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Fermi surface of UPt_3 from 3 to 30 T: Field-induced quasiparticle band polarization and the metamagnetic transition

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The magnetic-field dependence of the Fermi surface of UPt_3 has been studied up to 30 T from 17 to 150 mK using the quantum oscillatory magnetoresistance. Near the 20-T metamagnetic transition, rapid changes in the frequency spectrum indicate strong nonlinear magnetic splitting of the quasiparticle bands; weaker nonlinear splitting is seen at all fields. Above the transition the mass enhancement remains very large, and masses exceeding $100m_e$ have been observed. Below the transition a surface with mass $170m_e$ has been found.

The enormous magnetic susceptibility of the heavy fermion metals means that large magnetic polarizations can be induced by laboratory scale magnetic fields. For example, UPt_3 below 1 K (Ref. 1) is a Pauli paramagnet up to $H = 17$ T, and over this range the magnetic moment per uranium atom increases linearly from 0 to $\sim 0.3\mu_B$. Above 17 T the moment grows even faster, as the differential magnetic susceptibility, $\chi(H) = dM(H)/dH$, shows an upturn that culminates in a sharp peak at 20 T: the so-called metamagnetic transition.[2] The induced moment thus reaches $\sim 0.6\mu_B$ per uranium atom at 22 T,[3] a polarization comparable to that of ferromagnetic nickel.

A central question is whether the induced magnetization can be described in terms of a difference between the up- and down-spin Fermi surface volumes. This is the case, for example, with the spontaneous magnetization of iron at $T = 0$,[4] but not with gadolinium, where most of the moment arises from localized f electrons, which polarize the conduction bands by exchange.[5] At low fields the f electrons in UPt_3 are delocalized,[6,7] in which case large field-induced spin splitting of the bands should occur. Once the system is polarized, Luttinger's theorem can break down, however, and non-Fermi liquid magnetic degrees of freedom may appear accompanied by a change in the total Fermi surface volume.

We note that neither the underlying mechanism of the metamagnetic transition nor the nature of the high-field state are understood although several theories exist.[8] One of the major stumbling blocks is uncertainty over the role of quasiparticle band polarization in the transition.

In this paper we present measurements of the quantum oscillatory magnetoresistance of UPt_3 *both above and below the metamagnetic transition.* We have found compelling evidence that the quasiparticle bands are subject to strong nonlinear spin splitting at the metamagnetic transition. Furthermore, we have observed nonlinear spin splitting of the Fermi surface *when $M(H)$ is itself linear* (i.e., far from the metamagnetic transition), suggesting that the Pauli paramagnetism of heavy fermion metals is not a straightforward phenomenon. Finally, our measurements confirm by direct observation, that above the metamagnetic transition the polarized quasiparticles remain very massive, the largest mass seen above 20 T being $110m_e$. These findings stem from observations of the large, heavy, *thermodynamically dominant* Fermi surface sheets in the system, a condition which must be satisfied before rigorous comparisons with either thermodynamic measurements or band structure and many-body calculations can be made. We note that these conditions have not yet been reached in other heavy fermion systems with field-induced transitions.[2,9]

Magnetoresistance oscillations [sometimes called the Shubnikov–de Haas (SdH) effect] are produced by modulation of the density of states as Landau levels pass through extremal cross sections of the Fermi surface. When an extremal cross section A_σ is field *independent* the modulation is periodic in $1/H$ with frequency $F_\sigma = (\hbar/2\pi e)A_\sigma$,[10] but a central feature of our measurements is the observation of field *dependence* of the Fermi surface, in which case the experimentally measured quantity is the "projected" frequency[11] $F_{p\sigma}(H) = F_\sigma(H) - H\,dF_\sigma(H)/dH + \cdots$. In order to illustrate this important technical point, we show in Fig. 1 the field dependence of the frequencies for a very simplistic model of the magnetization process in UPt_3 in which the difference between the up- and down-pseudospin Fermi surface extremal areas, $A_\uparrow(H) - A_\downarrow(H)$, is proportional to $M(H)$. Evidently, $F_{p\sigma}$ may be very different from F_σ. When F_σ varies linearly with H, $F_{p\sigma}$ is constant. When F_σ has upward (downward) curvature, $F_{p\sigma}$ *falls (rises)*, the rise or fall being disproportionately large when H is large. The insensitivity to linear changes in the Fermi surface with field means that we are unable to compare directly the induced moment with the band splitting, so that supporting computational Fermi surface modeling will be required in order to test for localized f electron

