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2002 J. Phys.: Condens. Matter 14 L377

(http://iopscience.iop.org/0953-8984/14/21/102)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **14** (2002) L377–L383 PII: S0953-8984(02)36178-2

LETTER TO THE EDITOR

Pressure-induced superconductivity in an antiferromagnet $CeRh_2Si_2$

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Received 24 April 2002
Published 16 May 2002
Online at stacks.iop.org/JPhysCM/14/L377

Abstract
We studied the pressure effect by measuring the electrical resistivity for a high-quality single crystal of an antiferromagnet $CeRh_2Si_2$ with the tetragonal structure. The Néel temperature becomes zero around 1 GPa. An onset of superconductivity appears below 0.5 K in a relatively wide pressure region from 0.97 to 1.20 GPa, while the resistivity zero, which appears below 0.4 K, is found to be located in an extremely narrow region from 1.03 to 1.08 GPa. Correspondingly the A value of the Fermi liquid relation in the electrical resistivity ($\rho = \rho_0 + AT^2$) becomes maximum around 1 GPa. Superconductivity is realized in a heavy fermion state.

The f electrons of cerium and uranium compounds exhibit a variety of characteristics including spin and valence fluctuations, heavy fermions, Kondo insulators and anisotropic superconductivity [1]. Recently, a new aspect of these compounds with magnetic ordering has been discovered. When pressure P is applied to the cerium compounds with antiferromagnetic ordering such as $CeIn_3$, $CePd_2Si_2$ [2], $CeCu_2Ge_2$ [3] and $CeRhIn_5$ [4], the Néel temperature $T_N \to 0$ is reached at the quantum critical pressure P_c. Superconductivity and/or the non-Fermi liquid nature are observed around P_c. Very recently superconductivity was also found in a ferromagnetic state in UGe_2 [5] and URhGe [6]. The crossover from the magnetic ordered state to the non-magnetic state under pressure, crossing the quantum critical point, is the most interesting issue in strongly correlated f-electron systems.

$CeRh_2Si_2$ crystallizes in the tetragonal $ThCr_2Si_2$-type structure and orders antiferromagnetically below the Néel temperature $T_{N1} = 36$ K at ambient pressure. Its magnetic structure was investigated by the neutron diffraction measurements [7]. Below T_{N1}, the propagation

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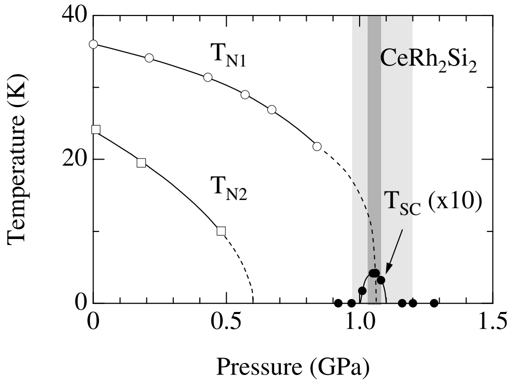

Figure 1. Pressure dependence of T_{N1}, T_{N2} and T_{sc} in $CeRh_2Si_2$. The temperature of T_{sc} is enlarged by ten times compared to those of T_{N1} and T_{N2}. The data on T_{N2} are cited from [7].

vector is $\boldsymbol{q}_1 = (1/2, 1/2, 0)$ and the magnetic moment is oriented along the [001] direction. Furthermore, the magnetic structure changes to the 4$\boldsymbol{q}$-structure with two propagation vectors $\boldsymbol{q}_1$ and $\boldsymbol{q}_2 = (1/2, 1/2, 1/2)$ below $T_{N2} = 25$ K. The magnitude of the magnetic moment is 1.42 μ_B at the corner-Ce site of the tetragonal structure and 1.34 μ_B at the body-centred Ce site.

When pressure is applied to $CeRh_2Si_2$, the Néel temperature $T_{N1} = 36$ K and the magnetic transition temperature T_{N2} are suppressed to 0 K at $P_{c1} = 1.0$–1.1 GPa and $P_{c2} \simeq 0.6$ GPa, respectively, as shown in figure 1. The T_{N1} value and the superconducting transition temperature T_{sc} are our data, determined from the electrical resistivity, as shown later. The value of T_{sc} is enlarged by ten times compared to T_{N1} and T_{N2}. The T_{N2} value is cited from [7].

