QUASI-TWO DIMENSIONAL FERMI SURFACES IN RARE EARTH AND URANIUM COMPOUNDS: UX_2 , CeTIn₅ AND UTGa₅*

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We present the quasi-two dimensional Fermi surface studies in rare earth and uranium compounds such as UX_2 (X: Bi, Sb, As and P) and $RTIn_5$ (R: rare earth, T: Co, Rh and Ir), together with $UTGa_5$. The present quasi-two dimensionality is closely related to the magnetic unit cell and/or the unique crystal structure elongated along the tetragonal [001] direction, which bring about a flat Brillouin zone and produce cylindrical but highly corrugated Fermi surfaces along [001].

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1. Introduction

The f electrons of rare earth and uranium compounds exhibit a variety of characteristics including spin and valence fluctuations, heavy fermions and anisotropic superconductivity [1]. In these compounds, both the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the Kondo effect compete with each other. Competition between the RKKY interaction and the Kondo effect was discussed by Doniach in terms of a function of $|J_{cf}| D(\varepsilon_F)$, where $|J_{cf}|$ is a magnitude of the magnetic exchange interaction and $D(\varepsilon_F)$ is the electronic density of states at the Fermi energy ε_F [2]. Most of the cerium compounds order magnetically, because the RKKY interaction overcomes the Kondo effect at low temperatures. On the other hand, some cerium compounds such as CeCu₆ and CeRu₂Si₂ indicate no long-range magnetic order, forming the heavy fermion state at low temperatures.

The heavy fermion state is roughly understood as follows. The 4f levels of the Ce ions are generally split into three Crystalline Electric Field (CEF)-doublets at high temperatures because the 4f electrons in the Ce compounds are almost localized in nature. At low temperatures, the magnetic entropy of the ground-state doublet in the 4f levels, $R \ln 2$, is expressed by integrating the magnetic specific heat $C_{\rm m}$ in the form of $C_{\rm m}/T$ over the temperature. When the magnetic specific heat $C_{\rm m}$ is changed into the electronic specific heat γT via the many-body Kondo effect, the heavy fermion state is formed below the Kondo temperature $T_{\rm K}$: $\gamma = R \ln 2/T_{\rm K} \simeq$ $10^4/T_{\rm K} ({\rm mJ/K}^2 {\rm mol})$ [1,3]. In fact, the electronic specific heat coefficient γ and the Kondo temperature are 1600 mJ/K² mol and 5 K in CeCu₆, and 350 mJ/K² mol and 20 K in CeRu₂Si₂, respectively.

The de Haas-van Alphen (dHvA) effect provides a powerful tool for determining the topology of the Fermi surface, the cyclotron effective mass m_c^* and the scattering lifetime of the conduction electron. Here, the dHvA voltage $V_{\rm osc}$ is obtained in the so-called 2ω detection of the field modulation method, following the Lifshitz-Kosevich formula: [1]

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

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

$$\alpha = \frac{2\pi^2 c k_{\rm B}}{e\hbar}, \qquad (3)$$

and

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

where $J_2(x)$ is the Bessel function which depends on the dHvA frequency F,

the modulation field h and the magnetic field strength H. The dHvA frequency $F \left(=\frac{\hbar c}{2\pi e} S_{\rm F}\right)$ is proportional to the extremal (maximum or minimum) cross-sectional area $S_{\rm F}$ of the Fermi surface and $T_{\rm D} \left(=\frac{\hbar}{2\pi k_{\rm B}} \tau^{-1}\right)$ is the Dingle temperature which is inversely proportional to the scattering lifetime τ .

Recently quasi-two dimensional Fermi surfaces were observed even in rare earth and uranium compounds. This quasi-two dimensionality is closely related to the magnetic unit cell and/or the crystal structure elongated along the *c*-axis of the tetragonal or hexagonal crystal structure, which bring about a flat Brillouin zone and produce cylindrical Fermi surfaces along the *c*-axis. Typical examples are UX₂ (X: Bi, Sb, As, P) [4] and CePtX(X:As, P) [5]. Furthermore, new heavy fermion superconductors of CeIrIn₅ and CeCoIn₅ or a pressure-induced superconductor of CeRhIn₅ with the tetragonal crystal structure are also quasi-two dimensional. We present in this paper quasitwo dimensional Fermi surfaces of UX₂ and CeTIn₅ (T: transition atom), together with UTGa₅.

