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LETTER TO THE EDITOR

A de Haas–van Alphen experiment under pressure on $CeCoIn_5$: deviation from the quantum critical region

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Abstract
We have carried out a de Haas–van Alphen experiment under pressure on a heavy-fermion superconductor, $CeCoIn_5$. Large cyclotron masses of 15 m_0 (m_0: the rest mass of an electron) in a nearly cylindrical Fermi surface called α_i ($i = 1, 2, 3$) and 60 m_0 in a similar cylindrical Fermi surface called β_2 at ambient pressure are found to be strongly reduced to 7 and 40 m_0, respectively, at 3 GPa. Correspondingly, the field-dependent cyclotron mass at ambient pressure becomes almost independent of the field at high pressures. These results indicate that $CeCoIn_5$, which is in the vicinity of the quantum critical region at ambient pressure, is changed into a usual heavy-fermion state under high pressures of about 3 GPa.

The interplay between magnetism and superconductivity is one of the most important issues remaining to be clarified in modern condensed matter physics. $CeCoIn_5$, which was discovered only recently [1], is a strong-coupling superconductor with a huge jump of the specific heat $\Delta C/C_e(T_c) = 4.5$ (e.g. a BCS value is 1.43), together with the highest transition temperature, $T_c = 2.3$ K, among the heavy-fermion superconductors [1, 2].

The temperature dependences of the specific heat [1, 2], thermal conductivity [3] and NMR relaxation rate [4] in the superconducting state indicate a power-law dependence, suggesting unconventional superconductivity. Even-parity pairing was furthermore clarified from the NMR Knight shift measurement [4], and the gap symmetry is of the $k_x^2 - k_y^2$ type as shown by angle-resolved thermal conductivity measurements [5], indicating that the pair interaction is mediated by magnetic fluctuations.

In addition to these characteristic features, and more interesting, is the non-Fermi liquid behaviour. The electrical resistivity shows a linear temperature dependence from $T_c = 2.3$ K

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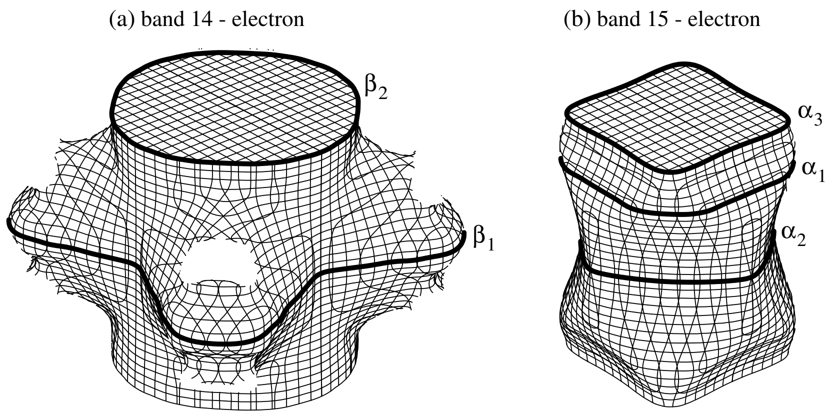

Figure 1. Bands 14- and 15-electron Fermi surfaces in $CeCoIn_5$.

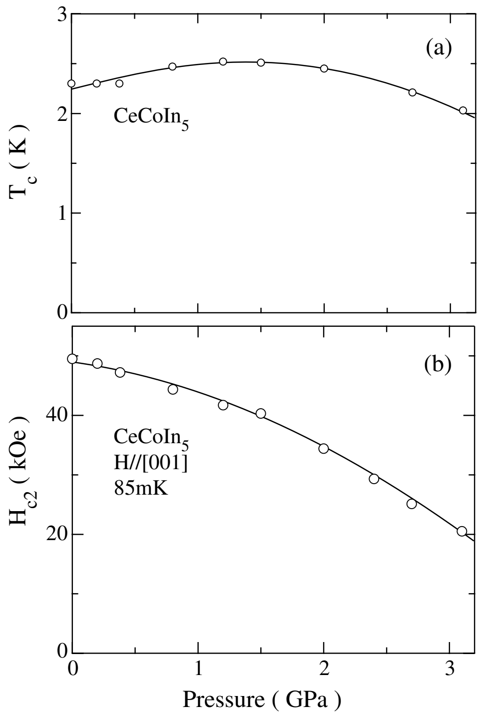

Figure 2. Pressure dependences of the superconductivity transition temperature T_c and upper critical field H_{c2} of $CeCoIn_5$.

