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De Haas-van Alphen effect in ytterbium and uranium compounds

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Abstract

We have measured the de Haas-van Alphen (dHvA) effects of $YbSn_3$, $YbZn_{11}$ and UGa_2 in the ^{4}He temperature range up to 15 T. The results for $YbSn_3$ show that the dHvA frequencies range from 60 to 3000 T and the cyclotron masses from $0.13m_0$ to $0.44m_0$, where m_0 is the free electron mass. For $YbZn_{11}$ the dHvA frequencies range from 200 to 11 000 T and the cyclotron masses are nearly the same as those of $YbSn_3$. These results indicate that $YbSn_3$ is a compensated metal and Yb ions in $YbSn_3$ and $YbZn_{11}$ are divalent. The dHvA frequencies of UGa_2 range from 20 to 1300 T. The observed cyclotron masses range from $0.3m_0$ to $3.3m_0$, which are larger than those of $YbSn_3$ and $YbZn_{11}$ by a factor of ten. Field dependence of the dHvA frequency is observed in the lowest frequency oscillations in UGa_2. These results indicate that the 5*f* electrons in UGa_2 hybridize appreciably with the conduction electrons. © 1998 Elsevier Science S.A.

Keywords: De Haas-van Alphen effect; Fermi surface; Cyclotron mass; $YbSn_3$; $YbZn_{11}$; UGa_2

1. Introduction

De Haas-van Alphen (dHvA) effect is one of the most useful tools for investigating dynamical properties of electrons near the Fermi level. It allows, not only to determine the shape of the Fermi surface, but also to estimate electron effective mass and electron life time. Until now much attention of the dHvA effect has been paid to light rare-earth compounds, particularly Ce compounds that show anomalous physical phenomena, such as mixed valency or heavy fermion properties due to interaction between 4*f* electrons and conduction electrons (see for example [1]). Yb compounds are also particularly interesting because the electron–hole symmetry exists between Ce and Yb compounds. In Yb compounds, 4*f* holes interact with conduction electrons; this interaction leads to valence instability and thus results in the same phenomena as above. However, the dHvA effect has been measured on only a few Yb compounds such as $YbIn_3$ [2], $YbSn_3$ [3,4], YbAs [5] and $YbGa_2$ [6], until now. Thus, we have aimed to measure the dHvA effect on various Yb compounds to study the effect of hybridization between 4*f* holes and conduction electrons. In this work we report on the results for $YbSn_3$ and $YbZn_{11}$.

Uranium compounds also show a variety of behavior including local magnetic order, Pauli itineracy, spin fluctuation and heavy fermion. UGa_2 is known as a localized ferromagnet with the Curie temperature T_c=125 K and as one of the most interesting U compounds with non-transition metal. However there has been controversy about whether the 5*f* electrons are localized or delocalized in UGa_2 among the experimental results such as magnetization [7], neutron diffraction [8] and photoemission spectroscopy [9]. In the present work, we have measured the dHvA effect in UGa_2 to characterize the 5*f* electrons as well as to study the Fermi surface properties using the dHvA effect.

2. Experimental

Single crystals of $YbSn_3$ and $YbZn_{11}$ were grown from metallic fluxes [10]. The purity of Yb was 99.99% and 99.999% for Sn or Zn. In the growth of $YbSn_3$, the starting material of Yb:Sn in an atomic ratio of 1:9 was sealed into a quartz ampoule under high vacuum. The ampoule was heated to 750°C and cooled gradually to 600°C for 2–4 days. For $YbZn_{11}$, the starting material of Yb:Zn=4:96 was used and the temperature was changed slowly from 750°C to 500°C for 2–3 days. UGa_2 single crystals were made by tri-arc Czochralski method from high purity U (99.99%) and Ga (99.9999%). The crystal structures of these compounds were checked by an X-ray powder

