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LETTER TO THE EDITOR

Pressure-induced superconductivity in a ferromagnet UGe_2

N Tateiwa[1], T C Kobayashi[1,5], K Hanazono[2], K Amaya[2], Y Haga[3], R Settai[4] and Y Ōnuki[4,3]

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

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Abstract
We confirmed bulk-superconductivity of a ferromagnet UGe_2 by the specific heat measurement, together with the measurements of the electrical resistivity and ac susceptibility, in a pressure range from $p = 1.0$ to 1.5 GPa, where the Curie temperature $T_C(= 22–36$ K) is still high, but another characteristic temperature T^* is close to zero. In this pressure range, the heavy fermion state is found to be formed at low temperatures.

Cerium and uranium compounds indicate a variety of phenomena including magnetic and quadrupolar ordering, heavy fermion and anisotropic superconductivity [1]. In these compounds, the RKKY interaction and the Kondo effect compete with each other. The former interaction enhances the long-range magnetic order, while the latter effect quenches the magnetic moments of localized *f* electrons. Most of the cerium and uranium compounds order magnetically, where the former interaction overcomes the latter effect. When the magnetic ordering temperature is low enough or close to zero, the heavy fermion state is formed at low temperatures. The conduction electrons in the heavy fermion state are highly different from bare electrons. They are interacting electrons, moving slowly in the crystal, which correspond to a large effective mass m^* or a large electronic specific heat coefficient γ.

When pressure p is applied to the cerium compounds with antiferromagnetic ordering such as $CeIn_3$ and $CePd_2Si_2$, the Néel tempereture T_N shifts to lower temperatures, and the magnetic quantum critical point corresponding to the extrapolation $T_N \to 0$ is reached at $p = p_c$ [2]. Superconductivity appears around p_c. Correspondingly, the heavy fermion state is formed as p approaches p_c. This seems to be a general feature, although the sample quality is essentially important for the appearance of superconductivity. This is because superconductivity is most likely to be magnetically-mediated or of a non-s-wave type and then the breaking of Cooper-pairs is mainly due to impurities and crystal defects.

[5] To whom correspondence should be addressed (e-mail: tckobayashi@mp.es.osaka-u.ac.jp)

Recently it was reported that a ferromagnet UGe_2 is a pressure-induced superconductor [3]. This is the first case where the ferromagnet becomes a superconductor under pressure. Superconductivity was observed in the pressure range from 1.0 to 1.5 GPa by measuring the electrical resistivity and ac susceptibility. Surprisingly, in this pressure range UGe_2 is still in the ferromagnetic state.

We previously studied the fundamental properties of UGe_2 [4–9]. It is a highly anisotropic ferromagnet with a Curie temperature $T_C = 52$ K. A saturated magnetic moment is 1.4 μ_B/U, directed along the a-axis in the orthorhombic structure. The magnetization is of an Ising type, being highly anisotropic. We note that the crystal structure was once believed to be orthorhombic ($Cmcm$), but later it was altered to $Cmmm$ ($a = 4.0089$ Å, $b = 15.0889$ Å and $c = 4.0950$ Å) [10]. The notation of a-, b- and c-axes in the previous reports are changed to the c-, b- and a-axes, respectively.

Ferromagnetic ordering in UGe_2 is not simple. For example, the electrical resistivity for the current along the a- and c-axes decreases steeply below T_C, as in the usual ferromagnet, while the resistivity for the current along the b-axis indicates a peak structure below T_C, as in the formation of the spin-density wave for an antiferromagnet [4]. This might be related to a change of the Fermi surface below T_C, from a paramagnetic Fermi surface to a ferromagnetic one. The carriers have moderately large effective masses in the range from 1.4 to 21 m_0 at low temperatures, reflecting $\gamma = 30$ mJ K^{-2}mol [5, 6]. The main Fermi surface observed in the de Haas–van Alphen experiment is a cylindrical one whose axis is along the b-axis [5]. Recent energy band calculations indicate that the cross-section of the cylinder is not circular but rather rectangular [11].

