Evolution of pressure-induced heavy fermion state and superconductivity in $CeRhIn_5$: A high-pressure Fermi surface study

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Evolution of the pressure-induced heavy fermion state and superconductivity in $CeRhIn_5$ have been established by high-pressure Fermi surface studies using dHvA technique. The effective mass of the conduction electrons named branch β_2 increases from 5.5 m_0 at ambient pressure to 20 m_0 at 1.6 GPa where superconductivity sets in. This heavy electron mass continues to increase steeply up to 2.1 GPa, 45 m_0 at 2.1 GPa, with a concomitant increase of the superconducting transition temperature. However, the underlying Fermi surface remains approximately same even at 2.1 GPa, strongly suggesting that antiferromagnetic fluctuations of Ce moments do not contribute to the volume of the Fermi surface, but enhance the cyclotron mass of the conduction electrons.

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INTRODUCTION

The *f* electrons of cerium and uranium compounds exhibit a variety of phenomena including spin and valence fluctuations, heavy fermions, and anisotropic superconductivity. In these compounds, both the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the Kondo effect compete with each other.[1] The former leads to long-range magnetic order, in which *f* electrons with magnetic moments are treated as localized electrons and the indirect *f*-*f* interaction mediated by the conduction electrons plays a predominant role. On the other hand, the latter quenches the magnetic moments of the localized electrons via spin polarization of conduction electrons, consequently producing a singlet state with a binding energy k_BT_K, where T_K is the Kondo temperature. Competition between the RKKY interaction and the Kondo effect was discussed by Doniach[2] 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.

Recent studies[3,4] indicate that many interesting phase transitions occur in *f*-electron compounds under pressure. When pressure *p* is applied to the *f*-electron compounds with antiferromagnetic order, the Néel temperature T_N decreases, and a magnetic quantum critical point corresponding to the limit $T_N \rightarrow 0$ is reached at $p = p_c$. Here, $|J_{cf}|D(\varepsilon_F)$ in the Doniach model can be replaced by pressure. Surprisingly, superconductivity and non-Fermi-liquid behavior appear around p_c. The crossover from the antiferromagnetic state to the nonmagnetic state under pressure is one of the most interesting issues in strongly correlated *f*-electron systems. The purpose of the present study is to report a pressure-induced electronic state in the superconducting phase of $CeRhIn_5$ by probing the Fermi surface using the de Haas–van Alphen (dHvA) experiment on a high-quality crystal of $CeRhIn_5$.

$CeRhIn_5$ as well as related compounds $CeCoIn_5$ and $CeIrIn_5$ crystallize in the tetragonal structure with alternating layers of $CeIn_3$ and $RhIn_2$, stacked sequentially along the [001] direction (*c* axis). Hegger *et al.* found that $CeRhIn_5$ orders antiferromagnetically below $T_N = 3.8$ K, but reveals an antiferromagnetic to superconducting transitions at a relatively low pressure $p^* = 1.63$ GPa.[5] The superconducting transition temperature $T_{sc} = 2.2$ K at 2.5 GPa is the highest value in the pressure-induced superconductors, as shown in Fig. 1 (Refs. 6–8). The superconducting phase is dominant in a wide pressure region from 1.6 to 5 GPa. On the other hand, $CeCoIn_5$ and $CeIrIn_5$ mentioned above are superconductors at ambient pressure, with $T_{sc} = 2.3$ and 0.4 K, respectively.[9]

The topology of main Fermi surfaces in the antiferromagnet $CeRhIn_5$ is nearly cylindrical, and is found to be in good agreement with that of a reference non-4*f* compound $LaRhIn_5$, indicating that the 4*f* electrons in $CeRhIn_5$ are localized and do not contribute to the volume of the Fermi surface.[10,11] A magnetic moment of 0.374 μ_B/Ce in $CeRhIn_5$ resides on the Ce ion, and forms a helical spin structure.[12]

Many cerium compounds show local moment magnetism

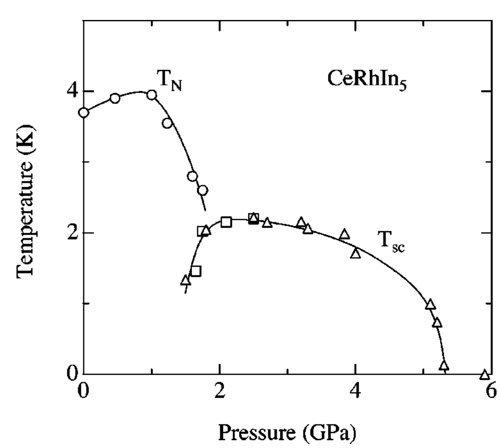

FIG. 1. Pressure dependence of the Néel temperature T_N and the superconducting transition temperature T_{sc}, obtained from the the resistivity data (△) (Ref. 6) and the NMR and ac-susceptibility data (○, □) (Ref. 7,8).

