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Authors
Taillefer, L
Newbury, R
Lonzarich, GG
et al.

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DIRECT OBSERVATION OF HEAVY QUASIPARTICLES IN UPt_3 VIA THE dHvA EFFECT

L TAILLEFER, R NEWBURY, G G LONZARICH, Z FISK* and J L SMITH*

Cavendish Laboratory, Cambridge, CB3 0HE, UK

**Los Alamos National Laboratories, Los Alamos, NM 87545, USA*

We present the results of an investigation of the de Haas-van Alphen (dHvA) effect in the heavy fermion superconductor UPt_3

Oscillations composed of up to 8 frequency components corresponding to cyclotron orbits in a plane normal to the a-axis, have been detected in a high purity single crystal and a study of their amplitude as a function of temperature and magnetic field has been performed in the intervals 20–150 mK and 40–115 kG, respectively From this study we obtain estimates of the cyclotron masses, found to range approximately from 25 to 90 times the bare electron mass, and of the mean free path, found to be in excess of 1000 Å The relationship between these findings and the results of conventional energy band calculations is discussed

1. Introduction

The exotic low temperature behaviour of the intermetallic compounds called "heavy fermion superconductors", has been attributed to the existence of fermion quasiparticles with effective masses of unprecedented magnitude, of order 100 times greater than the bare electron mass [1–6] Of fundamental interest is the precise nature of these quasiparticles and the origin of the effective interactions which lead to the formation of the enigmatic superconducting ground states

Although a great deal of indirect evidence for the existence of heavy quasiparticles has been collected, no measurement so far has allowed direct probing and unambiguous detection In this paper we report the first direct observation of these particles in a heavy fermion superconductor, namely in UPt_3, via the de Haas–van Alphen (dHvA) technique

The dHvA effect consists of an oscillatory variation $\tilde{M}$ of the magnetisation as a function of the inverse of the applied magnetic field H In accordance with the traditional theory for a Fermi liquid (see e g refs [7,8]) the frequency and amplitude of $\tilde{M}$ provide a direct measure of the principal properties of quasiparticles near the Fermi level (i e those quasiparticles which are responsible for the low temperature behaviour) In the simplest case of a paramagnet with one partly filled isotropic conduction band, the fundamental component of $\tilde{M}$ at a temperature T may be expressed as

$$\tilde{M} = \frac{\alpha T \exp(-\pi\hbar c k_0/e l_0 H)}{\sqrt{H}\sinh(2\pi^2 m^* c k_B T/e\hbar H)} \sin\left(\frac{\pi\hbar c k_0^2}{eH} + \phi\right), \quad (1)$$

where α and ϕ are quantities which are normally independent of T and H, and $\hbar k_0$, m^* and l_0 are, respectively, the momentum, the effective mass, and the effective mean free path of quasiparticles near the Fermi level The velocity v_0 of the quasiparticles at the Fermi level is given by $m^* v_0 = \hbar k_0$ From eq (1) it is seen that k_0 is determined from the frequency of the oscillations, m^* from the temperature dependence of the (relative) amplitude at fixed H, and l_0 from the field dependence of the (relative) amplitude at fixed T

In the more general case of several partly filled anisotropic conduction bands an oscillatory magnetisation $\tilde{\boldsymbol{M}}$ is associated with each extremal cross-sectional area $\mathscr{A}$ of the Fermi surface lying in a plane normal to $\boldsymbol{H}$ The fundamental component of $\tilde{\boldsymbol{M}}$ parallel to $\boldsymbol{H}$ for a given area $\mathscr{A}$ is in general still given by eq (1), where k_0, m^* and l_0 are now appropriate averages associated with the orbit around $\mathscr{A}$ Specifically, k_0 is the average Fermi wavevector defined by $\mathscr{A} = \pi k_0^2$, m^* is the cyclotron mass defined by $m^* = \hbar k_0/v_0$, where $1/v_0$ is the average of the inverse of the quasipar-

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ticle velocity on the orbit, and $1/l_0$ is the average of the inverse of the quasiparticle mean free path on the orbit

Hence for each group of quasiparticles associated with an extremal area $\mathscr{A}$ of the Fermi surface, the corresponding dHvA frequency $F = \hbar c\mathscr{A}/2\pi e$ and amplitude A will yield three orbitally averaged quasiparticle parameters a Fermi wavevector k_0, a cyclotron mass m^* and a mean free path l_0 These will be the focus of our attention

2. Details of the experiment

High purity single crystals of UPt_3 were prepared in an ultrahigh vacuum by zone-refining a rod of the compound, contained in a water-cooled copper crucible, with radio frequency heating Our purification procedure, previously used for transition metal intermetallic compounds [9–11], yielded bulk single crystals with residual resistivity ratios (extrapolated to absolute zero from the temperature interval of 1 2 to 10 K) in excess of 400 Low strain samples, 0 5 mm thick and 3 0 mm in diameter, were cut from the purest parts of the ingots by means of spark erosion followed by chemical etching