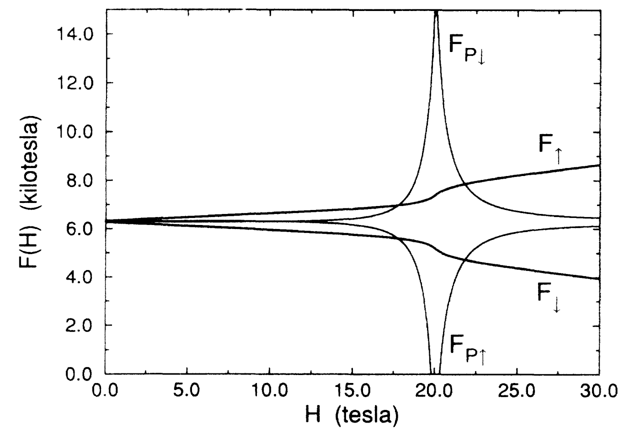

FIG. 1. Variation with field of the "instantaneous" frequency $F_\sigma(H)$, compared to that of the measured frequency $F_{p\sigma}(H)$, for a simplistic model of band magnetism in which the difference in up- and down-pseudospin extremal areas is proportional to the magnetization. Note that the larger surface can produce the smaller measured frequency.

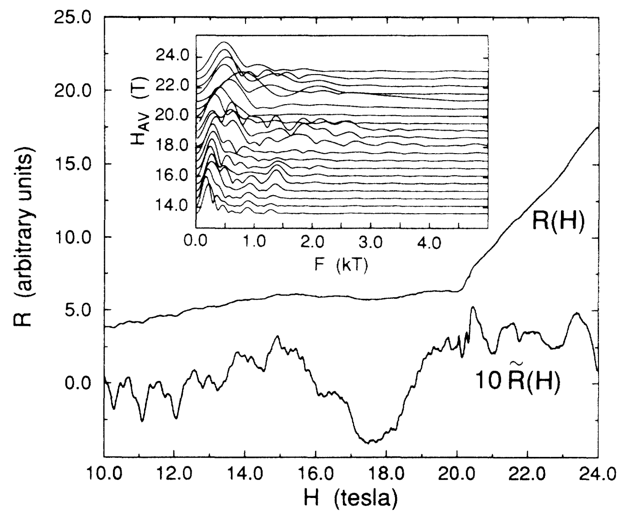

FIG. 2. Variation from 10 to 24 T of the magnetoresistance before $[R(H)]$ and after $[\tilde{R}(H)]$ subtraction of the nonoscillatory background. Inset: the corresponding variation with field of the frequency spectrum. The spectra are the Fourier transforms (in $1/H$) of consecutive, overlapping, 2-T wide sections, centered on H_{av}. A striking modification of the spectrum is seen as the transition is approached. The field is 22° from the a axis in the basal plane, at $T \approx 60$ mK.

magnetic degrees of freedom.

Our results consist of transverse magnetoresistance measurements on ultrapure single-crystal whiskers of UPt_3. The magnetic field was applied in the basal plane of the hexagonal structure, and the current was parallel to the c axis. Measurements at temperatures between 17 and 100 mK, and fields from 0 to 14.4 T, were made at the Cavendish Laboratory using a top-loading dilution refrigerator and superconducting magnet system specially designed for low-noise quantum oscillation experiments. Data at temperatures between 30 and 150 mK, from 10 to 30 T, were taken at the Nijmegen High Field Magnet Laboratory. The sample was cooled by a plastic dilution refrigerator, which was placed into a ^{4}He cryostat mounted in a 30-T hybrid magnet. To vary *in situ* the angle of the field with respect to the crystal axes the sample was mounted below the mixing chamber on a rotating platform, controlled by a wire from the top of the system. The sample was heat sunk with a silver wire to a piece of sintered silver powder in the mixing chamber.