A few measurements under pressure were done for $CeRh_2Si_2$. The specific heat measurement indicated that the electronic specific heat coefficient γ increases almost linearly with increasing pressure from 23 mJ K^{-2} mol^{-1} at ambient pressure to about 80 mJ K^{-2} mol^{-1} at P_{c1} [8]. Above P_{c1}, the γ value decreases slightly with increasing pressure. Similarly, the A coefficient in the electrical resistivity ($\rho = \rho_0 + AT^2$) also indicated a maximum around P_{c1} [9]. Moreover, we recently clarified from the de Haas–van Alphen (dHvA) experiment that the topology of the Fermi surface changes discontinuously at P_{c1}, indicating that the 4f electrons change from localized to itinerant at P_{c1} [10].

Superconductivity in $CeRh_2Si_2$ was first found by Movshovich *et al* [11] for a polycrystal sample. The transition temperature, determined from an onset of the resistivity drop, is $T_{sc} = 0.35$ K, where superconductivity appears around P_{c1}, namely in the pressure range from 0.6 to 1.6 GPa. We searched for superconductivity in a single crystal with the residual resistivity ratio (RRR) RRR $\simeq 30$, but no evidence of superconductivity was observed. On the other hand, the superconducting resistivity drop was observed below $T_{sc} = 0.6$ K for a polycrystal sample with RRR $= 62$ in the pressure range from 0.7 to 2.9 GPa [12]. The T_{sc} value was unchanged in this pressure range, but the degree of the resistivity drop had a maximum around P_{c1}. Recently, the superconducting resistivity drop was observed at 1.1 GPa for a high quality (RRR $= 110$) single crystalline sample, where T_{sc} value, determined from the onset of resistivity drop, was 0.38 K but the resistivity had a finite value even at 35 mK [13].

We continued the investigation of superconductivity for a high quality single crystal sample. The resistivity zero was confirmed in an extremely narrow pressure region around P_{c1}.

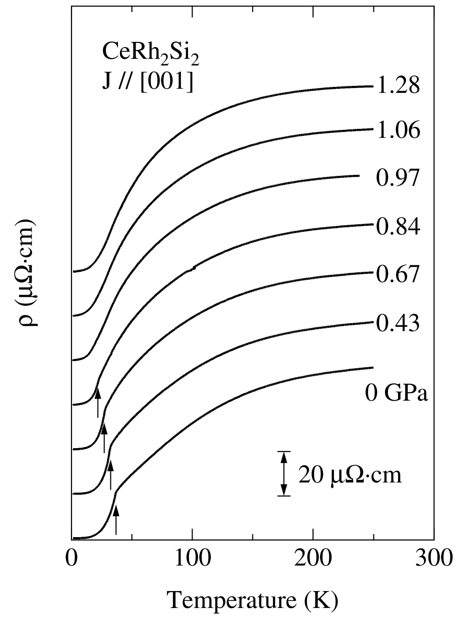

Figure 2. Temperature dependence of the electrical resistivity in $CeRh_2Si_2$ under various pressures. The Néel temperature T_{N1} is indicated by arrows.

Single crystals of $CeRh_2Si_2$ were grown by the Czochralski pulling method in a tetra-arc furnace, as described in [14]. Starting materials were 4N (99.99% pure)-Ce, 4N-Rh and 5N-Si. The electrical resistivity was measured by a four-probe ac resistance bridge (Linear Research, LR-700) at low temperatures down to about 100 mK with a dilution refrigerator and by a four-probe dc-method at high temperatures with a ^{4}He cryostat. The current was directed along the [001] direction. Pressures up to 1.28 GPa were applied by utilizing a BeCu piston-cylinder cell with a 1:1 mixture of commercial Daphne oil (7373) and kerosene as a pressure-transmitting medium. The pressure was calibrated by the T_{sc} value of Sn.

Figure 2 shows the temperature dependence of the electrical resistivity under several pressures up to 1.28 GPa. The resistivity data under pressure are shifted with regard to each other for convenience. The residual resistivity ρ_0 and the RRR at ambient pressure are $\rho_0 = 0.75\ \mu\Omega$cm and RRR $= 100$, respectively.

The steep resistivity drop below $T_{N1} = 36$ K at ambient pressure, shown by an arrow in figure 2, is due to the antiferromagnetic ordering. T_{N1} decreases with increasing pressure and is not observed above 0.97 GPa. On the other hand, no anomaly at T_{N2}, which corresponds to the change of the magnetic structure, is observed in the present resistivity. We note that the anomaly at T_{N2} was observed in the resistivity measurement for the current along the [100] direction for a relatively low-quality sample (typically RRR $= 30$) [14]. In the neutron experiments under pressure [7], both transitions were clearly observed: T_{N1} was observed up to 1.08 GPa and T_{N2} up to 0.48 GPa. The T_{N2} data, determined from the neutron experiment, are plotted in figure 1.