The dHvA experiments were performed in a superconducting magnet-dilution refrigerator system with maximum field of 115 kG and minimum temperature of 20 mK The dHvA magnetisation was detected by means of a low frequency (8 to 25 Hz) field modulation method, the main details of which have been described previously [12] Measurement of the sample temperature, which is particularly critical in the determination of the cyclotron masses, was facilitated by immersion of the sample directly in the dilute liquid phase of the mixing chamber, *and* by means of a thin silver heat link between the sample and a calibrated germanium thermometer The thermometer was embedded in a sintered silver matrix located in the mixing chamber and in a zero field region of the magnet Measurements of the cyclotron masses were carried out as a function of the amplitude and frequency of the modulation field The results presented here were obtained by an extrapolation in the limit of zero amplitude and frequency (i e zero eddy current heating of the sample by the modulation field) The errors quoted for the cyclotron masses arise entirely from the uncertainty in the sample temperature

3. Results of the experiment

Our experimental results, for a field direction along the a axis of the hexagonal ($SnNi_3$) structure (parallel to the ΓK direction in reciprocal space), are presented in figs 1 to 4 and in table 1 Fig 1 shows a typical recording of the dHvA magnetisation and its associated Fourier spectrum at 20 mK Five dominant and several weaker frequency components are evident, pointing to the existence of a complex Fermi surface As shown in table 1, the range of frequencies corresponds to a range of average Fermi wavevectors k_0 from 0 121 to 0 425 Å^{-1} The latter value is 67% of the Brillouin zone wavevector ΓA (fig 5) and, as we shall see in the next section, is comparable to the largest average wavevector expected from conventional band theory for any closed sheet of the Fermi surface of UPt_3

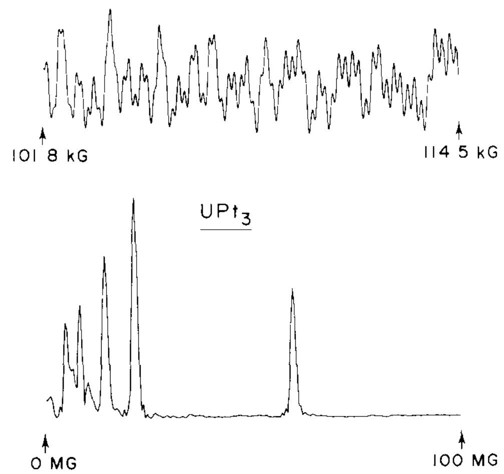


Fig 1 Typical oscillatory variation of the dHvA magnetisation in UPt_3 (upper trace) and the corresponding Fourier spectrum (lower trace) at 20 mK for an applied magnetic field $\boldsymbol{H}$ parallel to the a-axis The signal is detected by a low frequency and low amplitude field modulation technique

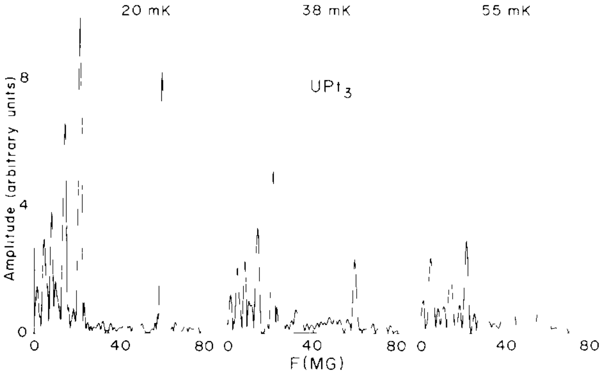


Fig 2 The dHvA amplitude spectrum for a magnetic field interval from 102 9 to 114 2 kG at temperatures of 20, 38 and 55 mK The strong temperature variation of the amplitudes indicates the presence of carriers of unusually high masses

The most dramatic finding, illustrated in figs 2 and 3, is the extremely rapid temperature dependence of the amplitude of all dHvA components observed In particular, the highest frequency component, although strong at 20 mK, collapses almost entirely in the noise when the temperature is raised above 50 mK near 110 kG (fig 2) From eq (1) it follows that this is the behaviour expected for *quasiparticles with masses up to two orders of magnitude stronger than in the simple metals*

