T_0)$$

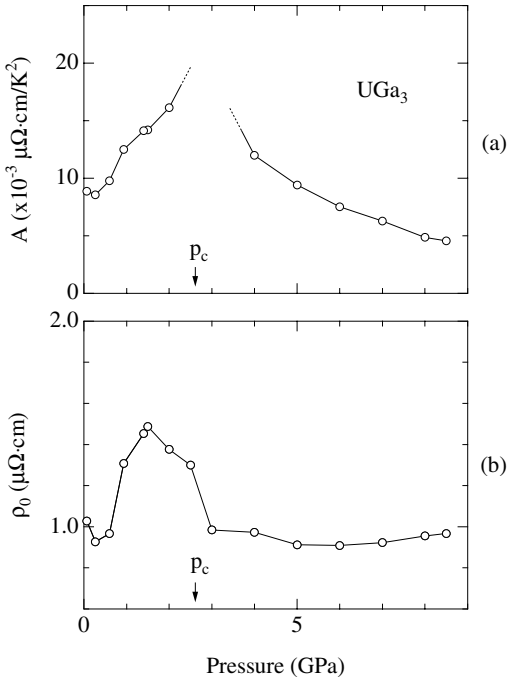

Figure 4. Pressure dependences of A- and ρ_0-values for UGa_3.

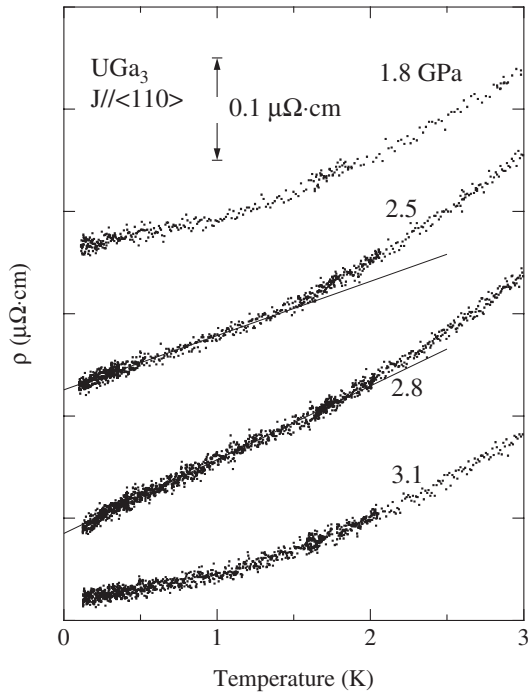

Figure 5. Temperature dependences of the electrical resistivity under pressure in UGa_3.

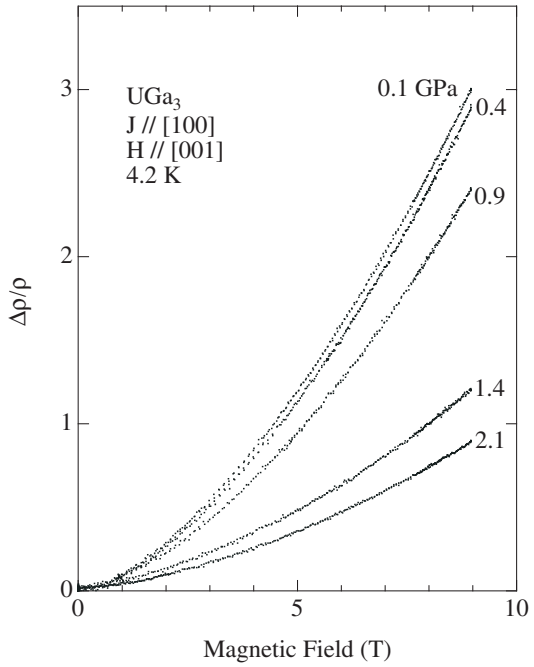

Figure 6. Transverse magnetoresistances under pressure in UGa_3.

and a strong temperature dependence of the magnetic susceptibility χ as $T \to 0$. Furthermore, the electrical resistivity ρ deviates from the T^2-dependence, indicating a linear T-dependence, for example [15].

