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Heavy fermions survive the metamagnetic transition in UPd_2Al_3

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The electronic structure above the metamagnetic transition (H_m) in UPd_2Al_3 is investigated by means of the de Haas–van Alphen (dHvA) effect. Two dHvA frequencies, 1270 and 3000 T, are observed for $H \| [100]$. The cyclotron effective masses are found to be $5.4m_0$ and $31m_0$, respectively. The heavy mass of the latter branch indicates that the heavy-fermion state persists above H_m. The angle dependence of both frequencies is determined and compared with the dHvA frequency branches below H_m. [S0163-1829(97)50720-X]

Metamagnetic transitions in heavy-fermion compounds such as $CeRu_2Si_2$, UPt_3, URu_2Si_2, and UPd_2Al_3 are one of the central issues in the current study of heavy-fermion physics. Those transitions clearly differ from classical metamagnetic transitions associated with spin reorientation in localized magnetic moment systems. They are strongly related to the fact that the heavy-fermion state sits close to magnetic instability and thus study of them may bring us a deeper understanding of the heavy-fermion state.

The metamagnetic transition and accompanying changes in the electronic structure have been investigated fairly well in $CeRu_2Si_2$ and UPt_3. The former having a nonmagnetic nonsuperconducting ground state exhibits a metamagnetic transition at $H_m = 7.7$ T for the magnetic field parallel to the c axis of the tetragonal structure,[1] while the latter, in which an antiferromagnetic order ($T_N = 5.0$ K) with a small magnetic moment of $0.03\mu_B$ and superconductivity ($T_c = 0.55$ K) coexist, shows a metamagnetic transition at $H_m = 20$ T for the field in the hexagonal basal plane.[2] The electronic specific heat coefficient γ shows an enhancement at H_m in both compounds. However, γ above H_m exhibits quite contrasting behavior between two compounds. The coefficient γ in $CeRu_2Si_2$ decreases down to about 20% of the zero-field value (350 mJ/mole K^2) at 20 T above H_m,[3] while γ in UPt_3 remains almost as large as the zero-field value (450 mJ/mole K^2) above H_m.[4] de Haas–van Alphen (dHvA) effect studies of $CeRu_2Si_2$ carried out by Aoki *et al.* showed that the Fermi-surface topology below and above H_m can be explained by band-structure calculations based on an itinerant and a localized f-electron model, respectively.[5] The cyclotron effective masses are largely reduced above H_m: the heaviest mass above H_m is only about $10m_0$ compared with $120m_0$ below H_m, m_0 being the free-electron mass. Accordingly, Aoki *et al.* have claimed a transition of the f-electron nature from itinerant to localized at H_m. The Fermi surface in UPt_3 below H_m studied by the dHvA effect is fairly well described by an itinerant f-electron band-structure calculation.[6] Shubnikov–de Haas effect measurements above H_m have confirmed a topological change in the Fermi surface.[7] However, in contrast to $CeRu_2Si_2$, heavy effective masses exceeding $100m_0$ are observed both below and above H_m in line with almost the same value of γ below and above H_m.[7] In the case of URu_2Si_2, little is known about the metamagnetic transition because of high transition fields. It shows three-step metamagnetic transitions around 40 T for the magnetic field parallel to the tetragonal c axis.[8] On the basis of a detailed analysis of the magnetization process, Sugiyama *et al.* argued that the heavy-fermion state was destroyed above the metamagnetic transitions.[8] However, the lack of an electronic-structure study such as the dHvA effect or specific-heat measurements leaves the nature of the metamagnetic transitions in URu_2Si_2 an open question.

Thus an individual interpretation of the metamagnetic transition has been proposed for $CeRu_2Si_2$, UPt_3, and URu_2Si_2 for the present. To get more insight into the metamagnetic transition in heavy-fermion compounds and to draw a unified picture of the underlying physics, which we expect to exist, it is helpful to explore the metamagnetic transition in another compound. UPd_2Al_3 perfectly fits this purpose: it is one of the most intensively studied uranium-based heavy-fermion compounds and its metamagnetic transition is relatively easy to access. UPd_2Al_3 is an antiferromagnetic heavy-fermion superconductor ($T_N = 14$ K, $T_c = 2.0$ K) with a moderately large γ of 150 mJ/mole K^2.[9] A well-developed ordered moment of $0.85\mu_B$ is noticeable compared with the small moment in UPt_3.[10] A sharp metamagnetic transition takes place at $H_m = 18$ T for the magnetic field in the hexagonal basal plane, the easy plane of the magnetization.[11] The magnetization jump at H_m amounts to $0.92\mu_B$. A detailed dHvA effect study below H_m was carried out by Inada *et al.*[12,13] and their results are in good agreement with band-structure calculations treating the $5f$ electron as itinerant.[14] However, the electronic structure above H_m has not been investigated so far by either the dHvA effect or specific heat. In this paper, we present the first dHvA measurements of UPd_2Al_3 above H_m. Two dHvA