The behavior of the SdH frequencies near 20 T supports the notion that the bands split nonlinearly at the metamagnetic transition. In Fig. 2 a typical magnetoresistance scan from 10 to 24 T is shown, with the corresponding quantum oscillation spectrum as a function of field. Below 16 T the oscillation frequencies show only weak field dependence (discussed below). But as the metamagnetic transition is approached a radical modification occurs, as the oscillations first weaken near 17 T, and then reappear with a completely different spectrum which contains higher frequencies and is strongly field dependent. This modification is reproducible, and is much too large to be due to the contribution of the nonlinear magnetization to the internal field, since $\mu_0 M(H)$ changes by only about 0.03 T across the transition. While some of this modification may arise from "magnetic breakdown,"[10] the natural explanation of the high frequencies seen near the transition is back projection of nonlinearly changing extremal areas (as per $F_{p\downarrow}$ in Fig. 1), compelling evidence that the bands split nonlinearly near the transition. This implies that theoretical models of the heavy fermion metamagnetic transition cannot ignore the magnetism of the quasiparticle bands, whether or not additional magnetic degrees of freedom are assumed to be present. Furthermore, from these data it is clear that polarization of the Fermi surface associated with the metamagnetic transition is spread out over a range of several tesla, although a further stepwise change at 20 T cannot at this stage be ruled out.

The quasiparticle masses derived from our measurements show that heavy quasiparticles survive into the high-field state. Figure 3 shows typical magnetoresistance data between 20 and 30 T with the field parallel to the b axis, along with the corresponding Fourier spectrum. The effective mass associated with each frequency was determined from the temperature dependence of the oscillation amplitudes using the Lifshitz-Kosevich formula,[10] thus assuming that the system is a Fermi liquid. The peak at 5.0 kT has an effective mass of $(110 \pm 30)m_e$, which is one of the largest ever observed. All of the masses, both in Fig. 3 and on the a axis,[12] show that the quasiparticle mass renormalization has a similar strength above the transition as it has below (masses below the transition are given in Ref. 6), *confirming that the high-field state is a heavy electron liquid,* in which most or all of the large linear specific heat observed above 20 T (Ref. 13) can be explained in terms of massive charged Fermion quasiparticles.

In Fig. 4 the dependence of the frequency spectrum on field orientation in the basal plane is shown, from which topological features of the Fermi surface can be determined. Figure 4(a) shows the clearest features above the metamagnetic transition, while for comparison Fig. 4(b)