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where the Fermi surface is quite similar to the non-4f lanthanum compound. For example, the topology of the Fermi surface in $CeAl_2$ and CeB_6 is similar to that in $LaAl_2$ and LaB_6, respectively, although the cyclotron mass of $CeAl_2$ and CeB_6 is one to two orders of magnitude larger than that of $LaAl_2$ and LaB_6.[13,14] The cyclotron masses of main Fermi surfaces in $CeRhIn_5$ are seven to nine times larger than the corresponding cyclotron masses in $LaRhIn_5$.[10,11] This is consistent with an electronic specific heat coefficient γ: $\gamma \simeq 50$ mJ/K^2 mol in $CeRhIn_5$ and 5.7 mJ/K^2 mol in $LaRhIn_5$. The degree of mass enhancement depends on the degree of competition between the RKKY interaction and the Kondo effect, which in turn depends on the degree of hybridization of the 4f electrons with the conduction electrons.

On the other hand, main Fermi surfaces in nonmagnetic compounds $CeCoIn_5$ and $CeIrIn_5$ are also nearly cylindrical but are identified by the 4f-itinerant band model.[11,15,16] The topology of two kinds of cylindrical Fermi surfaces of $CeCoIn_5$ is similar to that of $CeRhIn_5$, but the volume of the Fermi surfaces of $CeCoIn_5$ is larger than that of $CeRhIn_5$ because one 4f electron in each Ce site becomes a band electron in $CeCoIn_5$. The detected cyclotron masses of 5–87 m_0 in $CeCoIn_5$ are extremely large, and correspond to a large γ value of 1000 mJ/K^2 mol. A large cyclotron mass over 100 m_0 was also detected in $CeRu_2Si_2$ and UPt_3.[17–19] These compounds including $CeCoIn_5$ are called heavy fermion compounds since they have a large γ value $\gamma \simeq 10^4/T_K$ (mJ/K^2 mol), where T_K is the Kondo temperature. This means that the Kondo singlet state in each cerium (uranium) site forms an f-derived heavy band at low temperatures.[1,19]

Previous dHvA works were mainly carried out for magnetic and nonmagnetic f-electron compounds and were compared to the results of the corresponding non-4f lanthanum compounds and/or energy band calculations. In order to elucidate the nature of the electronic state in the pressure-induced superconductor $CeRhIn_5$, the present dHvA work was carried out from ambient pressure to 2.1 GPa in magnetic fields up to 170 kOe and at low temperatures down to 80 mK.

EXPERIMENT

Single crystals were grown by the self-flux method.[11] The residual resistivity ρ_0 and residual resistivity ratio ρ_{RT}/ρ_0 were 0.18 $\mu\Omega$ cm and 330, respectively, indicating a high-quality sample. The dHvA experiment was done by the standard field modulation method with a modulation frequency of 5 Hz and a modulation field of 80 Oe. Pressure was applied by utilizing a MP35N piston-cylinder cell with a 1:1 mixture of commercial Daphne oil (7373) and kerosene.

EXPERIMENTAL RESULTS AND ANALYSES

Figure 2 shows the dHvA oscillation at 1.3 GPa and the corresponding fast Fourier transformation (FFT) spectrum, together with the FFT spectra under 1.8 and 2.0 GPa in the magnetic field along [001]. Here, the detected dHvA signal V_{osc} for the field H is simply written as follows:[1]