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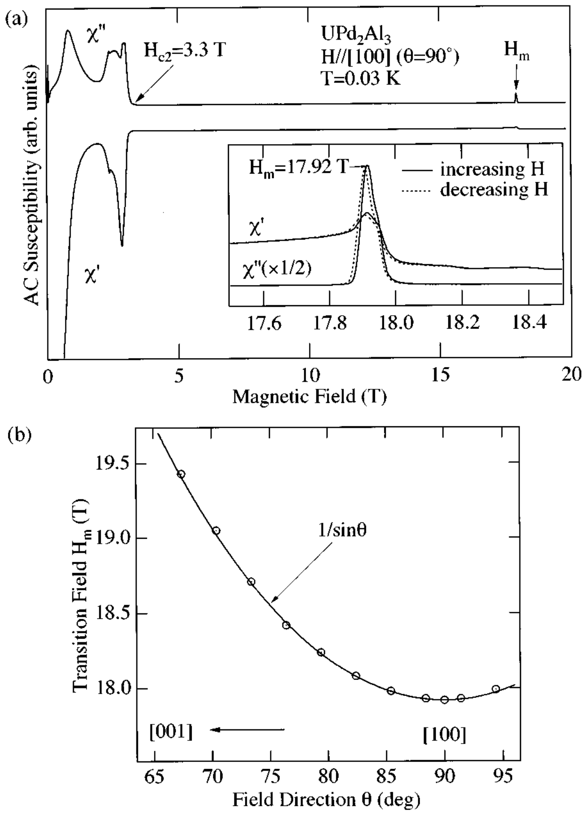

FIG. 1. (a) ac susceptibility of UPd_2Al_3 measured at 0.03 K with the magnetic field parallel to [100]. (b) Angle dependence of the metamagnetic transition field H_m.

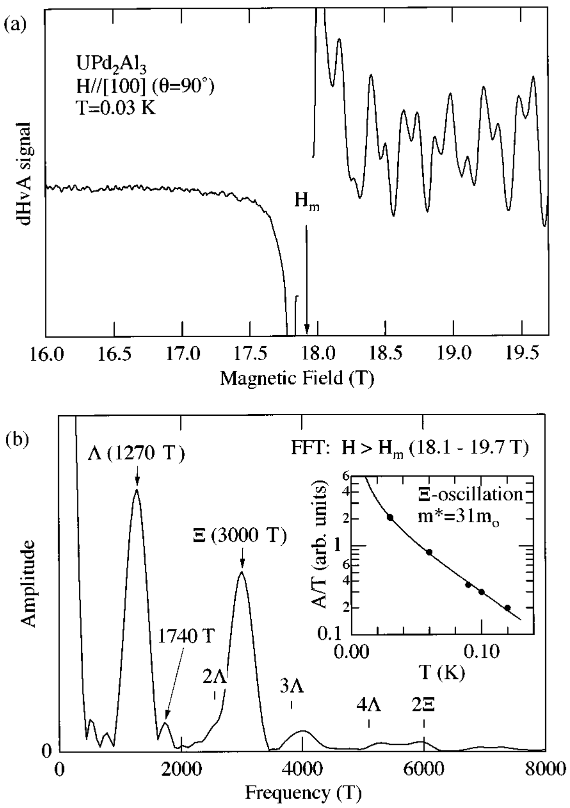

FIG. 2. (a) dHvA signal measured at 0.03 K with the magnetic field parallel to [100]. (b) Fourier transform of the signal above H_m and mass plot for Ξ (inset).

frequency branches are successfully observed above H_m. One of the branches is found to have a heavy effective mass of $31m_0$, indicating that the heavy-fermion state survives the metamagnetic transition.

The single crystal used in this study was grown in a tri-arc furnace using the Czochralski pulling method as described in Ref. 15. The dHvA effect measurements down to 0.03 K and up to 20 THz were carried out with a dilution refrigerator and a superconducting magnet. The standard field modulation technique was used, and the dHvA signal was detected at the fundamental frequency or the second harmonic of the modulation frequency. The former corresponds to usual ac susceptibility. The magnetic field was rotated from [001] (c^* axis) to [100] (a^* axis) in the reciprocal space and the field direction θ is measured from [001].[16]

Figure 1(a) shows the ac susceptibility at 0.03 K with the magnetic field applied parallel to [100] in the basal plane. The upper critical field H_{c2} of the superconductivity was estimated to be 3.3 THz from the point where the susceptibility deviates from the high-field straight line. The present H_{c2} value ranks relatively high among previous reports,[15,17] indicating good sample quality. Complex features appearing between about 2 THz and H_{c2} are ascribed to the peak effect. The metamagnetic transition is observed at $H_m = 17.92$ T, where the imaginary part shows a sharp peak. Despite the large change in the magnetic moment associated with the transition, the transition width is fairly narrow, less than 0.2 THz, and the hysteresis is very small, about 0.01 T. As the field is tilted from [100] to [001], the metamagnetic transition field varies as $1/\sin\theta$ [Fig. 1(b)]. This indicates that only the in-plane component of the magnetic field is effective to cause the metamagnetic transition.