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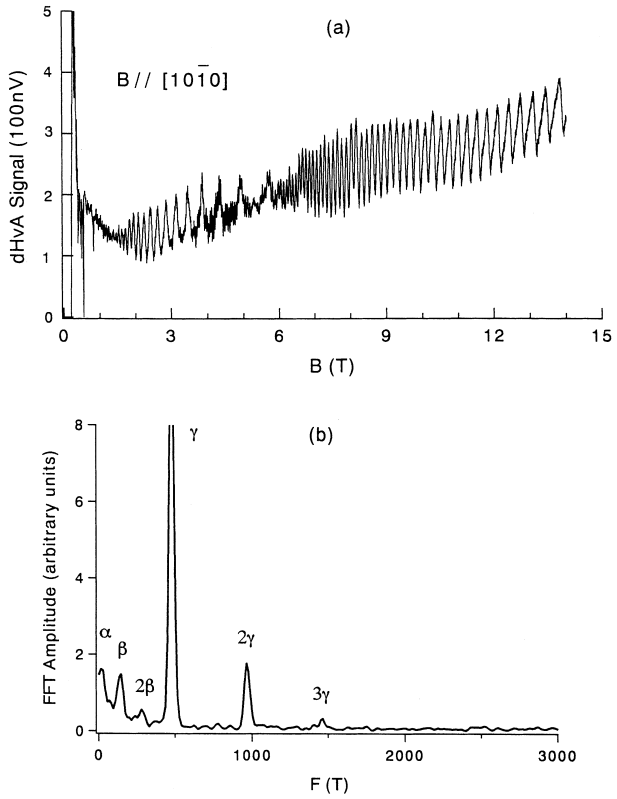

Fig. 1. (a) Typical dHvA oscillations of UGa_2 for the [10$\bar{1}$0] direction. (b) FFT spectrum of the oscillations.

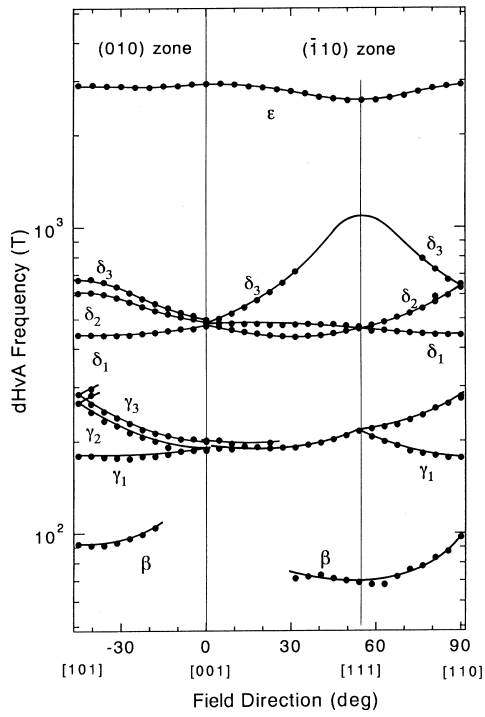

Fig. 2. Variation of the dHvA frequencies with direction of the magnetic field for $YbSn_3$.

diffraction method with Cu $K\alpha$ radiation. The residual resistivity ratios of the crystals were greater than 150. This indicates that the crystals have a sufficient quality to be used as samples for measurements of the dHvA effect. The dHvA effect was measured in the temperature range 1.5–4.2 K up to 15 T by a modulation method. The details of the measurement are described in [11]. In Fig. 1(a) the example of a recorder trace of dHvA oscillations measured in UGa_2 at 1.5 K with a magnetic field B parallel to the [10$\bar{1}$0] direction is shown. An FFT spectrum of these oscillations is shown as a function of the dHvA frequency F in Fig. 1(b). The spectrum contains three fundamental frequencies denoted by α, β and γ, and their higher harmonics 2β, 2γ and 3γ.

3. Results and discussion

3.1. $YbSn_3$

X-ray diffraction of powdered $YbSn_3$ crystals presented sharp Cu_3Au reflection lines with traces of β-Sn reflection lines. But the $YbSn_3$ phase was found not to be stable in air: when the powder was exposed to air for several days, the diffraction pattern revealed that $YbSn_3$ seems to change into β-Sn, together with Yb_2O_3, by a reaction with oxygen in air.