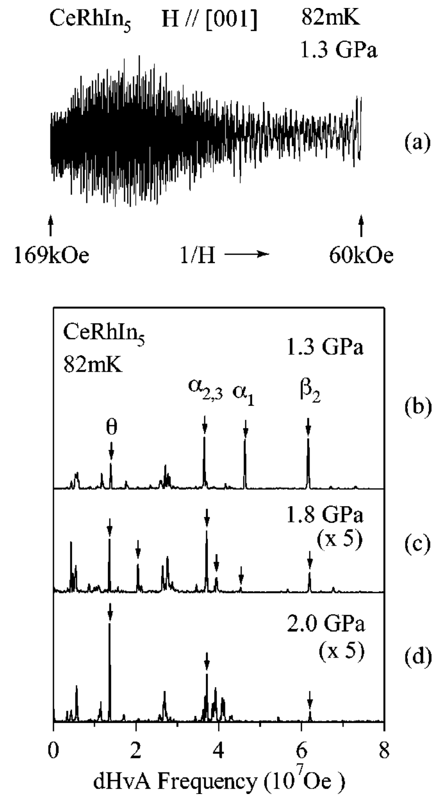

FIG. 2. (a) dHvA oscillation at 1.3 GPa and (b) its FFT spectrum, (c) and (d) are FFT spectra at 1.8 and 2.0 GPa, respectively.

$$V_{osc} = A \sin\left(\frac{2\pi F}{H} + \phi\right), \tag{1}$$

$$A \propto J_2(x) T H^{-1/2} \frac{\exp\left(-\alpha m_c^* \frac{T_D}{H}\right)}{\sinh\left(\alpha m_c^* \frac{T}{H}\right)}, \tag{2}$$

$$\alpha = \frac{2\pi^2 c k_B}{e\hbar}, \tag{3}$$

and

$$x = \frac{2\pi F h}{H^2}, \tag{4}$$

where the dHvA frequency F $(=\hbar c S_F/2\pi e)$ in the horizontal scale in Fig. 2(b), which is expressed as a unit of magnetic field, corresponds to the extremal (maximum or minimum) cross-sectional area of the Fermi surface S_F, $J_2(x)$ is the Bessel function, possessing a maximum at 3.1, h is the modulation field, m_c^* is the cyclotron effective mass, and $T_D[=(\hbar/2\pi k_B)(1/\tau)]$ is the Dingle temperature which is inversely proportional to the scattering lifetime of the conduction electron τ.

The main three branches named α_1, $\alpha_{2,3}$, and β_2 in Figs. 2(b)–2(d) are due to a band 15-electron Fermi surface (α_i) and a band 14-electron Fermi surface (β_2), respectively. Both Fermi surfaces are corrugated but cylindrical along

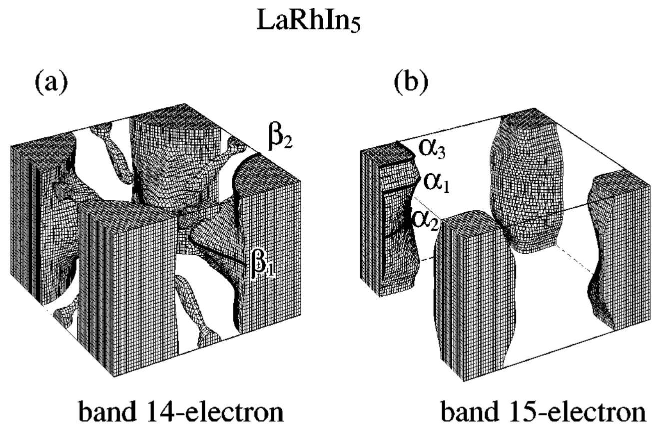

FIG. 3. Bands 14- and 15-electron Fermi surfaces in $LaRhIn_5$, taken from Ref. 11.

[001] as shown in Fig. 3. For these Fermi surfaces, it is enough to study the dHvA experiment under pressure only for $H \| [001]$. The other branches including branch θ are most likely produced by antiferromagnetic ordering. Detailed identification of each branch is described in Ref. 11. The FFT spectrum at $p = 1.3$ GPa is almost the same as that at ambient pressure.

With increasing pressure, the dHvA amplitude is strongly reduced. The FFT spectra at $p = 1.8$ and 2.0 GPa in Figs. 2(c) and 2(d) are enlarged by five times compared to that at $p = 1.3$ GPa because the dHvA amplitude A is reduced by a steep increase of the cyclotron mass above 1.6 GPa, shown later. The typical four branches θ, $\alpha_{2,3}$, α_1, and β_2 are observed at 1.8 GPa, together with appearance of new branches with $F = 2.1 \times 10^7$ and 4.0×10^7 Oe, while at 2.0 GPa branch α_1 disappears completely and new branches also appear around the dHvA frequency of branch $\alpha_{2,3}$. The reason for the appearance of these new branches for $p > 1.8$ GPa is not understood at this moment. We show in Fig. 4 the pressure dependence of the dHvA frequency for the four branches.