Furthermore, it is noted that other characteristic features were observed under pressure. The Curie temperature disappears for $p > 1.8$–2.0 GPa [7]. Moreover, a characteristic transition temperature T^* was observed below T_C [8, 9]. This anomaly was observed at $T^* \simeq 30$ K as a round shoulder in the electrical resistivity for the current along the a- and c-axes, and was also reflected clearly in the thermal expansion coefficient. This temperature T^* becomes close to zero around 1.2 GPa [9], where superconductivity was found below 0.7 K [3].

We continued studying the electronic state under pressure for a high-quality sample. To confirm bulk-superconductivity we measured the specific heat, together with the electrical resistivity and ac susceptibility.

A single crystal was grown by the Czochralski method in a tetra-arc furnace as described in [6]. The purity of the starting materials was 99.98% for U and 99.999% for Ge. The ingot was 4 mm in diameter and 80 mm in length, and it was annealed at 800 °C in a high vacuum of 5×10^{-11} torr for seven days. As for the present sample, the residual resistivity ρ_0 and the residual resistivity ratio RRR($= \rho_{RT}/\rho_0$) were 0.26 $\mu\Omega\cdot$cm and 600, respectively, at ambient pressure, indicating a high-quality sample.

The electrical resistivity and the ac susceptibility were measured by the ac four-probe and Hartshorn bridge methods, respectively. The specific heat measurement was done by the adiabatic heat pulse method. Pressure was applied by utilizing a CuBe piston-cylinder cell with a Daphne oil (7373) as a pressure-transmitting medium.

Figure 1 shows the temperature dependence of the electrical resistivity ρ at 1.0 GPa for the current along the c-axis. A steep decrease in the resistivity below $T_C = 34$ K is due to the ferromagnetic ordering. Furthermore, another decrease in the resistivity is found clearly below 11 K, which corresponds to the characteristic temperature T^* mentioned above. Both T_C and T^* decrease with increasing pressure.

From these measurements we formed the phase diagram, as shown in figure 2. Both T_C and T^* decrease approximately as a function of $(p_c - p)^n (n \simeq 0.5$–$0.4)$. Solid and broken lines are a guide to eyes. Superconductivity is observed in the pressure range from 1.0 to 1.5

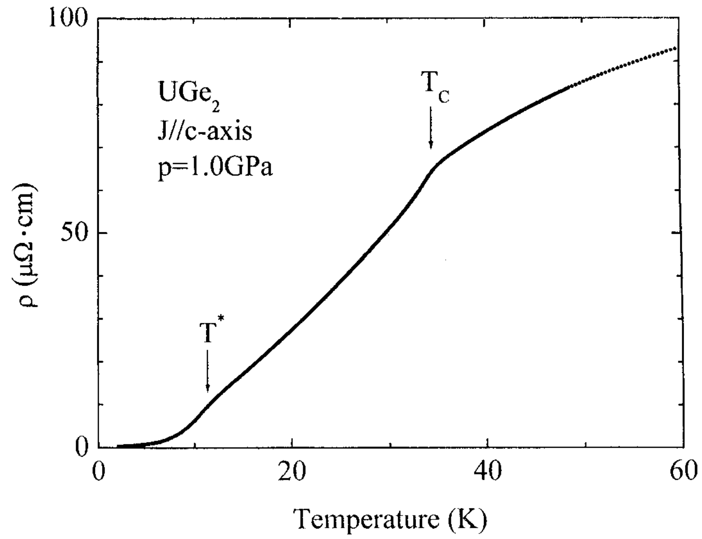

Figure 1. Temperature dependence of the electrical resistivity for the current along the c-axis at 1.0 GPa in UGe_2.

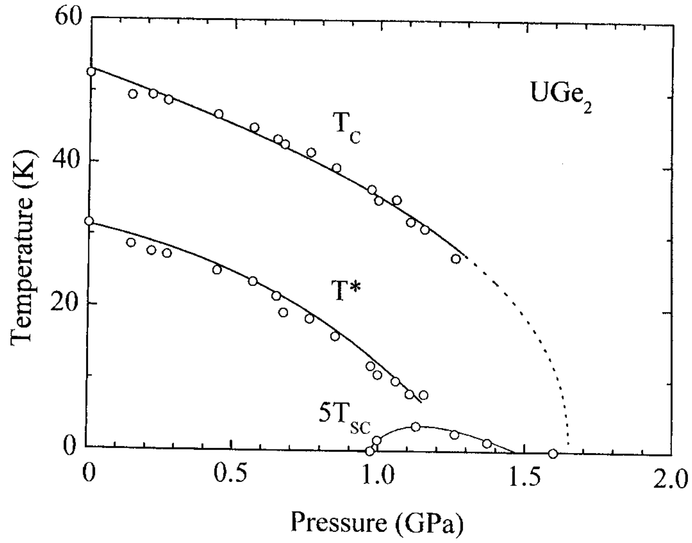

Figure 2. Phase diagram under pressure in UGe_2.

