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LETTER TO THE EDITOR

Quasi-two-dimensional Fermi surfaces and the de Haas–van Alphen oscillation in both the normal and superconducting mixed states of $CeCoIn_5$

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Abstract

We observed de Haas–van Alphen (dHvA) oscillation in both the normal and superconducting mixed states of a heavy-fermion superconductor $CeCoIn_5$. The Fermi surfaces are found to consist of nearly cylindrical Fermi surfaces and small ellipsoidal ones, reflecting the unique tetragonal crystal structure. The detected cyclotron masses of 5–87 m_0 for these Fermi surfaces are extremely large, and correspond to a large electronic specific heat coefficient of about 1000 mJ K^{-2} mol^{-1}. The cyclotron masses are also found to be field dependent in both the normal and mixed states.

The 4f electrons of rare-earth compounds exhibit a variety of characteristics including spin fluctuations, heavy fermions, and anisotropic superconductivity [1]. In the cerium compounds, the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction and the Kondo effect compete with each other. The former enhances the long-range magnetic order, while the latter quenches the magnetic moments of the localized 4f electrons by the spin polarization of the conduction electrons, consequently producing quasiparticles with a large effective mass called heavy fermions at low temperatures.

Recently it was reported that $CeIrIn_5$ and $CeCoIn_5$ are heavy-fermion superconductors [2–4]. The transition temperature T_c and the electronic specific heat coefficient C/T are 0.4 K and 680 mJ K^{-2} mol^{-1} for $CeIrIn_5$, and 2.3 K and 300–1000 mJ K^{-2} mol^{-1} for $CeCoIn_5$. Here, the C/T value of $CeCoIn_5$ is about 300 mJ K^{-2} mol^{-1} at T_c under $H = 0$, while it increases to about 1000 mJ K^{-2} mol^{-1} at 0.1 K under 50 kOe [2]. On the other hand, $CeRhIn_5$,

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ordering antiferromagnetically below $T_N = 3.8$ K, exhibits superconductivity under pressure $p > 1.6$ GPa [5].

These characteristic properties are closely related to the unique tetragonal crystal structure ($P4/mmm$, No 123, D^1_{4h}) with alternating layers of $CeIn_3$ and TIn_2 (T: Co, Rh, and Ir), stacked sequentially along the [001] direction (c-axis). The lattice parameters a and c are 4.614 Å and 7.552 Å in $CeCoIn_5$, 4.652 Å and 7.542 Å in $CeRhIn_5$, and 4.668 Å and 7.515 Å in $CeIrIn_5$, respectively [2–5]. The c/a value decreases monotonically in the series Co, Rh, Ir.

Recently we clarified that the Fermi surface in $CeIrIn_5$ is quasi-two-dimensional, reflecting the unique crystal structure [6]. That is, the 5d electrons of Ir hybridize with the 5p electrons of In sited at 4i, forming bonding and anti-bonding bands, which results in a small density of states around the Fermi energy. This means that conduction electrons are scarce in the Ir layers. Quasi-two-dimensionality in the electronic structure might be a common feature of a series of these compounds because the layer of TIn_2 is closely related to low dimensionality.

We continued investigating the Fermi surface properties of $CeCoIn_5$ with the highest values of T_c and C/T for $CeTIn_5$. Correspondingly, the upper critical field H_{c2} in superconductivity is large [2–4], and there is a possibility that de Haas–van Alphen (dHvA) oscillation is observed in both the normal and superconducting mixed states. We stress in this work that the heavy-fermion state is destroyed to a remarkable extent in magnetic fields, indicating that the large cyclotron mass of about 100 m_0 for the main Fermi surface is reduced to half at 160 kOe. The detected dHvA oscillation in the mixed state will also shed light on a new aspect of the exotic superconducting state characteristic of the unique crystal structure.

Single crystals of $CeCoIn_5$ were grown by the so-called self-flux method [2–6]. The lattice parameters were $a = 4.612$ Å and $c = 7.549$ Å. The fractional coordinates of indium at the 4i site were determined as (0, 0.5, 0.305). See reference [6] for details of the crystal structure.

In order to clarify the heavy-fermion state, we carried out the dHvA experiment by the usual field-modulation method at low temperatures down to 30 mK and in high magnetic fields up to 170 kOe.