Figure 2(a) shows the dHvA signal measured at 0.03 K with the field parallel to [100]. While no dHvA oscillation can be discerned below H_m, clear oscillations appear above H_m. The Fourier transform of the signal above H_m shows two distinct fundamental frequencies Λ (1270 THz) and Ξ (3000 T) with their harmonics [Fig. 2(b)]. A small peak at 1740 T may also be a fundamental frequency. The cyclotron effective masses of Λ and Ξ were determined from the temperature dependence of the oscillation amplitude. The determined masses are $5.4m_0$ and $31m_0$ for Λ and Ξ, respectively. The latter value is comparable to the heaviest mass of $45m_0$ observed below H_m.[13] We also examined the field dependence of the dHvA frequencies and effective masses though the field range available is less than 2 T (18.1–19.7 T). The field dependence in this field range was found to be smaller than the experimental error, which is estimated to be about 3% and 6% for the frequency and mass, respectively.

The absence of dHvA oscillations below H_m is consistent with the previous paper, in which Inada *et al.* noted that the dHvA oscillation amplitude is strongly damped as the field is tilted from [001] to the basal plane because of increasing Dingle temperature (the Dingle temperature is a measure of electron scattering).[13] Indeed we could not detect dHvA oscillations below H_m for field directions within 20° of [100]. Inada *et al.* suspected that the increasing Dingle temperature might be ascribed to some structural imperfection that pre-

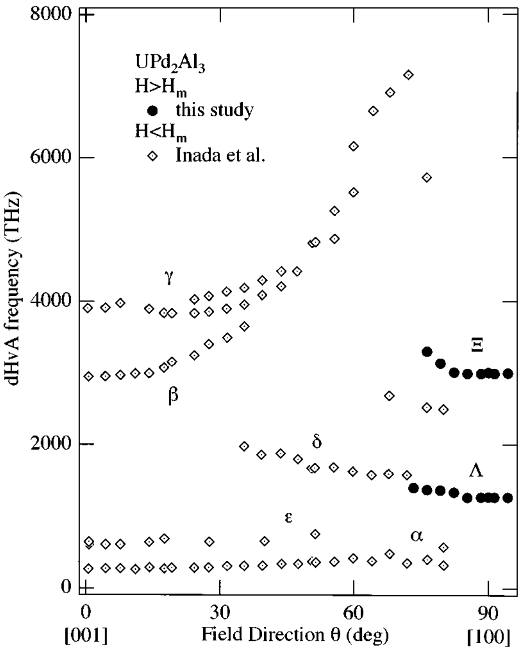

FIG. 3. Angle dependence of the dHvA frequency branches. The branches observed below H_m are taken from Inada *et al.* (Ref. 13).

vents cyclotron orbiting perpendicular to the basal plane.[13] However, since the dHvA oscillation clearly shows up above H_m, which indicates a substantial suppression of electron scattering above H_m, the increased Dingle temperature below H_m should be of magnetic origin. It is likely that electron scattering due to some magnetic fluctuation, e.g., fluctuation of moment direction or length, is quenched above H_m. The fact that the c-axis magnetoresistance shows a sharp and large (~50%) drop at H_m may be related to the suppression of electron scattering.[18]

We show the angle dependences of the two dHvA frequency branches Λ and Ξ above H_m in Fig. 3 though the data are limited in a narrow range of field directions close to [100] because of the increasing H_m. The frequency branches below H_m reported by Inada *et al.*,[13] which have also been confirmed at a few field directions in the present study, are shown together for comparison. The Ξ branch above H_m has no counterpart below H_m, thus providing direct evidence of modification of the Fermi surface at H_m. In the case of the Λ branch, judging from the frequency, it might correspond to the δ branch below H_m. However, the effective mass of δ is rather heavy, i.e., $33m_0$ at $\theta\approx50°$, while that of Λ for $H\|[100]$ is $5.4m_0$. Thus the correspondence between the frequencies is probably an accidental one.

Our results clearly indicate that, although the shape of the Fermi surface is modified, the heavy-fermion state still persists above H_m. In this sense, the metamagnetic transition in UPd_2Al_3 resembles that in UPt_3. A naive interpretation of the results could be assuming that the $5f$ electrons remain itinerant above H_m and attributing the metamagnetic transition to an antiferromagnetic to ferromagnetic transition within the framework of band magnetism. This interpretation was previously suggested by Sandratskii *et al.* on the basis of the band-structure calculations.[14] They showed that, as far as spins lie in the basal plane, the difference in the energy between the antiferromagnetic and ferromagnetic spin configurations are very small. However, apparently, it is too early to draw a definite conclusion about the nature of the metamagnetic transition in UPd_2Al_3. It is necessary to extend dHvA effect measurements to a wider range of field directions. Magnetic-field-dependent band-structure calculations will also be very helpful.

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