GPa, where the vertical temperature scale is enlarged by five times for the superconducting temperature T_{SC}. The maximum T_{SC} is 0.7 K. The phase diagram is in good agreement with the recent result by Saxena *et al* [3].

We observed superconductivity, as shown in the inset of figure 3. The transition temperature T_{SC} is 0.47 K at 1.26 GPa. The onset temperature is 0.55 K, and the zero-resistivity temperature is 0.45 K. We define here the transition temperature T_{SC} as the temperature where the resistivity is close to zero, as shown by an arrow in the inset of figure 3. We also confirmed superconductivity for the sample with RRR = 60. The onset temperature is 0.5 K, but the

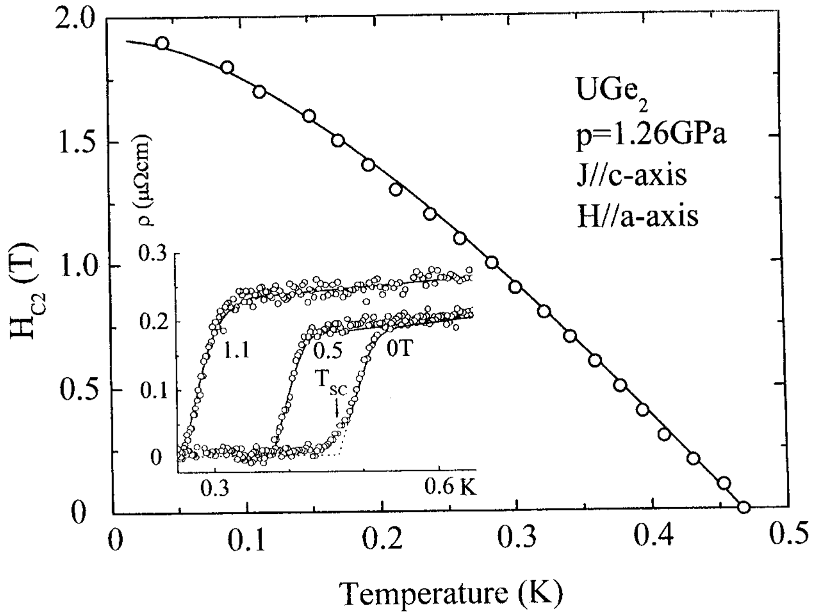

Figure 3. Upper critical field at 1.26 GPa for the field along the a-axis in UGe_2. The inset shows the temperature dependence of the electrical resistivity under the fields of 0, 0.5 and 1.1 T in UGe_2.

zero-resistivity is attained at a lower temperature of 0.1 K. This indicates that the present superconductivity is very sensitive to the sample quality.

The inset of figure 3 shows the temperature dependence of the electrical resistivity under a magnetic field. From these data, the upper critical field H_{c2} is determined as a function of the corresponding temperature. The superconducting phase diagram is obtained, as shown in figure 3. H_{c2} at zero temperature is estimated as 1.9 T. The solid line in figure 3 is a guide to eyes, calculated on the basis of the well known WHH theory [12]. The coherence length ξ is thus estimated as 130 Å from H_{c2} $(= \phi_0/2\pi\xi^2)$, where ϕ_0 is a quantum fluxoid.