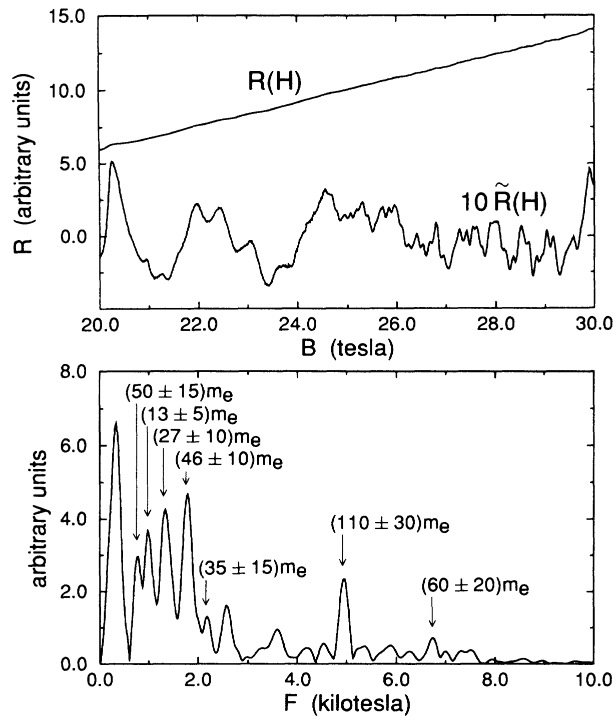

FIG. 3. Variation from 20 to 30 T of the magnetoresistance before [$R(H)$] and after [$\tilde{R}(H)$] subtraction of the nonoscillatory background, with the field parallel to the b axis. The frequency spectrum for $25 < H < 30$ T is also shown, with the effective mass given for those prominent peaks which are not harmonics. The weak, unlabeled peaks appear only at the high field end of the scan, and we believe them to be harmonics of the larger peaks, or possibly "quantum interference" (Ref. 14) orbits.

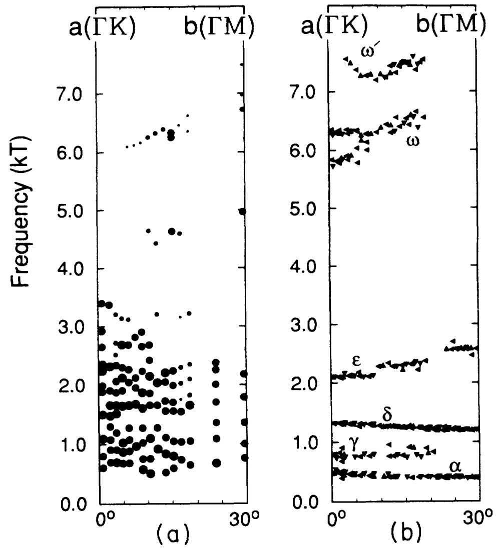

FIG. 4. Variation of the fundamental SdH frequencies with field orientation in the basal plane: (a) for $25 < H < 30$ T, at $50 < T < 100$ mK, (b) for $H < 14$ T and $T = 17$ mK. Some simplification has been achieved by including only those peaks that appear over a range of adjacent angles. In (a) the size of each point reflects the intensity of the peak, the smallest points arising from weak signals that appear only near 30 T. The frequency branch marked ω in (b) appears to survive unchanged into the high-field state. The branch marked ω', not seen in a previous de Haas–van Alphen study (Ref. 6), has a mass of $(170 \pm 35)m_e$—the largest value found in any system to date. We believe the ω and ω' branches to be the spin-split components of one Fermi surface sheet.

shows those below. The high-field data are more difficult to interpret, perhaps because variations in the projected frequency at these fields may be more the result of variations in $H\,dF(H)/dH$ than in $F(H)$, but also simply because more peaks appear above the transition than below. This is likely due to spin splitting of the Fermi surface (as in Fig. 1, where what appears to be one frequency below the transition is resolved into two frequencies above), although some peaks may be due to magnetic breakdown. Nevertheless, several features are clear. A frequency starting at 1.5 kT on the a axis rises gradually to 1.65 kT on the b axis. Several strong frequencies appear between 2.0 and 2.5 kT and persist across most of the diagram. Furthermore, a strong peak at 5.0 kT is seen on the b axis (also shown in Fig. 3) and a series of weaker peaks, rising from 6.2 to 6.5 kT, is observed as the field is rotated away from the a axis. This latter feature matches the band marked ω on Fig. 4(b), implying that this frequency may be unaffected by the metamagnetic transition, a remarkable result. Calculation of the field-dependent band structure is required to determine if the observed frequencies arise from magnetic splitting of, or magnetic breakdown between, sheets which retain the topology of the low-field Fermi surface, or whether a topological rearrangement or, more radically, a change in volume, of the Fermi surface has occurred.

