Transition of f Electron Nature from Itinerant to Localized: Metamagnetic Transition in $CeRu_2Si_2$ Studied via the de Haas–van Alphen Effect

H. Aoki and S. Uji
National Research Institute for Metals, 2-3-12 Nakameguro, Meguro-ku, Tokyo 153, Japan
A. K. Albessard and Y. Ōnuki
Institute of Materials Science, University of Tsukuba, Tsukuba, Ibaraki 305, Japan
(Received 22 April 1993)

We report the measurements of the de Haas–van Alphen (dHvA) effect for field ranges below and above the metamagnetic transition field (H_m) in $CeRu_2Si_2$. The dHvA frequency branches and the effective masses ranging from $1.5m_0$ to $120m_0$ observed below H_m agree with the predictions of the itinerant f electron model, whereas those above H_m can be explained well with the localized f electron model. All the dHvA frequencies change abruptly around H_m. The effective masses decrease considerably around H_m and then continue to decrease with increasing field.

PACS numbers: 71.28.+d, 71.25.Hc, 75.20.Hr

The interesting and anomalous properties of the heavy fermion compounds (HFC) originate from the highly correlated f electrons which reside near the borderline between localized and itinerant. The nature of the f electrons is a fundamental issue to be clarified for discussing the physics of HFC's.

The properties of HFC's are sensitive to external variables such as pressure and magnetic field. Most HFC's are known to exhibit metamagnetic transition (MT) in fields. Since the MT of $CeRu_2Si_2$ takes place at an accessible field in laboratories [about 7.7 T (H_m) for the field direction parallel to [001]], it is a very suitable compound to study the MT and the interplay among interactions involved in HFC's. It crystallizes in the $ThCr_2Si_2$ type structure and the linear term of the specific heat amounts to 350 mJ/moleK^2 [1,2]. Neither superconductivity nor magnetic order has been found down to 20 mK. The thermal, electrical, and other physical properties [3–6] have been found to change drastically across the MT, implying that the MT is closely correlated with the electronic structure change. In this paper we will report a dramatic change of the Fermi surface (FS) properties associated with the MT and particularly the transition of the $4f$ electron nature from itinerant to localized.

The de Haas–van Alphen (dHvA) effect was measured and analyzed by the fast Fourier transform separately for the fields below and above H_m. For the details of the experimental method and the sample preparation, refer to our papers [6,7]. Figure 1 shows the dHvA frequencies and their angular dependences in the (010) plane. The open circles denote the frequency branches observed for the fields below H_m and the closed triangles those above H_m. The frequencies thus obtained are "average frequencies" over the field ranges used for the Fourier analyses, because the dHvA frequencies have been found to change with field, particularly for the fields near H_m. The field dependences of the frequencies and effective masses for the [001] direction will be described later. The frequencies and the effective masses (m^*) in the high and low field limits of the present measurements are listed in Table I.

The oscillations observed below H_m are classified in three categories from the point of view of m^*, i.e., the β and γ oscillations with small m^* of the order of $(1-2)m_0$, α and κ with those of the order of $(10-20)m_0$, and ψ exceeding $100m_0$. The α, β, and γ oscillations are the same as those previously reported by Lonzarich [8] and by us [6]. The κ and ψ oscillations have been observed for the first time in the present measurements. The observation and the details of the mass determination of ψ have already been reported elsewhere [7].

For the fields above H_m, four frequency branches, i.e., β', γ', δ, and ω are observed. The β' and γ' oscillations obviously correspond to the β and γ oscillations below H_m

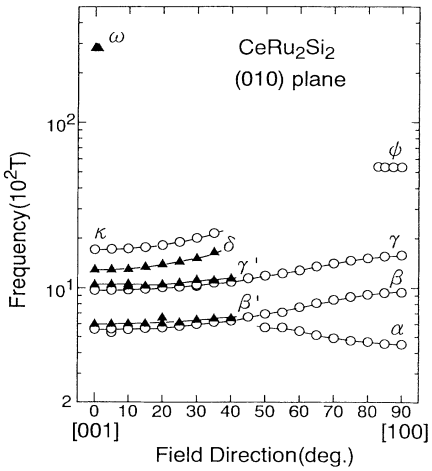

FIG. 1. Angular dependence of the dHvA frequencies in the (010) plane. The open circles and closed triangles denote the frequency branches observed for the fields below and above H_m, respectively.

0031-9007/93/71(13)/2110(4)$06.00

TABLE I. Frequencies and effective masses determined in the high and low field limits of the present measurements for fields parallel to the [001] direction. In the parentheses are given the field strengths where the measurements were made.