In Fig. 2 the angular dependence of the dHvA frequencies in the (010) and ($\bar{1}$10) zones is shown. The detected frequencies range from 150 to 3000 T. The highest one corresponds to 16% of the square base of the Brillouin zone. The frequency variations are well described by four groups of branches denoted β, γ, δ and ε. The δ_3 branch is tentatively depicted around the [111] direction.

Since the branch ε forms a continuous line and shows very slight variation with the field orientation, the Fermi surfaces of the ε branch is expected to be nearly a sphere and to be centered on the Γ point of the Brillouin zone. The δ group of the branches is composed of three lines. They meet each other in the [001] direction, while they seem to form a singlet and a doublet in the [111] and [110] directions. This behavior suggests that the Fermi surfaces of the δ branch group are on the $\langle 111 \rangle$ axes of the Brillouin zone and are composed of at least eight small pockets. Each pocket should have form similar to a tetrahedron with its trigonal-axis parallel to the axis on which the pocket is placed. From this Fermi surface model we can explain qualitatively why the higher frequency branch, the δ_3 branch, could not be observed around the [111] direction by taking into account the curvature factor of this Fermi surface [11]. For the γ group, we can speculate that the Fermi surfaces are on the $\langle 100 \rangle$ axes of the Brillouin zone.

The volumes of the Fermi surfaces corresponding to the

branch groups ε, δ and γ are estimated to be 4.5%, 0.32% and 0.08% of the Brillouin zone, respectively. The carrier concentration deduced from these values is very low and equal to at most 0.1 per formula unit. The cyclotron masses measured with $B//[101]$ are listed in Table 1 together with the dHvA frequencies F. They range from $0.19m_0$ to $0.44m_0$ and are nearly the same as those of La compounds (see for example [11]). From these observations we conclude that $YbSn_3$ is a compensated metal with Fermi surfaces arising from the slightly overlapped electron and hole bands, and that the Yb ion in $YbSn_3$ is divalent. This valence value is consistent with the magnetic measurement of $YbSn_3$ by [12].

3.2. $YbZn_{11}$

X-ray diffraction analysis ascertained that $YbZn_{11}$ crystals used in the present study have the $BaCd_{11}$ tetragonal structure with space group $I4_1/amd$.

In Fig. 3 the angular dependence of the dHvA frequencies in the (010) and $(\bar{1}10)$ zones is shown. The detected frequencies range from 150 to 11 000 T. The θ_1 frequency corresponds to 60% of the Brillouin zone base area. Noteworthy features of the results are: (i) the branch η_1 disappears around the [001] direction; (ii) the disappearance is also seen in the θ_1 and θ_2 branches; (iii) instead, the branches ε_1, ε_2 and ε_3 appear around that direction. These features suggest that the Fermi surfaces corresponding to η_1, θ_1 and θ_2 are multiply connected through the Brillouin zone boundaries. The Fermi surfaces corresponding to other branches such as α_i, β_i, δ and ζ_i are expected to be closed.

The cyclotron masses measured with $B//[100]$ are presented in Table 1. The m_c^* values of $YbZn_{11}$ are nearly the same as those of $YbSn_3$. This indicates that the Yb ion in $YbZn_{11}$ is divalent. This divalency has been predicted by [13] from the measurements of lattice parameters and magnetization.