Figure 1 shows the dHvA oscillation for the field along [001] and the corresponding fast-Fourier-transformation (FFT) spectrum. Branches α_i ($i = 1, 2, 3$) and β_i ($i = 1, 2$) correspond to the main Fermi surfaces. The angular dependence of the dHvA frequency for these branches is shown in figure 2. 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. As shown in figure 2, branches α_i follow approximately a $1/\cos\theta$ dependence, indicating nearly cylindrical Fermi surfaces, where θ is the angle by which the field is tilted away from [001] towards [100] and [110]. The dHvA branches in figure 2 are well explained by the Fermi surfaces shown in figure 3, calculated on the basis of the itinerant 4f band model as for $CeIrIn_5$ [6]:

(1) branches α_i: a nearly cylindrical 15-electron band Fermi surface.
(2) branches β_i and η: a complicated 14-hole band Fermi surface, stretching along [001].

An outer orbit called c in the 14-hole band Fermi surface and two kinds of small 15-electron band Fermi surfaces centred at Z and R are, however, not observed experimentally as in $CeIrIn_5$. The identity of the other branches called γ, ε, and ζ is not clear. They might be due to ellipsoidal Fermi surfaces, although these branches are experimentally not observed in the whole angular region. Branches ε and γ might correspond to two kinds of 13-hole band Fermi surfaces centred at Γ and X points, respectively.

We will consider the Fermi surface property from another viewpoint—that based on the upper critical field H_{c2} in superconductivity. H_{c2} (=49.5 kOe) is clearly reflected as a dip in the dHvA signal, shown by an arrow in figure 1. H_{c2} is anisotropic, as shown in the inset of figure 4. Assuming that the Fermi surface is simply an ellipsoid of revolution, $H_{c2}(\theta)$ for the

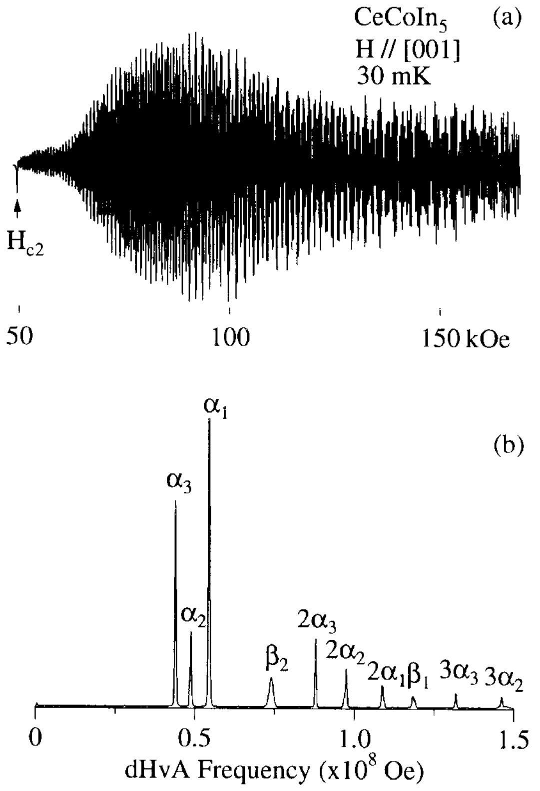

Figure 1. (a) dHvA oscillation and (b) its FFT spectrum in CeCoIn$_5$.

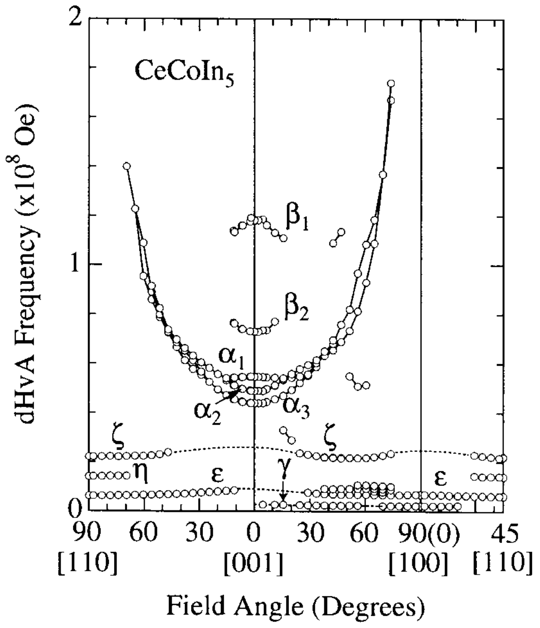

Figure 2. Angular dependence of the dHvA frequency in CeCoIn$_5$. Solid and dotted lines are guides to the eyes.