$H < H_m$	$H > H_m$
Frequencies (T)	
β: 5.36×10^2 (3 T)	β': 6.13×10^3 (13.1 T)
γ: 9.8×10^2 (5.4 T)	γ': 1.04×10^3 (12.5 T)
κ: 1.70×10^3 (5–7 T)	δ: 1.24×10^3 (12.6 T)
ψ: not measured (5.36×10^3 [100])	ω: 2.82×10^4 (15.65–15.9 T)
Effective masses (m_0)	
β: 1.5 (3 T)	β': 0.97 (13.1 T)
γ: 1.6 (5.4 T)	γ': 1.1 (12.5 T)
κ: 20 (5.65–7.1 T)	δ: 3.6 (12.6 T)
ψ: not measured (120 [100])	ω: 8.2 (15.65–15.9 T)

and have slightly higher frequencies and smaller m^* than those of β and γ, respectively. Although no trace of δ has been observed for the fields below H_m, a relatively strong signal of it is observed above H_m. The angular dependence of the δ frequency is very similar to that of the κ frequency. The ω oscillation is observed for the first time in the present measurements. The signal intensity of the ω oscillation rapidly decreases with the tilting angle from the [001] direction and could be observed only for the field directions within 1° from the [001] direction.

The observed frequency branches below H_m agree well with the predictions of the band structure calculations based on the itinerant f electron model [9,10]. The band structure calculations predict three ellipsoidal hole surfaces (HS) centered at the Z point, a large spherical HS centered at the Z point, and a multiply connected electron surface (ES). The β and γ oscillations are attributed to the two of the three ellipsoidal HS's [6,8–10]. The small m^* observed are consistent with those predicted by the renormalized band structure calculation and reflect the small f contents of these FS's.

Judging from the frequencies, their angular dependences, and the relatively large m^*'s of the α and κ oscillations, they are assigned to the orbits on the multiply connected ES. The ψ oscillation is assigned to the orbit on the large HS centered at the Z point [7]. The m^* of $120m_0$ for [100] is consistent with the prediction of the renormalized band structure calculation [9] and reflects the rich f content of the large HS.

On the other hand, the frequency branches above H_m can be explained very well by the localized f electron model. There are no band structure calculations of the localized f electron model for $CeRu_2Si_2$. However, both the band structure calculation [11] and the dHvA effect measurements [6] have been performed for the reference material $LaRu_2Si_2$. According to these studies, the three ellipsoidal HS's similar to those of $CeRu_2Si_2$ below H_m also exist in $LaRu_2Si_2$. The β' and γ' oscillations can be attributed to these HS's. The multiply connected ES shrinks and becomes disconnected. However, a similar orbit to that of κ remains in $LaRu_2Si_2$ as an inner orbit of a ringlike ES centered at the Γ point. The δ oscillation can be assigned to this orbit.

The HS centered at the Z point is much larger than that below H_m of $CeRu_2Si_2$. The frequency of 2.72×10^4 T along the [001] direction in $LaRu_2Si_2$ is close to that of ω, i.e., 2.82×10^4 T. The signal amplitude of the oscillation from the large HS in $LaRu_2Si_2$ has been found to decay very rapidly as the field direction is tilted from the [001] direction [6] because of the so-called curvature factor [12]. This is also consistent with the observation that the ω oscillation could be observed only near the [001] direction. The ω oscillation can be attributed to this FS.

The m^*'s observed above H_m are larger than those of $LaRu_2Si_2$, but comparable to those of $CeRu_2Ge_2$ [13] whose $4f$ electrons are found to be localized and which has nearly the same FS topology as that of $LaRu_2Si_2$.

It has been shown that the electronic structures are different between the field ranges below and above H_m. However, one to one correspondences between the orbits on the FS's below and above H_m can be made as described above. To see how the $4f$ electrons change their nature from itinerant to localized as a function of field, the frequencies and effective masses for the sets of the dHvA oscillations $\beta-\beta'$, $\gamma-\gamma'$, $\kappa-\delta$, and for the ω oscillation have been measured separately as a function of field for the [001] direction.

Before presenting the field dependences, we will briefly describe a characteristic feature of the dHvA oscillations in $CeRu_2Si_2$ which affects the analysis and interpretation of the observed effective masses and frequencies. Figure 2 shows the recorder traces of the β oscillations at various temperatures. It is noted that the amplitude of the oscillation changes with field and takes a minimum at about 6.3 T for 70 mK. Such field dependence of the amplitude is observed for other oscillations for the fields near H_m and can be understood in terms of field dependent effective masses as follows.