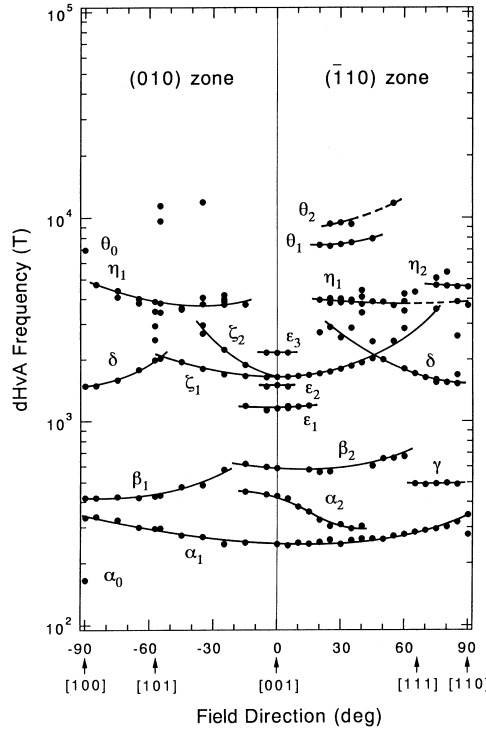

Fig. 3. Variation of the dHvA frequencies with direction of the magnetic field for $YbZn_{11}$.

3.3. UGa_2

X-ray diffraction examination ascertained that UGa_2 crystals used in the present study have the AlB2 hexagonal structure with space group $P6/mmm$.

In Fig. 4 the angular dependence of the dHvA frequencies in $(1\bar{2}10)$ and (0001) zones is shown. The

Table 1
Cyclotron effective masses m_c^* of $YbSn_3$, $YbZn_{11}$ and UGa_2

$YbSn_3$ $B//[101]$		$YbZn_{11}$ $B//[100]$		UGa_2 $B//[0001]$		$B//[10\bar{1}0]$	
F (T)	m_c^*/m_0	F (T)	m_c^*/m_0	F (T)	m_c^*/m_0	F (T)	m_c^*/m_0
						32	0.17
179	0.19	158	0.14				
266	0.29						
		343	0.19				
443	0.24	422	0.29			478	0.82
607	0.40						
				1132	2.3		
				1341	3.3		
		1480	0.44				
				1780	3.0		
2896	0.44						
		6958	0.73				

m_0 is the electron mass and F (T) the dHvA frequency in units of tesla.

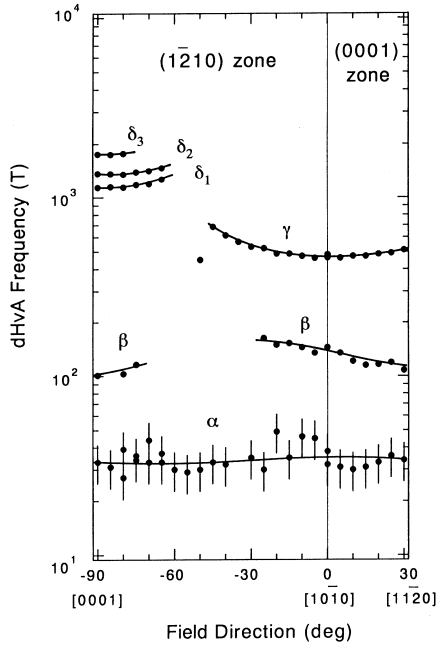

Fig. 4. Variation of the dHvA frequencies with direction of the magnetic field for UGa_2.

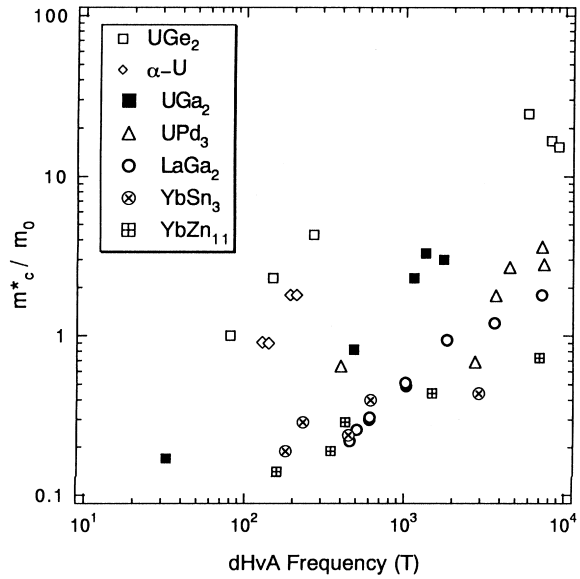

Fig. 5. Cyclotron mass m_c^* in units of the free electron mass m_0 as a function of dHvA frequency.