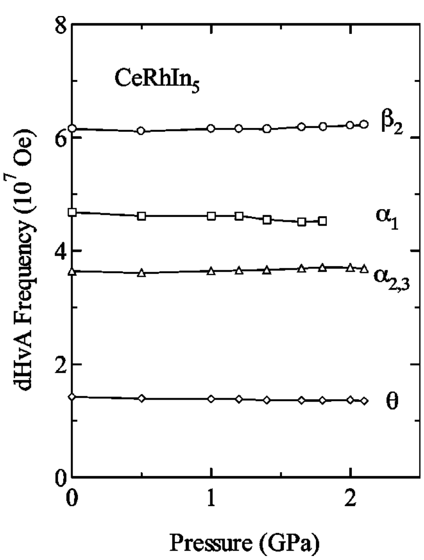

FIG. 4. Pressure dependence of the dHvA frequency for four dHvA branches in $CeRhIn_5$.

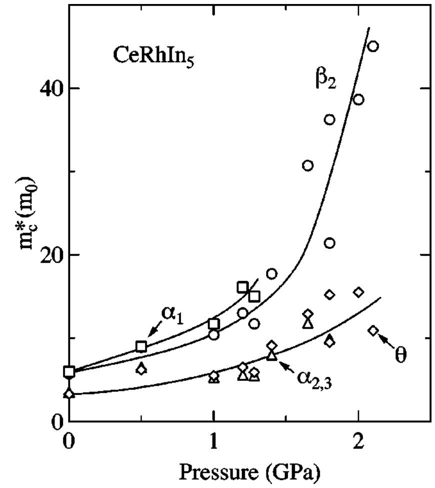

FIG. 5. Pressure dependence of the cyclotron mass for four dHvA branches in $CeRhIn_5$.

The magnitude of the dHvA frequency for at least three branches is unchanged in the pressure range from 0 to 2.1 GPa. The magnitude of the dHvA frequency of branch α_1 is also unchanged at least up to 1.8 GPa.

By using Eqs. (1)–(4), we determined the cyclotron effective mass m_c^* from the temperature dependence of the dHvA amplitude, namely, from the slope of a plot of $\ln[A\{1-\exp(-2\alpha m_c^* T/H)\}/T]$ vs T on the basis of successive approximations. Figure 5 shows the pressure dependence of the cyclotron mass, which was determined at $H = 120$ kOe. The cyclotron mass increases steeply above 1.6 GPa, where superconductivity sets in. For example, the cyclotron mass of branch β_2 is 5.5 m_0 at ambient pressure, 20 m_0 at 1.6 GPa and 45 m_0 at 2.1 GPa. Furthermore, we found that the cyclotron mass is strongly field dependent with increasing pressure, as shown in Fig. 6. At ambient pressure, the cyclotron mass of branch β_2 is not field dependent, but at 2.0 GPa the

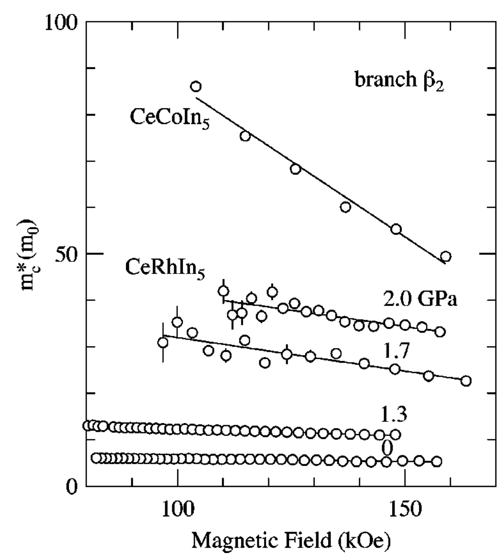

FIG. 6. Field dependence of the cyclotron mass for branch β_2 in $CeRhIn_5$. The data for $CeCoIn_5$ at ambient pressure are taken from Ref. 16.

cyclotron mass of 40 m_0 at 110 kOe is reduced to 33 m_0 at 158 kOe. This is also found in the other branches. In Fig. 6, a much steeper mass reduction due to magnetic fields is shown for $CeCoIn_5$ at ambient pressure.[16] The mass reduction due to magnetic fields is a characteristic phenomenon in heavy fermion compounds.[20] We point out that a considerable mass reduction is only observed above $p^* = 1.63$ GPa where superconductivity sets in.