Experimentally a non-Fermi-liquid relation was observed in several compounds: a quasi-linear resistivity of $\rho \propto T^{1.2}$ from a superconducting transition temperature $T_c \simeq 0.4$ K to high temperatures up to 40 K under $p = 3.05$ GPa in $CePd_2Si_2$ [16] and a linear T-dependence of $\rho \propto T$ from $T_c \simeq 2$ K to 15 K under $p = 15.6$ GPa in $CeCu_2Ge_2$ [17]. Not caused by pressure but caused by chemical pressure of alloying in $CeCu_{2.9}Au_{0.1}$, a linear T-dependence of the resistivity was also observed in the temperature region from 0.3–0.4 K to the lowest measured temperature of 15 mK [18].

One of the theories for the non-Fermi-liquid behaviour around the magnetic quantum critical point is based on the self-consistent renormalization (SCR) theory, taking into account the effect of couplings among the spin-fluctuation modes (mode–mode coupling) [19–21]. This does not, however, explain the linear T-dependence of the resistivity for three-dimensional magnetic systems. The reason for the linear temperature dependence of the resistivity is still an open question.

The non-Fermi-liquid behaviour is closely related to superconductivity as well as to a characteristic feature of the residual resistivity, as discussed for $CePd_2Si_2$ and $CeCu_2Ge_2$ [16, 17, 22]. In the present experiment on UGa_3 we observed a plausible resistivity drop related to superconductivity. Superconductivity under pressure is related to the characteristic features of both the A- and ρ_0-values. In particular, the ρ_0-value is closely related to superconductivity as for $CeCu_2Ge_2$ [22, 23]. In the present experiment the ρ_0-value has a maximum at 1.5 GPa, as shown in figure 4(b). At this pressure we observed a slight decrease of the resistivity below 0.3 K, as shown in figure 7. This resistivity drop might be related to superconductivity. UGa_3 is, however, still in the antiferromagnetic state with $T_N = 45$ K.

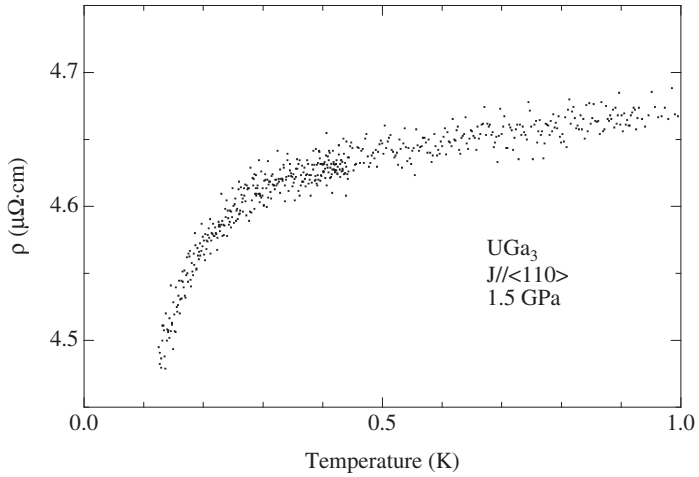

Figure 7. The resistivity drop below 0.3 K at 1.5 GPa in UGa_3.

It might be difficult to conceive of superconductivity being formed in such a state, although coexistence of superconductivity and ferromagnetism is realized in UGe_2 [6, 7].

Non-BCS superconductivity, where the Cooper-pair coupling is based on the spin fluctuations instead of phonons, is very sensitive to impurities, especially non-magnetic ones. Our sample, with a residual resistivity ratio of about 80, which is at present the highest value found in UGa_3, might not be sufficiently pure for the realization of superconductivity. It is furthermore necessary to confirm superconductivity with a better sample. This is left to future study.

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