The critical pressure for T_{N1} was estimated as $P_{c1} = 1.1$ GPa in the neutron scattering experiment and as 1.06 GPa in our previous dHvA experiment [10]. A dotted curve for T_{N1} in figure 1 is a guideline reaching $P_{c1} = 1.06$ GPa. It is noted that T_{N1} decreases abruptly when pressure approaches P_{c1} and becomes zero at P_{c1}. Following the results of our dHvA

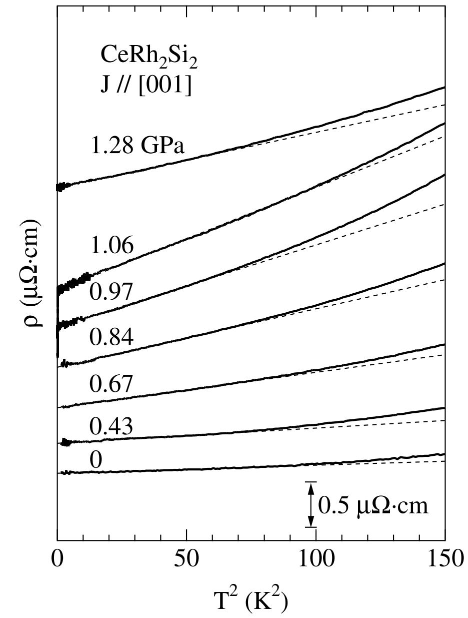

Figure 3. Quadratic temperature dependence of the electrical resistivity in $CeRh_2Si_2$ under various pressures.

experiment, the topology of the Fermi surface changes abruptly at P_{c1}. The phase boundary at P_{c1} is most likely of the first order. A similar result was observed in UGe_2 and was discussed from a viewpoint of the first-order phase transition [15, 16].

The low-temperature resistivity under pressure follows the Fermi liquid form ($\rho = \rho_0 + AT^2$) within an experimental error. Figure 3 shows the T^2-dependence of the electrical resistivity. It is noted that the T^2-dependence is satisfied even around P_{c1}, as was pointed out previously [9, 17]. This is a characteristic feature in $CeRh_2Si_2$.

We show in figure 4 the pressure dependence of A and ρ_0 values. The A value has a maximum at 1.0 GPa, very close to P_{c1}. The residual resistivity ρ_0 also has a small anomaly around P_{c1}.

Superconductivity appears around P_{c1}, as shown in figure 5. An indication of superconductivity appears in the pressure region from 0.97 to 1.20 GPa, which is shown in figure 1 as a grey region. As shown in figure 5, the resistivity at $P = 1.01$ and 1.16 GPa decreases gradually with decreasing temperature below 0.5 K. The resistivity zero is, however, not attained. The resistivity zero is observed in an extremely narrow pressure region around P_{c1}, which is shown in figure 1 as a dense-grey region. Namely, the resistivity at $P = 1.05$ and 1.06 GPa decreases below 0.5 K and becomes zero at 0.4 K. It is noted that the onset temperature in superconductivity is 0.4–0.5 K in the pressure region from 0.97 to 1.20 GPa, but the resistivity zero is attained in the pressure region from 1.03 to 1.08 GPa. This implies that homogeneous bulk-superconductivity is realized in an extremely narrow pressure region around P_{c1}.

We determined the upper critical field H_{c2} in superconductivity. Figure 6 shows the temperature dependence of the electrical resistivity under magnetic fields along the [001] direction. The superconducting temperature T_{sc} for each field is defined as the temperature obtained from the extrapolation of the resistivity drop, as shown at 0 T in figure 6.

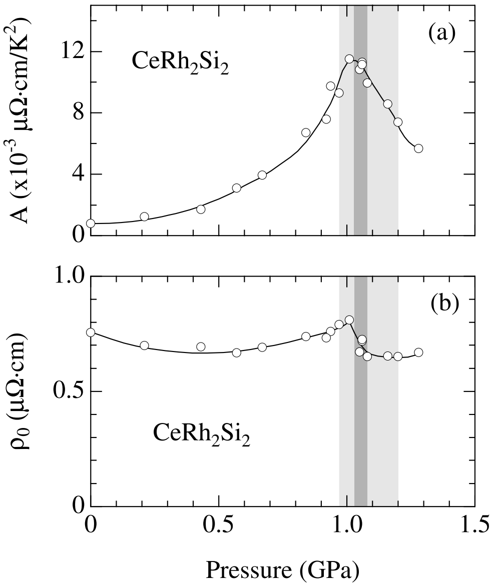

Figure 4. Pressure dependence of (a) the A and (b) ρ_0 values in $CeRh_2Si_2$.