When pressure is increased, the Néel temperature T_N slightly increases but starts to decrease above $p = 1.0$ GPa, as shown in Fig. 1. At the characteristic pressure $p^* = 1.63$ GPa, the internal field at the In site in the $CeIn_3$ plane, obtained by the ^{115}In NQR experiment,[7] becomes zero and the superconducting state sets in. Furthermore it was clarified that antiferromagnetic order disappears at least at 2.1 GPa but three dimensional antiferromagnetic fluctuations are dominant in the normal state from the NQR experiments showing the $\sqrt{T}$ dependence of the nuclear spin-lattice relaxation rate.[7,21] There is a possibility that the Fermi surface in $CeRhIn_5$ may change into an *f*-itinerant Fermi surface similar to that of $CeCoIn_5$ at $p > p^*$. The present result shows that this does not occur at least up to $p = 2.1$ GPa. It is thus suggested that the antiferromagnetic fluctuations of Ce moments do not contribute to the volume of the Fermi surface, but enhance the cyclotron mass of the conduction electrons, most likely via the many-body Kondo effect. We wish to recall here that in the case of local moment magnetism, one does not observe a change in the Fermi surface similar to the case of $CeAl_2$ and CeB_6 where the underlying Fermi surfaces of Ce compounds are quite similar to their La compounds.[13,14] The recent Fermi surface studies under pressure using dHvA technique for an antiferromagnet $CeRh_2Si_2$ indicated that the topology of the Fermi surface is found to be almost unchanged up to a critical pressure $p_c \simeq 1.0$ GPa but changes abruptly above p_c, meaning a first-order-like phase transition at p_c.[22,23] A new Fermi surface for $p > p_c$ is well explained by the $4f$-itinerant band model in $CeRh_2Si_2$.

On the other hand, the nature of the superconducting state in $CeRhIn_5$ around 2.5 GPa is almost the same as that of $CeCoIn_5$ at ambient pressure. Namely, the existence of a line node of the energy gap in the superconducting state was confirmed in both compounds.[7,8,21] Recently the node in $CeCoIn_5$ was determined from the thermal conductivity measurement under magnetic fields to be located at [110], being directed along [001] for nearly cylindrical Fermi surfaces, indicating symmetry of $d_{x^2-y^2}$ type.[24] The upper critical field in superconductivity for both compounds is about 100 kOe for the field perpendicular to [001].[6,16] Furthermore, it was clarified from the electrical resistivity, magnetic susceptibility and specific heat measurements that both compounds exhibit superconductivity in the non-Fermi liquid state at zero field.[11] From these experimental results, $CeRhIn_5$ around 2.5 GPa and $CeCoIn_5$ at ambient pressure are in the vicinity of the quantum critical point.

It is interesting to note that the topology of the Fermi surface is nevertheless different between $CeRhIn_5$ under pressures up to 2.1 GPa and $CeCoIn_5$ at ambient pressure. It is, however, expected that the topology of the Fermi surface in $CeRhIn_5$ will be similar to that of $CeCoIn_5$ either at pressures higher than 2.5 GPa at which the superconducting transition temperature T_{sc} becomes a maximum, or at pressures higher than 5 GPa at which T_{sc} becomes zero. Further studies are needed to settle the second issue.

SUMMARY

In conclusion we have shown that the topology of the Fermi surface of the antiferromagnet $CeRhIn_5$, which is similar to that of $LaRhIn_5$, is essentially unchanged up to $p = 2.1$ GPa. On the other hand, the cyclotron mass increases steeply above 1.6 GPa where superconductivity sets in: 5.5 m_0 at ambient pressure, 20 m_0 at 1.6 GPa, and 45 m_0 at 2.1 GPa for the cyclotron mass of a band 14-electron Fermi surface named branch β_2. The superconducting state is realized in the heavy fermion state.

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