2. dHvA studies in quasi-two dimensional compounds

2.1. Cylindrical Fermi surfaces formed by a flat magnetic Brillouin zone in uranium dipnictides

Uranium dipnictides UX₂ (X = Bi, Sb and As) crystallize in the tetragonal structure of anti-Cu₂Sb type (D_{4h}^7 or P4/nmm), where the crystal structure of UP₂ is slightly different from that of UX₂ [6]. They order antiferromagnetically. Magnetic moments of uranium ions are aligned ferromagnetically in the (001) planes, which are stacked along the [001] direction in an antiferromagnetic ($\uparrow\downarrow$) sequence in UBi₂ [7]. In the case of USb₂, UAs₂ and UP₂, this sequence is ($\uparrow\downarrow\downarrow\uparrow\uparrow$) [7–9]. It is worth mentioning here that the magnetic unit cell of USb₂, UAs₂ and UP₂ is doubled with respect to the chemical unit cell along [001], which brings about a flat magnetic Brillouin zone. Here ordered moments and the Néel temperatures are $\mu_{ord} = 2.1$, 1.88, 1.61 and 2.0 $\mu_{\rm B}/\rm U$, and $T_{\rm N} = 183$, 203, 273 and 201 K for X = Bi, Sb, As and P, respectively [10].

First we show the results of the dHvA effect in UBi₂. Figure 1 shows the angular dependence of the dHvA frequency in UBi₂. It follows the $1/\cos\theta$ -dependence for the branch β or β' up to 81°. A solid line for these branches in Fig. 1 shows the $1/\cos\theta$ -dependence. This result indicates that the branch β or β' originates from a cylindrical but slightly corrugated Fermi surface. In contrast to the branches β and β' , the dHvA frequency of the branch α is almost constant against the field angle, meaning that the branch α originates from a nearly spherical Fermi surface. We determined the cyclotron effective



Fig. 1. Angular dependence of the dHvA frequency in UBi_2 .

mass m_c^* from the temperature dependence of the dHvA amplitude by using Eq. (2). The cyclotron mass was determined as $9.2 m_0$ for the branch α , $6.3 m_0$ for β' and $4.4 m_0$ for β . The masses are rather large because the corresponding Fermi surfaces have small cross-sections.

Next we show the dHvA results for USb₂. Figure 2 shows the angular dependence of the dHvA frequency in USb₂. It follows the $1/\cos\theta$ -dependence for branches α , γ , δ and ε . These results also indicate that branches α , γ , δ and ε originate from cylindrical but slightly corrugated Fermi surfaces. The cyclotron mass was determined as $3.8 m_0$ for the branch α , $6.0 m_0$ for γ , $3.8 m_0$ for δ and $2.0 m_0$ for ε .

The Fermi surfaces in UBi₂ consist of a cylindrical Fermi surface named β and a spherical one named α . The volume of the cylindrical Fermi surface occupies 4.8 % of the magnetic Brillouin zone, whereas the spherical Fermi surface occupies 9.9 %. As the magnetic unit cell contains two molecules of UBi₂, UBi₂ is a compensated metal with equal carrier numbers of electrons and holes. If we assume that there are one spherical hole-Fermi surface and two cylindrical electron-Fermi surfaces in the Brillouin zone, the number of carriers is well compensated, where the holes and the electrons occupy 9.9 % and 9.6 % of its volume, respectively. We show in Fig. 3 (a) a Brillouin zone and the corresponding Fermi surfaces.



Fig. 2. Angular dependence of the dHvA frequency in USb_2 .



Fig. 3. Schematic magnetic Brillouin zone and the Fermi surfaces (a) for the branches α and β in UBi₂ (b) for the branches α , γ , δ and ε in USb₂.