to about 10 K [1, 6]. The electronic specific heat C_e in the form C_e/T at 60 kOe increases as a function of $\sqrt{T}$ below 0.8 K, suggesting a three-dimensional antiferromagnetic quantum critical fluctuation [2]. Note that the upper critical field H_{c2} of the superconductivity is 50 kOe in this measurement. The magnetization M in the form M/H at 10 kOe does not become constant at low temperatures, but can be well expressed by the form $\chi = \chi_0 + C/(T^\alpha + a)$ ($\alpha = 0.8$–1) below 15 K [7]. $CeCoIn_5$ is thus close to antiferromagnetic criticality.

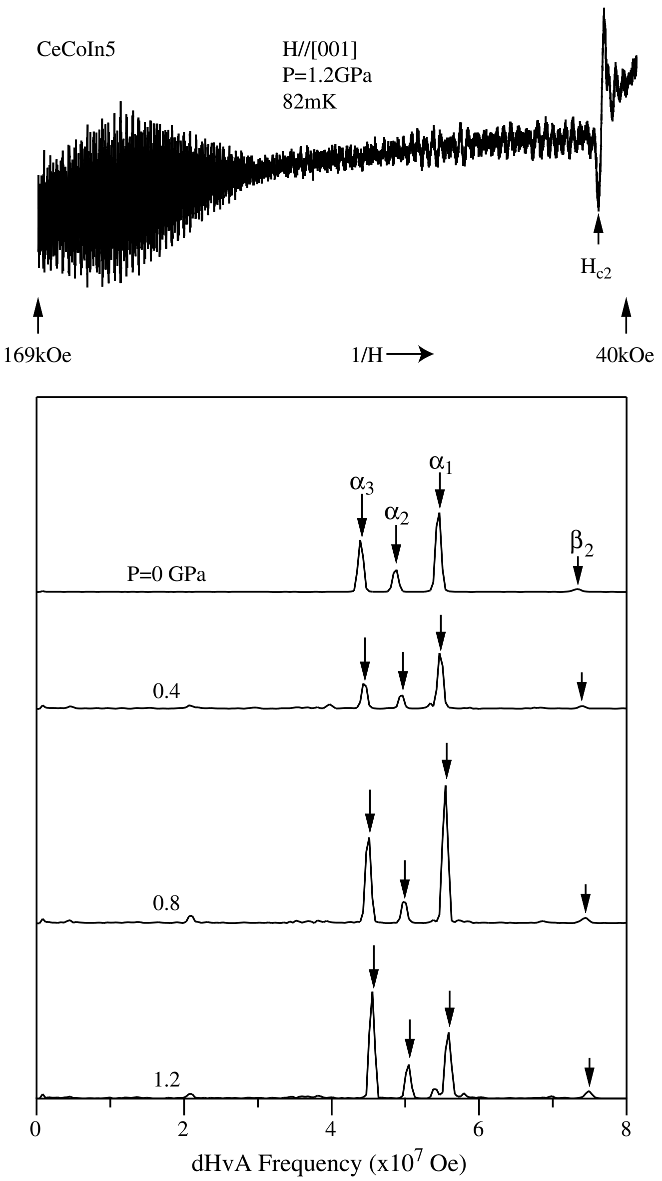

Figure 3. dHvA oscillation at 1.2 GPa for $H \parallel [001]$ under 1.2 GPa and FFT spectra at 0, 0.4, 0.8 and 1.2 GPa for $CeCoIn_5$.