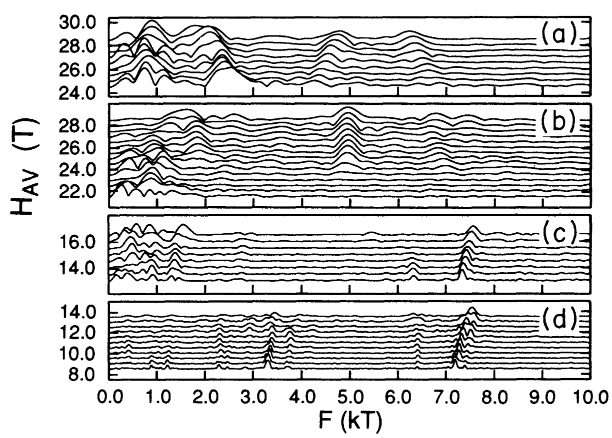

FIG. 5. Field dependence of the SdH frequencies in UPt_3 in field regions where $M(H)$ varies linearly with H. Each spectrum is from a 2-T-wide interval centered on H_{av}. The shifts are due to nonlinear change of the corresponding extremal area with field. The conditions for these scans were (a) $23 < H < 30$ T, $T = 70$ mK, H 18° from the a axis; (b) $20 < H < 30$ T, $T = 70$ mK, H parallel to the b axis; (c) $12 < H < 17.5$ T, $T \approx 40$ mK, H 10° from the a axis; (d) $7 < H < 14.4$ T, $T = 17$ mK, H 12° from the a axis.

Finally, we present evidence of nonlinear magnetic splitting of the quasiparticle bands at fields *where $M(H)$ is itself varying linearly with H.* This is a new phenomenon, distinct from the nonlinearities seen near 20 T, because there $M(H)$ is itself nonlinear.

Figures 5(a) and 5(b) illustrate the field dependence of the highest frequencies seen above the metamagnetic transition. Some frequencies show a clear field dependence [for example, the 2.5-, 4.5-, and 6.5-kT peaks in Fig. 5(a)], while others [the 5.0-kT frequency in Fig. 5(b)] do not.

Figures 5(c) and 5(d) show field-dependent frequencies below 20 T. The peak which rises from 7.2 to 7.5 kT in Fig. 5(d) corresponds to the ω' frequency in Fig. 4(b)—a frequency not seen in a previous de Haas–van Alphen study.[6] Its appearance was a surprise since local-density-approximation band-structure calculations predict only one large orbit when the field is in the basal plane, and this had already been identified with the ω frequency of Fig. 4(b) [the field-independent peak at 6.4 kT in Figs. 5(c) and 5(d)].[6,7] The clue to the origin of the ω' frequency lies in its quasilinear upward shift with increasing field: extrapolating this dependence to $H = 0$ we find that in zero field the ω and ω' frequencies coincide. This is compelling evidence that these are the spin-split components of one surface. Thus the roughly constant, negative second derivative with field of the ω' extremal area indicated by the behavior shown in Figs. 5(c) and 5(d), compared to the (at most) linear field dependence of the ω surface, demonstrates that subtle, nonlinear quasiparticle band splitting underlies the apparently simple Pauli paramagnetism of the heavy fermion metals, a point underscored by the differing masses of these surfaces: at 12 T, with the field 12° from the a axis, the ω orbit has an effective mass of $(120 \pm 30)m_e$, compared to $(170 \pm 35)m_e$ for the ω' surface, which is the largest quasiparticle mass observed to date in any system.

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[1] Throughout this discussion we assume that the applied field is in the basal plane, and we ignore the superconducting portion of the field-temperature phase diagram. At $T = 0$ K this requires fields larger than $H_{c2} \sim 3$ T.

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[10] See, for example, D. Shoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, Cambridge, 1984). Strictly speaking, the quantum oscillation frequency is a function of the internal field B, instead of the applied field H. But despite the large induced moment per uranium atom in UPt_3, the contribution of the magnetization to the internal field is negligible compared to H.

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[14] See, for example, A. B. Pippard, *Magnetoresistance in Metals* (Cambridge University Press, Cambridge, 1989).