Furthermore we calculated the γ value from these Fermi surfaces. The estimated γ values from the branches α and β are $\gamma_{\alpha} = 8.1$ and $\gamma_{\beta} = 4.9 \,\mathrm{mJ/K^2}$ mol, respectively. The total γ value is thus $\gamma_{m_c^*} = \gamma_{\alpha} + 2\gamma_{\beta} = 18 \,\mathrm{mJ/K^2}$ mol, which is in good agreement with $20 \,\mathrm{mJ/K^2}$ mol determined from the specific heat measurement.

A very flat magnetic Brillouin zone is realized in the antiferromagnetic state of USb_2 , although the Brillouin zone in the paramagnetic state is the same as that in UBi_2 . It is thus expected that the Fermi surface in USb_2 is approximately the same as that in UBi_2 in the paramagnetic state, although it is modified in the antiferromagnetic state due to the flat magnetic Brillouin zone.

From the angular dependence of the dHvA frequency in Fig. 2, it is seen that all the Fermi surfaces are cylindrical in USb₂. The detected cylindrical Fermi surfaces occupy in the magnetic Brillouin zone 16.8% for the branch α , 7.9% for the branch γ , 5.4% for the branch δ and 3.3% for the branch ε . As the magnetic unit cell contains four molecules of USb₂, USb₂ is a compensated metal with equal carrier numbers of electrons and holes. To compensate the numbers of the carriers, we assume as follows:

one hole-Fermi surface from the branch α , one hole-Fermi surface from the branch δ , two electron-Fermi surfaces from the branch γ and two electron-Fermi surfaces from the branch ε .

The electron-Fermi surfaces thus occupy 22 % and the hole-Fermi surfaces occupy 22 %, well compensated. Figure 3 (b) shows the flat magnetic Brillouin zone and the corresponding Fermi surfaces in USb₂. The γ value is also consistent with this scheme of the Fermi surfaces.

Experimental results are summarized as follows:

- (1) The Fermi surfaces of UBi₂ are found to consist of one spherical Fermi surface and two cylindrical ones.
- (2) Each Fermi surface in UBi₂ splits into two cylindrical Fermi surfaces in USb₂, which are well explained by the band-folding procedure in a flat magnetic Brillouin zone.
- (3) The quasi-two-dimensional character of these Fermi surfaces is mainly due to the conduction electrons in the U-plane, including the 5f electrons because the cyclotron mass is large.

2.2. Quasi-two dimensional Fermi surface originated from the unique tetragonal crystal structure in RTIn₅ and UTGa₅

CeRhIn₅ orders antiferromagnetically below $T_{\rm N} = 3.8$ K, whereas superconductivity was observed under pressure, p > 1.6 GPa [11]. It was also reported that CeIrIn₅ and CeCoIn₅ are heavy fermion superconductors at ambient pressure [12–15]. The transition temperature $T_{\rm c}$ and the γ value are 0.4 K and 680 mJ/K² mol in CeIrIn₅, and 2.3 K and 300-1000 mJ/K² mol in CeCoIn₅. Here, the γ value in CeCoIn₅ is about 300 mJ/K² mol at $T_{\rm c}$ but increases with decreasing the temperature, reaching about 1000 mJ/K² mol at 0.1 K, which was obtained by the specific heat measurement in magnetic fields [15].

These characteristic properties in CeTIn₅ are closely related to the unique tetragonal crystal structure (P4/mmm #123 D_{4h}^1) with alternating layers of CeIn₃ and TIn₂ (T: Co, Rh and Ir), stacked sequentially along the [001] direction (*c*-axis), as shown in Fig. 4.



Fig. 4. Tetragonal crystal structure of RTIn₅.

First we will show the dHvA experiment for a non-4f reference compound LaRhIn₅ [16]. Figure 5(a) shows the angular dependence of the dHvA frequency. The dHvA branches α_i (i=1, 2 and 3) as well as β_2 follow roughly the 1/cos θ -dependence, where θ means a field angle tilted from [001] to [100] or [110]. This angular dependence claims that the corresponding Fermi surface is nearly cylindrical.

Figure 5(b) shows the theoretical dHvA frequency calculated in the scheme of the FLAPW method within the local-density approximation. The magnitude and angular dependence of the dHvA frequency in Fig. 5(a) are the same as theoretical ones in Fig. 5(b), except dHvA branch ε_1 .