We also confirmed superconductivity from the specific heat measurement. Figure 4 shows the temperature dependence of the specific heat in the form of C/T for $p = 0$ and 1.13 GPa, together with the ac susceptibility. The ac susceptibility was measured for the same sample, simultaneously with the specific heat measurement. The γ value is 97 mJ K^{-2}mol for $p = 1.13$ GPa, which is about three times larger than 30 mJ K^{-2}mol determined at ambient pressure $p = 0$. At 0.7 K $(= T_{SC})$, we found a peak corresponding to the transition of superconductivity. The C/T value in figure 4 decreases approximately linearly below T_{SC}. The residual γ value of 71 mJ K^{-2}mol is, however, large. Correspondingly, $\Delta C/\gamma T_{SC}$ is 0.2–0.3, which is small, compared to a BCS value of 1.45. Here ΔC is the jump of the specific heat at T_{SC}. Simply thinking, 70% of the Cooper-pairs are broken, producing a large amount of quasiparticles.

Next we investigated the electronic state by the electrical resistivity and the specific heat measurements. In the Fermi liquid state, the resistivity at low temperatures follows $\rho = \rho_0 + AT^2$, and $\sqrt{A}$ is proportional to the γ value. Figure 5 shows the pressure dependence of A and γ. The A value increases steeply above $p = 1.0$ GPa, where superconductivity starts to appear, and has a maximum at p =1.3–1.4 GPa . In this pressure range, T^* becomes zero. Correspondingly, the γ value also has a large value of 100 mJ K^{-2}mol. The features of the A

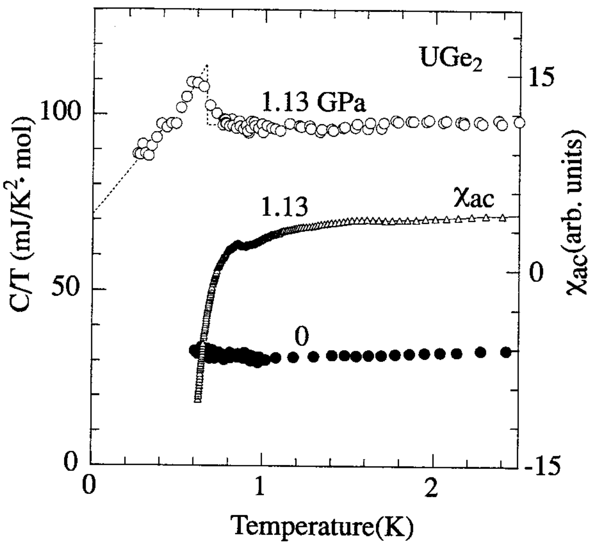

Figure 4. Temperature dependence of the specific heat in the form of C/T at 0 and 1.13 GPa, togehter with the ac susceptibility at 1.13 GPa in UGe_2.

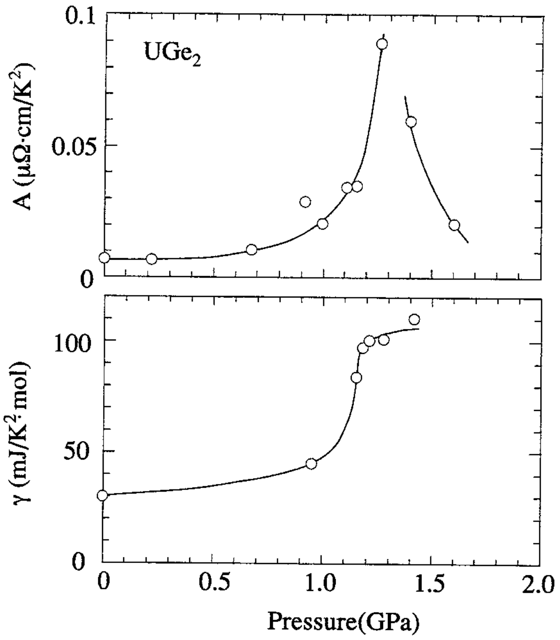

Figure 5. Pressure dependence of the A and γ values in UGe_2.

value as a function of pressure are almost the same as reported recently [3]. We note that the A value was reported previously to decrease steeply with increasing pressure ($p > 2$ GPa) [7].