observed frequencies range from 30 to 1300 T. The highest one corresponds to 5% of the hexagonal base of the Brillouin zone. Frequencies larger than 1300 T were not observed in the present experiments. The dependence of the δ_i and γ branches on the field direction are found to be very similar to those previously observed in $LaGa_2$ [11] which has the same crystal structure as UGa_2. The δ_i and γ branches in UGa_2 correspond to the ε and γ branches of $LaGa_2$, respectively. Thus we expect that the Fermi surfaces corresponding to these branches are similar to that of $LaGa_2$.

The cyclotron masses observed with $B//[0001]$ are listed in the 3rd and 4th columns of Table 1. The observed masses range from $0.2m_0$ to $3.3m_0$ and are larger than those of $YbSn_3$ and $YbZn_{11}$ by a factor of 4 to 10. The cyclotron masses versus the dHvA frequency are plotted in Fig. 5. For comparison, we have plotted the cyclotron masses of UGe_2 [14], UPd_3 [15], α-U [16], $LaGa_2$ [11] and Yb compounds of this work in the figure, where UGe_2 is known as a heavy fermion compound, UPd_3 as a ferromanget with localized 5*f* electrons, α-U as a metal with delocalized 5*f* electrons and $LaGa_2$ as a normal metal without *f* electrons. We can see that the cyclotron masses of UGa_2 are about twice as large as those of UPd_3. This indicates that 5*f* electrons hybridize with the conduction electrons appreciably and have delocalized characteristics. This picture is consistent with the observation by the photoemission spectroscopy [9] that 5*f* valence band emission exists near the Fermi level. However the effect of hybridization between 5*f* and conduction electrons in UGa_2 is less pronounced than in UGe_2 and α-U.

There is a noteworthy finding about the field dependence of dHvA frequencies. In Fig. 6 the field dependence of the α frequency observed with $B//[10\bar{1}0]$ at 1.5 K in UGa_2 is shown. The frequency increases with increasing B and tends to saturate to 35 T at higher fields. The γ frequency shows the similar field dependence at this direction. We consider this behavior as further evidence that 5*f* electrons hybridize with conduction electrons for the following reason. Magnetostriction of UGa_2 is known to be so large that UGa_2 undergoes a lattice distortion from the hexagonal to the orthorhombic structures below $T_c=125$ K [7]. Magnetic fields also induce a lattice distortion that affects 5*f* electron states of U ion in crystalline electronic fields. If the 5*f* electrons interact noticeably with the conduction electrons, the band structure of the conduction electrons is altered by the distortion. This results in a change in the Fermi surface size and thus the dHvA frequency. Therefore, we conclude that 5*f* electrons are partly delocalized, which leads to large mass enhancement and the field dependence of dHvA frequency.

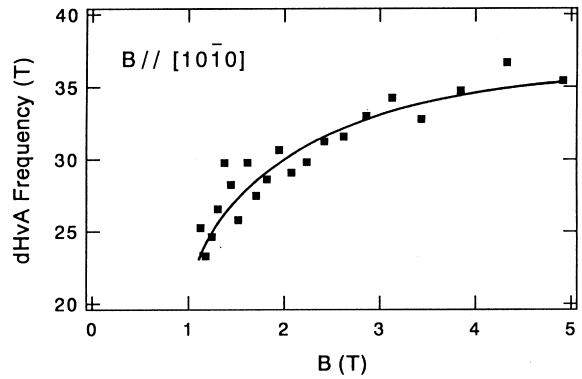

Fig. 6. Magnetic field dependence of the α frequency for UGa_2.

In summary, we have measured the dHvA effect in $YbSn_3$, $YbZn_{11}$ and UGa_2. From the branch diagrams of the dHvA oscillations, some topological features of the Fermi surface are drawn. The values of cyclotron masses suggests that Yb in YbSn3 and $YbZn_{11}$ are divalent and the $5f$ electrons in UGa_2 hybridize with conduction electrons appreciably.

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