These characteristic properties are closely related to the tetragonal crystal structure with alternating layers of $CeIn_3$ and $CoIn_2$, stacked sequentially along the [001] direction (*c*-axis). This unique crystal structure brings about a quasi-two-dimensional electronic state. That is, the 3d electrons hybridize with the 5p electrons of In in the $CoIn_2$ layer, forming bonding and antibonding bands, which results in a small density of states around the Fermi energy. This means that there are scarcely conduction electrons in the $CoIn_2$ layer, and hence the main Fermi surfaces with a dominant 4f-electron character consist of cylindrical but highly corrugated bands of 14- and 15-electron Fermi surfaces [6, 8], as shown in figure 1.

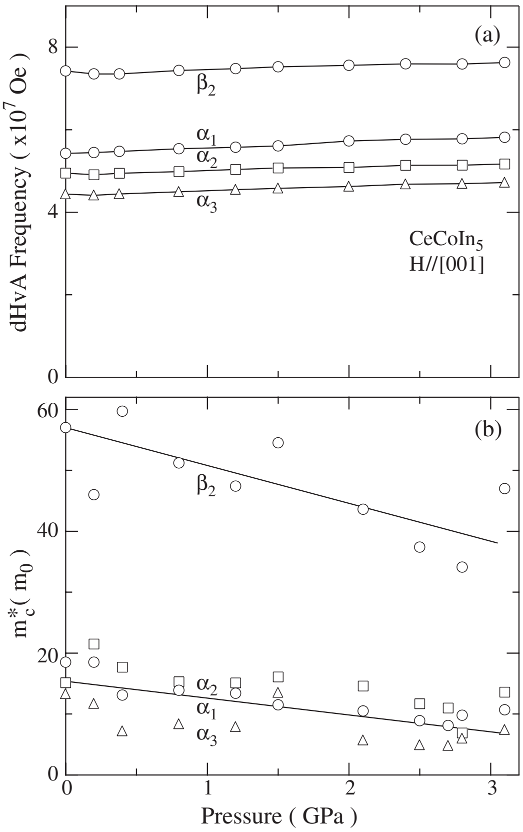

Figure 4. Pressure dependences of the dHvA frequency and cyclotron mass in $CeCoIn_5$.

It was clarified from our previous de Haas–van Alphen (dHvA) experiment that the cyclotron mass is extremely large, corresponding to a large C_e/T (=1 J K^{-2} mol^{-1}) value at 0.25 K under 60 kOe: 85 m_0 (m_0: the rest mass of an electron) at 90 kOe in branch β_2 of the band of the 14-electron Fermi surface and 17–30 m_0 at 50 kOe in branches α_i (i = 1, 2, 3) of the band of the 15-electron Fermi surface. These large cyclotron masses are found to be reduced steeply with increasing fields: 50 m_0 in branch β_2 and 8–18 m_0 in branches α_i at 160 kOe.

The electronic state can also be tuned by applying pressure [9]. The specific heat and electrical resistivity experiments under pressure revealed that the increase of C_e/T with decreasing temperature is reduced steeply with increasing pressure and the linear temperature dependence of the resistivity is changed into a usual Fermi liquid relation—a T^2-dependence—above 2.5 GPa. To clarify furthermore the electronic state under pressure via a microscopic probe, we have carried out a de Haas–van Alphen (dHvA) experiment at high pressures up to 3.1 GPa.

The dHvA experiment was done by the standard field modulation method with a modulation frequency of 3.4 Hz and a modulation field of 80 Oe. Pressure was applied by utilizing a MP35 piston–cylinder cell with a 1:2 mixture of commercial Daphne oil (7373) and petroleum ether. The single crystal was the same as the one used previously [6, 8].

To elucidate an effect of pressure on superconductivity and the electronic state, we show in figure 2 the pressure dependences of the transition temperature T_c and the upper critical

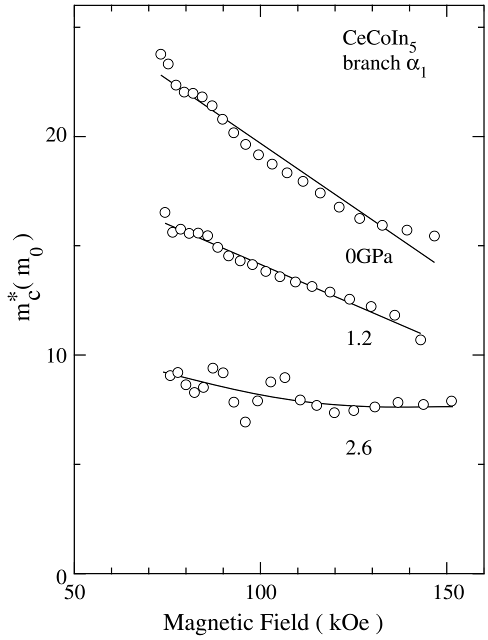

Figure 5. The magnetic field dependence of the cyclotron mass for branch α_1 in $CeCoIn_5$.