Here, the A value was obtained from the resistivity measurement, as shown in figure 6. We

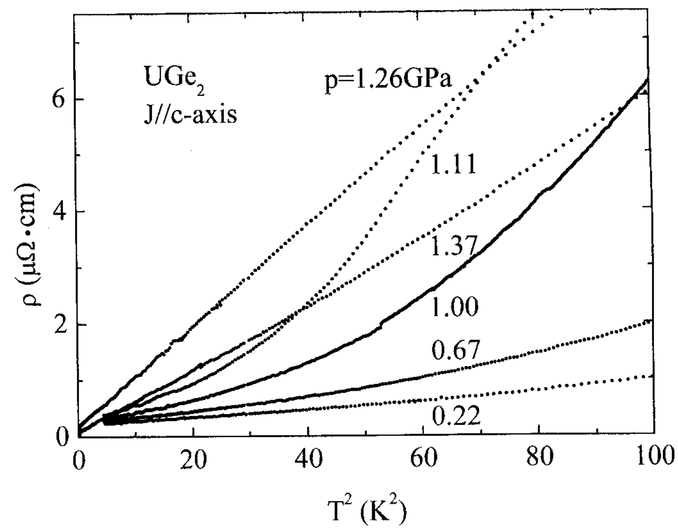

Figure 6. T^2-dependence of the electrical resistivity under pressure in UGe_2.

note that the A value depends on the anomaly at T^*. When the characteristic temperature T^* is high enough, the A value is not related to it, following the T^2-dependence below about 8 K, for example, for $p = 0.22$ GPa. With increasing pressure, the T^2-dependence is applicable below about 4 K for $p = 1.11$ GPa, because T^* is 8 K for this pressure. The characteristic phenomenon occurring at T^* merges into the T^2-dependence of the resistivity for $p = 1.26$ GPa, as seen in figure 6.

The origin of the phase transition or the phenomenon occuring at T^* is not clear at present. There is, however, a possibility that the charge-density wave or the spin-density wave is formed below T^* [3]. The main Fermi surface is rectangular in cross-section and cylindrical along the b-axis, which might favour the partial nesting of the Fermi surface along the a- and/or c-axes [11]. This phenomenon couples to the electronic and magnetic states because the 5f electrons become conduction electrons related to the main Fermi surface and also carry the magnetic moments in the ferromagnetic state. The 5f electrons have a dual nature. A large γ value for $p = 1.2$–1.6 GPa is closely related to this phenomenon and to the appearance of superconductivity.

We noticed that the characteristic phenomenon occuring at T^* depends on the magnetic field applied along the direction of the magnetic moment or the a-axis. We continue our investigation to clarify this phenomenon.

Superconductivity in UGe_2 was theoretically discussed on the basis of a non-unitary triplet pairing, which favours superconductivity in the ferromagnetic state [13]. To clarify the nature of superconductivity, we point out (1) a relation between a mean free path l and the coherence length ξ, and (2) the residual γ value. The mean free path is roughly estimated from the residual resistivity of 0.2–0.3$\mu\Omega$·cm for the present sample. More directly we can estimate the l value from the results of de Haas–van Alphen effect for the main cylindrical Fermi surface [14]. This Fermi surface is corrugated, having maximum and minimum cross-sections. One of them has a cyclotron effective mass of 10 m_0 and the l value of 1300 Å at ambient pressure, which is changed to 15 m_0 and 1400 Å at 1.0 GPa. Under 1.0 GPa, T_{SC} was 40 mK. The mean free path of 1400 Å is one order larger than the ξ value of 130 Å.

Next we confirmed bulk superconductivity from the specific heat experiment. The residual γ value, which amounts to 70% of the total γ value is, however, extremely large. It is a crucial problem whether a large residual γ value is intrinsic or not. This is closely related to a validity of the theory including the non-unitary triplet pairing mechanism mentioned above [13]. Usually, the residual γ value depends on the sample quality, namely the pair-breaking due to the crystal defects and impurities. We will check it for a higher-quality sample in the near future because the present sample with RRR $= 600$ does not exceed the previous one with RRR $= 910$.

In conclusion, we confirmed bulk superconductivity of UGe_2 from the specific heat measurement. The present superconductivity is closely related to the phenomenon occuring at $T^*(< T_C)$. In a pressure region from $p = 1.0$ to 1.5 GPa, where T^* is close to zero but T_C (= 22–36 K) is still high, the heavy-fermion superconducting state is formed, possessing a large value of $\gamma \simeq 100$ mJ K^{-2}mol.

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