Since each Landau level splits into two levels of the up and down spin electrons due to the Zeeman effect, the phases of the dHvA oscillations from the up and down spin electrons differ from each other by $|\pi(g_\uparrow m_\uparrow^* + g_\downarrow m_\downarrow^*)/2m_0|$, where g_σ and m_σ^* are the g factor and effective mass of the up ($\sigma=\uparrow$) or down spin ($\sigma=\downarrow$) electrons, respectively. The observed dHvA oscillation is the sum of the oscillations from the up and down spin electrons, and therefore the total amplitude depends on the phase difference. When the m^* changes considerably as a function of field as will be shown in Fig. 4, the phase difference and consequently the total amplitude change with field. Particularly when the phase difference becomes $|\pm\pi+2n\pi|$ (n is an integer) and both the amplitudes are equal, the total amplitude becomes zero (spin

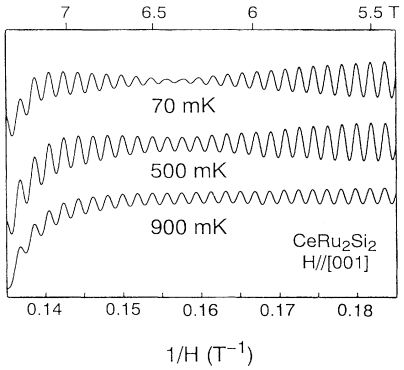

FIG. 2. Recorder traces of the β oscillations for the fields parallel to the [001] direction and below H_m. The temperatures are 70 mK (top), 500 mK (middle), and 900 mK (bottom), respectively. The vertical scale is different for each temperature. The dHvA oscillations were detected with a constant modulation field of 0.009 T and were transferred to a low frequency filter.

splitting zero [12]). However, it is noted that the amplitude becomes minimum rather than zero, and that the minimum becomes less obvious at higher temperatures while the minimum position stays nearly at the same field. Within the framework of the conventional Lifshitz-Kosevich formula [12], this observation means that the oscillation amplitudes from the up and down spin electrons are not equal and depend differently on temperature, i.e., that the m^*'s of the up and down electrons are different. Then, the effective masses and the frequencies determined by the conventional method are averages of those of the up and down spin electron oscillations. It can also be shown that other effects such as FS topology, a bicrystal structure, the band splitting of the up and down spin electrons, and the Bessel-zero effect [12] are not responsible for the observed field and temperature dependence of the amplitude by carefully examining the amplitude, the wave shape, and the Fourier spectra as functions of field, field direction, modulation field strength, and temperature.

Figure 3 shows the field dependences of the dHvA frequencies. The dHvA frequencies at each field of the β, β', γ, γ', and δ oscillations have been measured from the recorder traces by employing a low frequency filter. Since the measured frequencies are found to change considerably around H_m, the frequency in Fig. 3 was determined from the measured frequency by using the calculation procedure described in Refs. [14] and [15]. The data points near the minima are omitted from the calculation because a spurious frequency change takes place due to the phase change at the minimum position. The frequencies of the κ and ω oscillations were determined by the Fourier analyses. The ψ oscillation for the [001] direction could not be observed, probably because of the extremely high effective mass [7,9]. The broken lines are guides to the eye and the frequency of the ψ oscillation is the prediction of the band structure calculation [10]. The dHvA frequencies are nearly constant at low fields and change abruptly around H_m. Then they seem to change slightly with increasing field near H_m and again become constant at higher fields.

Figure 4 shows the effective masses of the dHvA oscillations as a function of field. The data points near the minima are omitted from the figure, because the temperature dependences of the amplitudes near the minima are

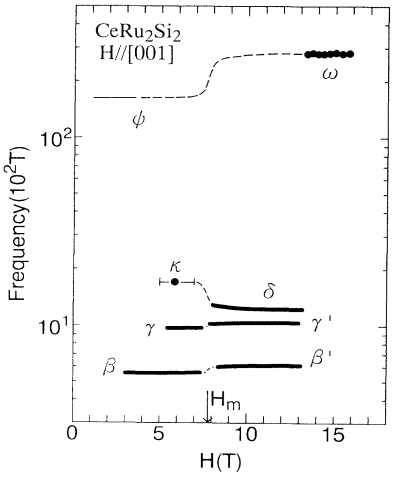

FIG. 3. Field dependences of the dHvA frequencies. The broken lines are guides to the eye. The error bar for the κ oscillation denotes the field range used for the Fourier analysis to determine the frequency. The field range used for the ω frequency is about as large as the symbol size.

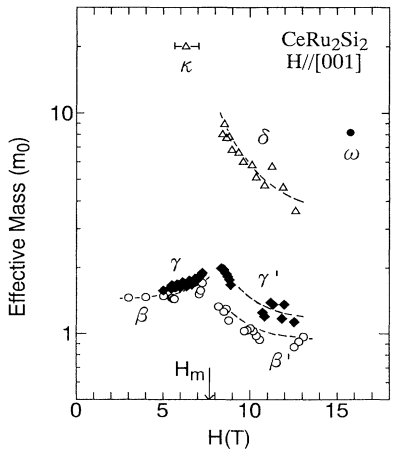

FIG. 4. Field dependences of the effective masses. The broken lines are guides to the eye. The error bar for the κ oscillation denotes the field range used for the Fourier analyses. The field range used for ω is as large as the symbol size.

such as shown in Fig. 2 that unphysically small m^* are derived there. The m^*'s are nearly constant at low fields and are enhanced around H_m, and then decrease with increasing field. The overall field dependences of the m^*'s are quite similar to that of the specific heat [5].