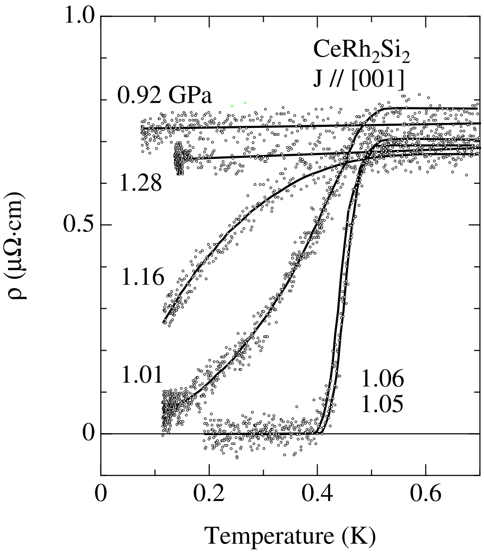

Figure 5. Low-temperature resistivity under pressure in $CeRh_2Si_2$.

Figure 7 shows the temperature dependence of H_{c2}. The solid line in figure 7 is a guide to eyes. The coherence length ξ is estimated as 340 Å from $H_{c2}(0)$ $(= \Phi_0/2\pi\xi^2)$, where Φ_0 is the quantum flux. We note that the mean free path for the present sample around 1.06 GPa is estimated from the dHvA experiment, being about 1000 Å. This indicates that the present sample is close to a clean limit.

Characteristic features at the quantum critical point, namely at P_{c1} are the most important issue to be clarified. As for $CeRh_2Si_2$, the present experimental results are summarized as follows:

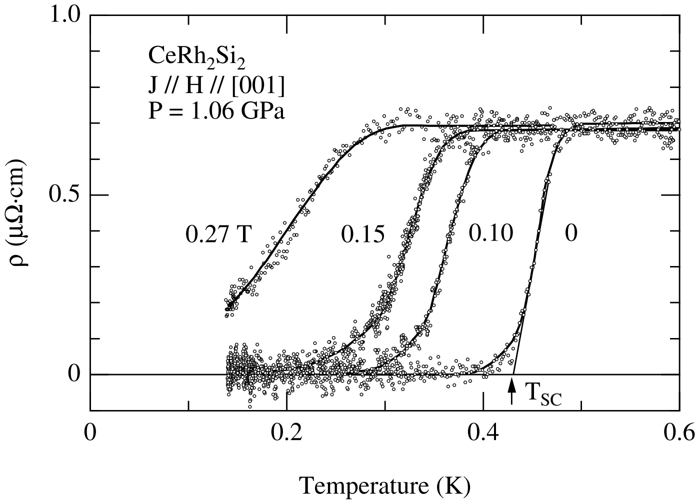

Figure 6. Low-temperature resistivity under various magnetic fields at 1.06 GPa in $CeRh_2Si_2$.

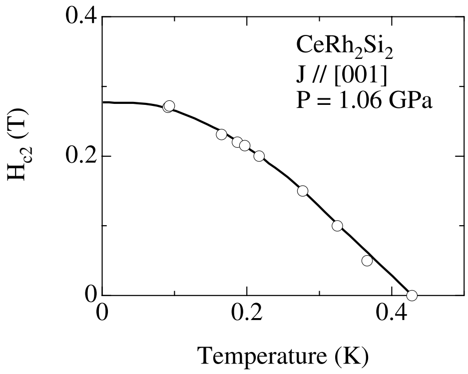

Figure 7. Temperature dependence of H_{c2} at 1.06 GPa in $CeRh_2Si_2$.

(i) The Fermi liquid nature of $\rho = \rho_0 + AT^2$ is satisfied in the whole pressure region, even at P_{c1}.
(ii) The pressure dependence of the A value has a maximum around P_{c1}.
(iii) Following the results of our previous dHvA experiment, the topology of the Fermi surface changes abruptly at P_{c1}, implying that the 4f electrons change from localized to itinerant when pressure crosses P_{c1}. This might be related to an abrupt decrease of T_{N1} when pressure approaches P_{c1}. These results indicate that the transition at P_{c1} is of the first order.
(iv) Homogeneous bulk-superconductivity appears in an extremely narrow pressure region around P_{c1}.
(v) The upper critical field was determined as $H_{c2}(0) = 0.28$ T for $H \parallel [001]$, implying that the coherence length ξ is 340 Å.

We are grateful to Professor K Miyake for helpful discussion. This work was financially supported by the Grant-in-Aid for COE Research (10CE2004) from the Ministry of Education, Culture, Sports, Science and Technology of Japan. SA has been supported by JSPS.

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