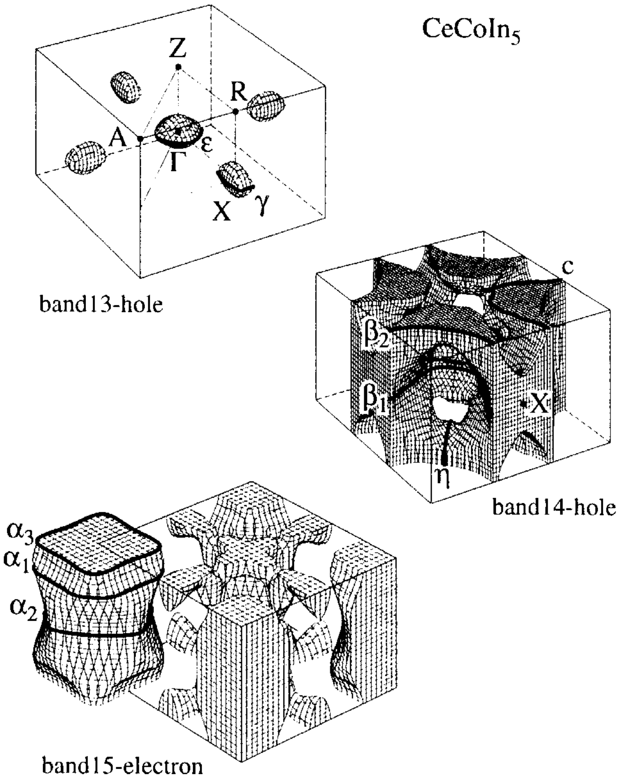

Figure 3. Fermi surfaces of CeCoIn$_5$ based on the itinerant 4f band model.

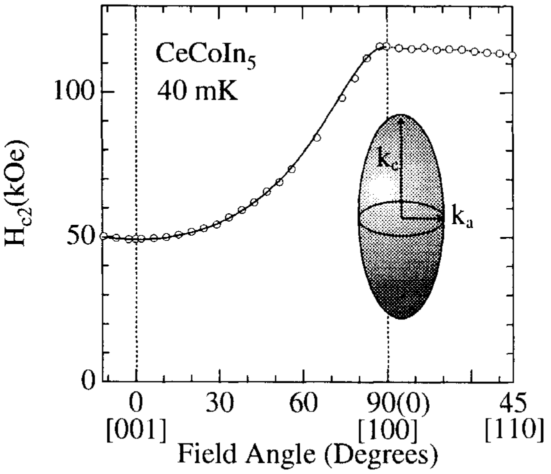

Figure 4. Angular dependence of H_{c2} at 40 mK in CeCoIn$_5$. The inset shows the corresponding Fermi surface.

field tilted away from [001] towards [100] by θ is defined as [7]

$$H_{c2}(\theta) = H_{c2}(\theta = 90^\circ) \Big/ \sqrt{\sin^2\theta + \frac{m_c^*}{m_a^*}\cos^2\theta} \qquad (1)$$

where $H_{c2}(\theta = 90^\circ) = 116$ kOe and $m_c^*/m_a^* = 5.6$. The m_c^*/m_a^* value simply corresponds to the ellipsoid of revolution with $k_c/k_a = 2.4$, where $2k_a$ and $2k_c$ are the caliper dimensions of the Fermi surface, as shown in figure 4. The subscripts a and c correspond to [100] (a-axis) and [001] (c-axis), respectively. The fitted solid line in figure 4 is in good agreement with the experimentally observed angular dependence of H_{c2}. This might simply suggest that the complicated quasi-two-dimensional Fermi surfaces in figure 3 are considered to be the ellipsoidal one in figure 4 from the viewpoint of H_{c2}. From these H_{c2}-values, the coherence length is estimated as $\xi_a = 82$ Å and $\xi_c = 35$ Å.

We determined the cyclotron mass from the temperature dependence of the dHvA amplitude in a constant field range—for example, from 150 to 169 kOe for $\boldsymbol{H} \parallel [001]$: about 50 m_0 for branches β_1 and β_2, 15 m_0 for α_1, and the largest value of 87 m_0 for the branch with 1.1×10^8 Oe, tilted by 42.5° from [001] towards [100]. A large cyclotron mass is actually detected, which corresponds to a large C/T value of about 1000 mJ K^{-2} mol^{-1}. We note that the corresponding band mass m_b is 3.48 and 1.73 m_0 for branches β_1 and β_2, respectively, and 1.78 m_0 for α_1, as shown in table 1. The ratio m_c^*/m_b is 14 and 28 for branches β_1 and β_2, respectively, and 8.4 for α_1. The corresponding ratio of the experimental electronic specific heat coefficient to the theoretical one is about 30, where the theoretical specific heat coefficient is 30 mJ K^{-2} mol^{-1}. There is a discrepancy between the mass ratio m_c^*/m_b and the ratio of the electronic specific heat coefficient to the theoretical one.