It is noted that the degree of the change in the frequency or the effective mass around H_m depends on the f content of the FS; i.e., the change is moderate for $\beta-\beta'$ and $\gamma-\gamma'$ of the ellipsoidal HS's, larger for $\kappa-\delta$ of the ES, and huge for $\psi-\omega$ of the HS. This observation is consistent with the assignments of the FS's below and above H_m and also supports that the nature of the $4f$ electrons changes from itinerant to localized around H_m. The dramatic change of the electronic structure observed via the dHvA effect is also consistent with the drastic changes in magnetic, electrical, thermal, and other physical properties around H_m [3–6].

Finally, we will compare the present observations with similar observations reported for other HFC's. The suppressions of the effective masses reported for CeB_6 [16] and $CeCu_6$ [17] are similar to those observed above H_m. The f electrons in CeB_6 are found to be localized. On the other hand, it is reported for $CeCu_6$ that the dHvA results observed above H_m ($=2$ T) are not in satisfactory agreement with the band structure calculations based on either the localized or itinerant f electron models [17], although we believe that at least in Ce compounds the f electrons will be localized at a sufficiently high field. The suppressions of the effective masses in these compounds were explained by the theory of the dHvA effect based on the periodic Anderson Hamiltonian [18]. This theory may be applied also to $CeRu_2Si_2$ at higher fields than H_m. For URu_2Si_2 the magnetic changes at the MT were successfully analyzed based on the destruction of the c-f mixing and the recovery of the localized magnetic moment [19]. On the other hand, for UPt_3 it is reported that the f electrons are itinerant and heavy electrons are present for the field ranges both below and above H_m [20], although many features of the physical properties around H_m are similar to those of $CeRu_2Si_2$ [21]. At present, the evolution of the f electron nature with field and the associated changes in the physical properties appear to be different for each HFC. Further experimental as well as theoretical studies are necessary to clarify whether or not the mechanism of the metamagnetic transition in HFC's can be described in a unified picture.

We thank Professor A. Hasegawa, Dr. H. Yamagami, Dr. K. Sato, and Dr. T. Shimizu for helpful discussions. One of us (Y.O.) acknowledges financial support from the Ministry of Education, Culture and Science and from Tsukuba Project Research.

[1] M. J. Benus *et al.*, Solid State Commun. **55**, 779 (1985).
[2] J. D. Thompson *et al.*, Solid State Commun. **56**, 169 (1985).
[3] P. Haen *et al.*, J. Low Temp. Phys. **67**, 391 (1987).
[4] C. Paulsen *et al.*, J. Low Temp. Phys. **81**, 317 (1990).
[5] H. P. van der Meulem *et al.*, Phys. Rev. B **44**, 814 (1991).
[6] Y. Ōnuki *et al.*, J. Phys. Soc. Jpn. **61**, 960 (1992).
[7] H. Aoki *et al.*, J. Phys. Soc. Jpn. **61**, 3457 (1992).
[8] G. G. Lonzarich, J. Magn. Magn. Mater. **76&77**, 1 (1988).
[9] G. Zwicknagl *et al.*, Physica (Amsterdam) **163B**, 97 (1990).
[10] H. Yamagami and A. Hasegawa, J. Phys. Soc. Jpn. **62**, 592 (1993).
[11] H. Yamagami and A. Hasegawa, J. Phys. Soc. Jpn. **61**, 2338 (1992).
[12] D. Shoenberg, *Magnetic Oscillations in Metals* (Cambridge Univ. Press, Cambridge, 1984).
[13] C. A. King and G. G. Lonzarich, Physica (Amsterdam) **171B**, 161 (1991).
[14] J. M. van Ruitenbeek *et al.*, J. Phys. F **12**, 2919 (1982).
[15] T. I. Sigfusson *et al.*, J. Phys. F **14**, 2141 (1984).
[16] W. Joss *et al.*, Phys. Rev. Lett. **59**, 1609 (1987).
[17] S. Chapman *et al.*, J. Phys. Condens. Matter **2**, 8123 (1990).
[18] A. Wasserman *et al.*, J. Phys. Condens. Matter **1**, 2669 (1989).
[19] K. Sugiyama *et al.*, J. Phys. Soc. Jpn. **59**, 3331 (1990).
[20] S. R. Julian *et al.*, Phys. Rev. B **46**, 9821 (1992).
[21] See, for example, H. P. van der Meulen *et al.*, Phys. Rev. B **41**, 9352 (1990), and references therein.