Fig. 5. (a) Angular dependence of the dHvA frequency and (b) the theoretical one in LaRhIn₅.

These dHvA branches are well identified by the theoretical Fermi surface in Fig. 6. The dHvA branches α_i are due to a band 15-electron Fermi surface whose topology is nearly cylindrical but is highly corrugated, having maximum and minimum cross-sections. Branches β_i are also due to a highlycorrugated band 14-electron Fermi surface. This Fermi surface has a convex part stretching along the [110] direction. This is a main reason why the dHvA frequency of branch β_1 has a minimum at about $\theta = 30^\circ$, tilted from [001] to [100] or [110]. Branches ε_i are due to a band 13-hole Fermi surface, forming a network or a lattice. The orbit ε_1 in Fig. 6(a) was, however, not detected experimentally, which is most likely due to the damping of the dHvA amplitude based on a curvature factor of this Fermi surface.

Next, the dHvA experiment for CeRhIn₅ was carried out in the antiferromagnetic state [16,17]. Figure 7 shows the angular dependence of the dHvA frequency. Main three branches are named β_2 , α_1 , and α_3 . To identify these branches, we compare them to those of LaRhIn₅ and CeCoIn₅, as shown in Fig. 8. If the 4*f* electrons in CeRhIn₅ are localized and the Fermi surface



Fig. 6. Theoretical (a) band 13-hole, (b) band 14-electron and (c) band 15-electron Fermi surfaces in LaRhIn₅.



Fig. 7. Angular dependence of the dHvA frequency in CeRhIn₅.

is not affected by the small magnetic Brillouin zone, the Fermi surface in CeRhIn₅ should be the same as that of LaRhIn₅. On the other hand, if the 4f electrons are itinerant as in CeIrIn₅ and CeCoIn₅, the Fermi surface of CeRhIn₅ should be the same as that of CeCoIn₅. Note that the topology of the Fermi surface is approximately the same between CeIrIn₅ and CeCoIn₅, which were well explained by the 4f-itinerant model as shown below [18,19].

A dHvA frequency with $F=6.13 \times 10^3$ T of branch β_2 in CeRhIn₅ is the same as 6.13×10^3 T in LaRhIn₃, but is smaller than 7.35×10^3 T in CeCoIn₅. The dHvA frequencies of branches α_1 and $\alpha_{2,3}$ in CeRhIn₅ are also the same as those of LaRhIn₅, but are smaller than those of CeCoIn₅, as shown in Fig. 8.



Fig. 8. Angular dependence of main dHvA frequencies in (a) LaRhIn₅, (b) CeRhIn₅ and (c) CeCoIn₅.

From these experimental results we can conclude that the contribution of the 4f electrons to the volume of the Fermi surface in CeRhIn₅ is negligibly small. We note that there are so many dHvA branches in CeRhIn₅ compared to those in LaRhIn₅. This might be approximated by a band-folding procedure where the paramagnetic Fermi surface of CeRhIn₅, which is almost the same as that of LaRhIn₅, is folded into a small magnetic Brillouin zone based on a large magnetic unit cell, producing small Fermi surfaces. Branches β_2 , α_1 and $\alpha_{2,3}$ are most likely formed by the conduction electrons which break through the antiferromagnetic Brillouin zone boundary and circulate around the original paramagnetic Fermi surface and/or some of these branches are not affected by the magnetic Brillouin zone.

On the other hand, the cyclotron effective mass of CeRhIn₅ is large compared to that of LaRhIn₅. The cyclotron mass is 5.5 m_0 in branch β_2 , 6.0 m_0 in branch α_1 and 3.5 m_0 in branch $\alpha_{2,3}$. The corresponding mass in LaRhIn₅ is 0.73 m_0 , 0.69 m_0 and 0.51&0.64 m_0 , respectively. The ratio of the mass of CeRhIn₅ to that of LaRhIn₅ is about 7–9. On the other hand, the ratio of the γ value of CeRhIn₅ to that of LaRhIn₅ is about 9, where $\gamma \simeq 50$ mJ/K² mol in CeRhIn₅ and 5.7 mJ/K² mol in LaRhIn₅. Both ratios are approximately consistent. These experimental results are almost the same as the recent results in CeRhIn₅ [20,21]. For example, main branches β_2 and $\alpha_{2,3}$ were observed: $F=6.120\times10^3$ T ($m_c^*=6.1\pm0.3 m_0$) and $F=3.600\times10^3$ T ($m_c^*=4.6\pm1.0 m_0$), respectively.