Table 1. de Haas–van Alphen frequency F and the cyclotron effective mass m_c^* at an effective field of $\bar{H} = 160$ kOe for $\boldsymbol{H} \parallel [001]$ and $\theta = 42.5^\circ$, $\bar{H} = 150$ kOe for $\boldsymbol{H} \parallel [110]$, and $\bar{H} = 45$ kOe for $\boldsymbol{H} \parallel [100]$ in CeCoIn$_5$.

	Experimental		Theoretical		
	F (10^7 Oe)	m_c^* (m_0)		F (10^7 Oe)	m_b (m_0)
$\boldsymbol{H} \parallel [001]$					
			c (14-hole band)	13.3	4.37
β_1	12.0	48	β_1 (14-hole band)	13.0	3.48
β_2	7.5	49	β_2 (14-hole band)	6.45	1.73
α_1	5.56	15	α_1 (15-electron band)	5.43	1.78
α_2	4.53	18	α_2 (15-electron band)	4.53	1.09
α_3	4.24	8.4	α_3 (15-electron band)	3.90	1.48
$\boldsymbol{H} \parallel [110]$					
ζ	2.21	33			
η	1.40	20			
ε	0.62	5.6			
$\boldsymbol{H} \parallel [100]$					
ε	0.67	12			
γ	0.23	4.3			
$\boldsymbol{H} \parallel$ 42.5° from [001] towards [100]					
	10.9	87			

The reason is as follows. The cyclotron mass is found to be field dependent, as shown in figure 5(a). Here, we determined the cyclotron effective mass in a relatively narrow field range containing 30–90 cycles of the oscillation. The cyclotron mass of branches α_i and β_i decreases with increasing the field for $\boldsymbol{H} \parallel [001]$: 30 m_0 at 51 kOe and 16 m_0 at 160 kOe for branch α_1, and 83 m_0 at 93 kOe and 49 m_0 at 160 kOe for branch β_2, as shown in figure 5(a). A cyclotron mass over 100 m_0 is expected for branches β_i at lower fields. We note that mass

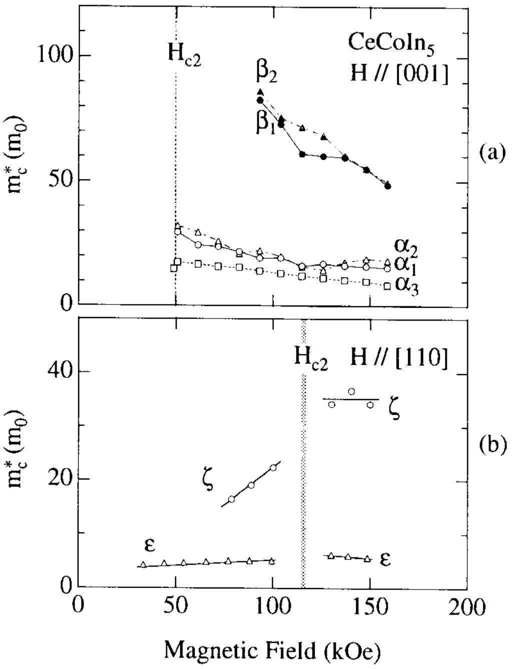

Figure 5. Field dependence of the cyclotron effective mass for the field along (a) [001] and (b) [110] in CeCoIn$_5$.

reduction due to magnetic fields is observed in heavy-fermion compounds such as CeCu$_6$; this was discussed by Chapman *et al* on the basis of the mean-field theory of the SU(N) Anderson lattice model [8].

Next we show the dHvA oscillation in figure 6 in both the normal and superconducting mixed states for the field along [001], where H_{c2} (49.5 kOe) is shown by an arrow in figure 6(a). The FFT spectra in both the mixed and normal states are shown in figures 6(b) and 6(c), respectively. In the normal state, branches α_1, α_2, and α_3 are clearly observed, while only one branch α_3 is found in the mixed state, close to H_{c2}.

On the other hand, when the field is directed along the (001) plane, we observed branches ζ, η, ε, and γ with relatively small dHvA frequencies and cyclotron masses, as shown in figure 2. As shown in figure 7, we can observe the dHvA oscillation down to low fields of 20–30 kOe in the mixed state, where H_{c2} is about 115 kOe.