field H_{c2}. The present data were obtained by the ac susceptibility measurement via the dHvA technique, as shown later. T_c increases slightly with increasing pressure, has a maximum of $T_c = 2.5$ K at 1.5 GPa and decreases gradually with further increasing pressure.

The change of T_c is thus small in this pressure region, whereas H_{c2} decreases steeply with increasing pressure: 50 kOe at ambient pressure and 20 kOe at 3.1 GPa. This is closely related to a decrease of the C_e/T value with increasing pressure, although it is clarified that the magnitude of H_{c2} at ambient pressure is strongly reduced by the paramagnetic effect [2, 7].

Figure 3 shows the typical dHvA oscillation under 1.2 GPa in the field range from 40 to 169 kOe at 82 mK, and its fast Fourier transformation (FFT) spectrum, together with spectra at 0, 0.4 and 0.8 GPa. The magnetic field is directed along [001]. H_{c2} is clearly observed in the dHvA oscillation, as shown by an arrow. The main branches, called β_2, α_1, α_2 and α_3, in the FFT spectra, mentioned above, are approximately unchanged as regards the magnitude of the dHvA frequency and also the amplitude. Here the dHvA frequency F $(=\hbar S_F/2\pi e)$ is proportional to the extremal (maximum or minimum) cross-sectional area of the Fermi surface S_F.

Figure 4 shows the pressure dependence of the dHvA frequency and the corresponding cyclotron mass m_c^*. The dHvA frequency is unchanged as a function of pressure, indicating that the topology of the Fermi surface is unchanged. On the other hand, the cyclotron masses decrease steeply with increasing pressure. The cyclotron masses of 58 m_0 for branch β_2 and 14–18 m_0 for branches α_i are reduced to 40 and 5–10 m_0, respectively, at 3 GPa. Here, the cyclotron mass was determined by the temperature dependence of the dHvA amplitude at 140 kOe for branch β_2 and at 110 kOe for branches α_i.

As mentioned above, the cyclotron mass at ambient pressure is strongly field dependent. We show in figure 5 the field dependence of the cyclotron mass for branch α_1. The cyclotron mass at ambient pressure is reduced from 24 m_0 at 73 kOe to 15 m_0 at 145 kOe. On the other hand, the reduction of the cyclotron mass at 2.6 GPa is very small. This is consistent with the recent results of electrical resistivity measurements under pressure, because the Fermi liquid relation of the T^2-dependence of the electrical resistivity is satisfied above 2.5 GPa [9]. The electronic state in the vicinity of the quantum critical region at ambient pressure is changed to a usual heavy-fermion state above 2.5 GPa.

The present characteristic features contrast with those of the antiferromagnet $CeRhIn_5$ under pressure [10–12]. Pressure-induced superconductivity appears above 1.6 GPa in $CeRhIn_5$. Correspondingly the cyclotron mass increases steeply above 1.6 GPa, at which superconductivity sets in. The electronic state of $CeRhIn_5$ at 2.5 GPa is very similar to that of $CeCoIn_5$ at ambient pressure.

From the result of the present dHvA experiment, together with the previous results of the electrical resistivity, specific heat and magnetization measurements, it is concluded that $CeCoIn_5$ at ambient pressure is in the quantum critical region. The huge cyclotron mass is strongly field dependent, and is extremely reduced by magnetic fields. This characteristic feature diminishes with increasing pressure. The cyclotron mass at high fields is almost field independent at 2.6 GPa. The electronic state deviates from the quantum critical region at ambient pressure to the usual Fermi liquid state above 2.5 GPa.

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