We would like to clarify the 4f-electronic nature in CeCoIn₅. Figure 9 shows the angular dependence of dHvA frequency in CeCoIn₅. The dHvA branches in Fig. 9(a) are well identified by the 4f-itinerant band model, where Figure 9(b) shows the angular dependence of the theoretical dHvA frequency in CeCoIn₅. Main branches α_i and β_i in Fig. 9 are identified by theoretical Fermi surfaces in Fig. 10.



Fig. 9. (a) Angular dependence of the dHvA frequency and (b) the theoretical one in $CeCoIn_5$.

Fermi surfaces in CeCoIn₅ are similar to those of LaRhIn₅, although the size of each Fermi surface is different between them. A similar relation is present between the Fermi surface of Pb with four valence electrons and that of Al with three valance electrons. If one 4f-electron in each Ce site becomes a conduction electron in CeCoIn₅, the volume of the band 13-hole Fermi surface in LaRhIn₅, shown in Fig. 6(a), is shrunken, changing into two kinds of small closed Fermi surfaces in CeCoIn₅, as shown in Fig. 10(a). Correspondingly, the band 14- and 15-electron Fermi surfaces are expanded



Fig. 10. Theoretical Fermi surfaces in CeCoIn₅.

in volume. The band 14- and 15-electron Fermi surfaces in Fig. 6(b) and (c) are changed into Fermi surfaces in Fig. 10(b) and (c), respectively. This is a reason why the dHvA frequencies of β_i and α_i in CeCoIn₅ are larger than those in LaRhIn₅.



Fig. 11. (a) Temperature dependence of the specific heat $C_{\rm e}$ in the form of $C_{\rm e}/T$ under H=0, 6 and 8 T. (b) Field dependence of the cyclotron mass in CeCoIn₅.

As shown in Fig. 11, the cyclotron mass of CeCoIn₅ is extremely large. For example, the mass of β_i is about 80 m_0 at 10 T. The cyclotron mass is strongly field-dependent, where 80 m_0 at 10 T is reduced to 50 m_0 at 16 T for branch β_i [19]. A large cyclotron mass over 100 m_0 is expected at lower fields than 10 T, consistent with the specific heat coefficient in magnetic fields. Similar dHvA results are obtained in CeIrIn₅ [18].

UTGa₅ (T: transition atom) has also the HoCoGa₅-type tetragonal crystal structure as in RTIn₅. Here we present dHvA results in a series of UTGa₅, namely Pauli paramagnets of UFeGa₅ and UCoGa₅, and an antiferromagnet UPtGa₅.

We show in Fig. 12(a) the angular dependence of the dHvA frequency in UFeGa₅ [22], together with the theoretical one in Fig. 12(b). Branches α_i show roughly a 1/cos θ -dependence of the dHvA frequency. Branches b_i are disconnected in the angular dependence, indicating a multiply-connected Fermi surface. The origin of these detected dHvA branches is well explained on the basis of the theoretical Fermi surfaces in Fig. 13. Namely, branches α_i are due to a highly corrugated but cylindrical Fermi surface, and a_i and b_i are due to a lattice like structure in the band 15.



Fig. 12. Angular dependence of the (a) dHvA frequency and (b) the theoretical one in UFeGa₅.