The dHvA oscillation in the mixed state is observed in CeRu$_2$, UPd$_2$Al$_3$, URu$_2$Si$_2$, and a few other compounds [9–11]. This is due to quasiparticles which are produced by the pair breaking in magnetic fields. See reference [9] for details. A characteristic Fermi surface property in the mixed state is a field dependence of the cyclotron mass below H_{c2}. We found that the cyclotron mass in the mixed state for $\boldsymbol{H} \parallel [110]$ decreases with decreasing field, as shown in figure 5(b). This mass reduction is extremely large for branch ζ with a relatively large mass. On the other hand, the mass reduction is small for branch ε with a small mass. As for branch α_3 in figure 6, a slight mass reduction is found even in fields close to H_{c2}: 15 m_0 in the field range from 48.5 to 49.5 kOe (the mixed state), but 18 m_0 in the field range from 50 to 52 kOe (the normal state), as shown in figure 5(a). The reduction of the cyclotron mass with decreasing field might also be one of the characteristic properties for the heavy-fermion superconductors [9]. In the mixed state, the number of quasiparticles decreases with decreasing

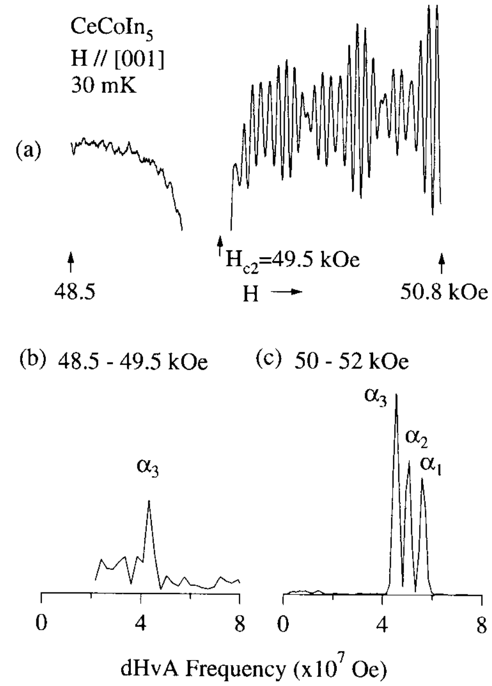

Figure 6. (a) dHvA oscillation, (b) the FFT spectrum in the field range from 48.5 to 49.5 kOe (superconducting mixed state), and (c) the FFT spectrum from 50 to 52 kOe (normal state) in $\mathrm{CeCoIn_5}$.

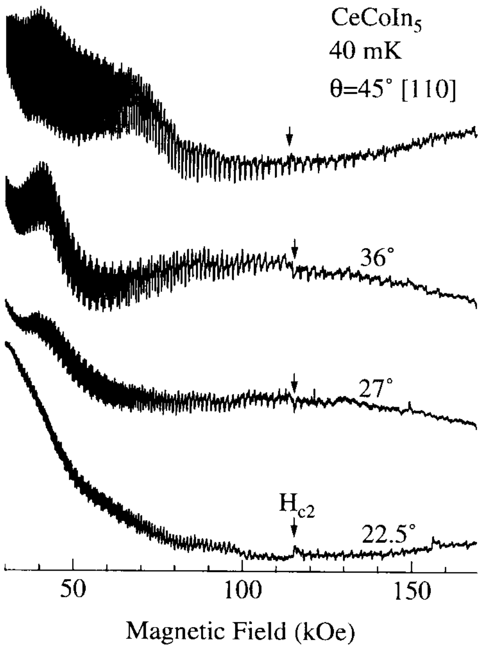

Figure 7. dHvA oscillation for several field directions in $\mathrm{CeCoIn_5}$. θ is the angle by which the field is tilted from [100] towards [110] on the (001) plane.

field below H_{c2}. Correspondingly, the correlation between quasiparticles might be weakened, which is most probably the origin of the mass reduction below H_{c2}. This is an interesting issue not fully clarified yet.

The cyclotron mass is thus strongly field dependent in both the normal and mixed states. This is a characteristic feature of $\mathrm{CeCoIn_5}$. Surprisingly, the dHvA oscillation due to branch ε in figures 5(b) and 7 is observed in low fields down to $H/H_{c2} \simeq 0.2$. On the other hand, the dHvA oscillation due to a main Fermi surface called α_i is observed in the mixed state, close to H_{c2}. The earlier theory of the dHvA effect in the mixed state is applicable near H_{c2} [10, 11]. It is questionable whether this small Fermi surface called ε has a superconducting energy gap in magnetic fields below H_{c2}. This might indicate the growth of a pair-breaking effect far below H_{c2} for this particular Fermi surface. More importantly, there is a possibility that the superconducting state for this Fermi surface is changed into a normal state far below H_{c2}. Clarifying this is left to future study.

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