We will compare the Fermi surface of UFeGa₅ with valence electrons of U($5f^{3}6d^{1}7s^{2}$), Fe($3d^{6}4s^{2}$) and Ga($4s^{2}4p^{1}$) to that of CeCoIn₅ with valence electrons of Ce($4f^{1}5d^{1}6s^{2}$), Co($3d^{7}4s^{2}$) and In($5s^{2}5p^{1}$). CeCoIn₅ is a compensated metal with equal volumes of electron- and hole-Fermi sur-



Fig. 13. Fermi surface of (a) band 14-holes, (b) band 15-electrons in UFeGa₅.

faces. If the band 14-hole Fermi surface in $CeCoIn_5$ is almost fully occupied by an electron and also the volume of the band 15-electron Fermi surface is slightly enlarged, these Fermi surfaces correspond to the band 14-small hole Fermi surfaces and the band 15-large electron ones, respectively, in UFeGa₅. In CeCoIn₅ we could not observe the lattice-structure like band 15-electron Fermi surface, while the corresponding Fermi surface was detected completely in UFeGa₅. We suppose that the lattice-structure like band 15-Fermi surface is not present and/or is changed into small closed Fermi surfaces in CeCoIn₅.

The 5f electrons in UFeGa₅ are highly itinerant compared to the 4f electrons in CeCoIn₅. The cyclotron mass in UFeGa₅ is relatively large: 9.2, 4.6 and 8.0 m_0 for α_i (*i*=1, 2 and 3), respectively.

In UCoGa₅ we observed dHvA branches with small dHvA frequencies, as shown in Fig. 14. This means that UCoGa₅ is a semimetal. The Fermi surface most likely consists of small band 15-hole Fermi surfaces and small band 16-electron Fermi surfaces, because one more valence electron is added, compared to that in UFeGa₅. In fact, a volume of one ellipsoidal Fermi surface (branch *a*) is equal to a volume of two pieces of ellipsoidal Fermi surfaces (branch *b*) and four pieces of ellipsoidal Fermi surfaces (branch *c*): $V_a = 2V_b + 4V_c$ (V_a =0.0059 $V_{\rm BZ}$, V_b =0.0019 $V_{\rm BZ}$ and V_c =0.0005 $V_{\rm BZ}$), where $V_{\rm BZ}$ is a volume of the Brillouin zone.

Finally we show in Fig. 15 the angular dependence of the dHvA frequency in an antiferromagnet UPtGa₅ with $T_{\rm N}=25$ K and an ordered moment of $0.25 \,\mu_{\rm B}/\rm{U}$. Most of the dHvA branches indicate the $1/\cos\theta$ -dependence, claiming cylindrical Fermi surfaces. The magnetic unit cell of UPtGa₅ is doubled with respect to the chemical unit cell along [001], which brings about a flat magnetic Brillouin zone as in USb₂. This is a main reason on the existence of the cylindrical Fermi surfaces in UPtGa₅. The cyclotron mass is in the range of 10 to 25 m_0 for [001], reflecting the γ value of 57 mJ/K² mol.



Fig. 14. Angular dependence of the dHvA frequency in UCoGa₅.



Fig. 15. Angular dependence of the dHvA frequency in UPtGa₅.

3. Concluding remark

High-quality single crystals, low temperatures and strong magnetic fields are fundamentally necessary to demonstrate the dHvA measurement for the strongly correlated electron systems. At present, the carrier with a large cyclotron mass over 100 m_0 is detected in the dHvA experiment for CeRu₂Si₂, UPt₃ and CeCoIn₅, reflecting the γ value of 57 mJ/K² mol.

Quasi-two dimensionality in rare earth and uranium compounds is closely related to the magnetic unit cell and/or the unique crystal structure as well as the corresponding electronic state. In UX₂, the conduction electrons are 5f, 6d and 7s electrons in the U-plane. It is expected that these conductive U-planes are separated by the nearly non-conductive X-planes, bringing about the quasi-two dimensional electronic state. The magnetic unit cell, doubled with respect to the chemical unit cell along [001], enhances two dimensionality because the unit length along [001] in the Brillouin zone becomes half and correspondingly each Fermi surface is band-folded in the flat Brillouin zone.

The similar quasi-two dimensionality is also realized in the unique tetragonal crystal structure of RTIn₅ and UTGa₅. It was demonstrated that the topology of the Fermi surface in UFeGa₅ is well explained by the 5f-itinerant band model, compared to that in CeCoIn₅. This indicates that the 5felectrons are itinerant as the 3d electrons, while the 4f electrons become itinerant via the many-body Kondo effect at low temperatures. A different mechanism between them results in a different cyclotron effective mass between them.

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