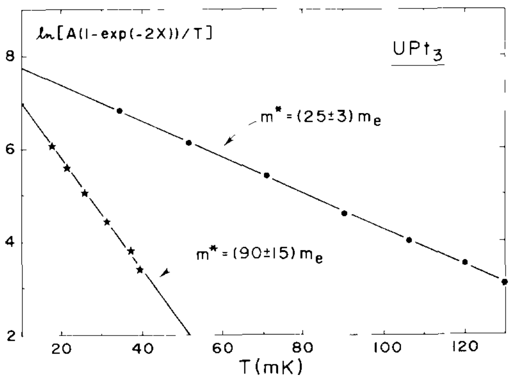


Fig 3 Semi-logarithmic plot of the dHvA amplitude A versus temperature T for the oscillatory components with the lowest (4 8 Mg O) and the highest (59 5 MG *) frequency observed For each component, the effective mass m^* is derived iteratively from eq (1) as described in the text (with $X = 2\pi^2 m^* c k_B T / e\hbar H$) The masses for the above two components are $(25 \pm 3)m_e$ and $(90 \pm 15)m_e$, respectively The components with intermediate frequencies have masses intermediate between these two values (table 1)

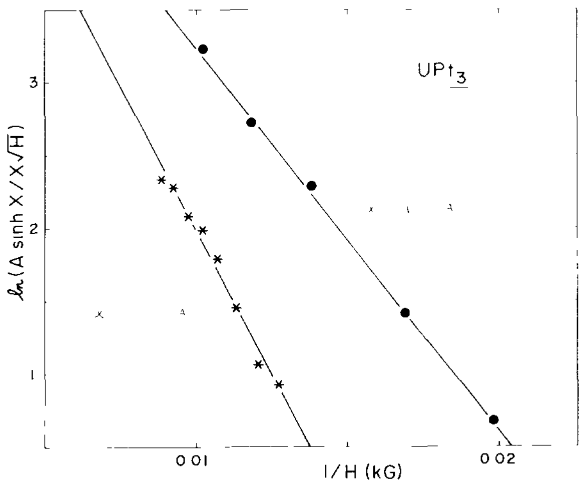


Fig 4 Semi-logarithmic plot of the dHvA amplitude A versus inverse magnetic field for the oscillatory components defined in the caption of fig 3 For each component, an effective mean free path l_0 is derived from eq (1) as described in the text (with X defined in the caption of fig 3) The mean free paths for the above two components are (1000 ± 200) Å and (2200 ± 500) Å, respectively The corresponding scattering (Dingle) temperatures defined by eq (2) are (70 ± 15) mK and (30 ± 7) mK, respectively

A detailed study of the temperature dependence of the dHvA amplitudes (fig 3) yields, from eq (1), the cyclotron masses given in table 2, which range from $25m_e$ for the lowest dHvA frequencies, to $50m_e$ for the intermediate fre-

Table 1
Summary of dHvA results for a magnetic field applied along the a-axis (parallel to the ΓK direction of the reciprocal lattice) F is the dHvA frequency, k_0 is the average effective wavevector characterising the extremal area associated with F, m^* is the cyclotron mass derived from the temperature dependence of the dHvA amplitude, and l_0 is an effective mean free path derived from the field dependence of the amplitude An average effective Fermi velocity v_0 may be defined for each extremal area by $\hbar k_0 = m^* v_0$

F(MG)	k_0(Å^{-1})	$m^*(m_e)$	l_0(10^3 Å)
4 8(3)	0 121(3)	25(4)	1 0(2)
6 1(3)	0 136(3)	–	≥ 1
8 0(4)	0 156(3)	40(7)	≥ 1
14 2(3)	0 208(3)	50(8)	≥ 1
21 4(3)	0 255(2)	60(8)	1 5(4)
59 5(5)	0 425(3)	90(15)	2 2(5)

(1 A = 0 641 Å^{-1})

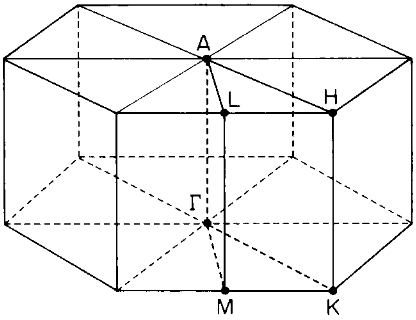


Fig 5 The first Brillouin zone of the hexagonal close packed structure

quencies, and up to $90m_e$ for the highest frequency

In addition to the average wavevectors k_0 and masses m^* of the orbits, we obtain, from the field dependence of the amplitude (eq (1)), estimates of the mean free path l_0 From fig 4 and table 1 we see that in all cases a lower bound for l_0 is approximately 1000 Å, a value comparable to that obtained in many other pure metals which we have investigated We point out that an effective scattering time τ_0 and a scattering (Dingle) temperature T_D for the quasiparticles can be defined by [8]

$$\hbar/\tau_0 = 2\pi k_B T_D = \hbar v_0/l_0 = \hbar^2 k_0/m^* l_0 \qquad (2)$$

Table 2
Comparison of band calculated [17] and experimental values of the average wavevector k_0 (Fermi surface area $\mathcal{A} = \pi k_0^2$) and of the cyclotron masses The ratio of experimental to calculated masses is the largest thus far reported in any system and is in all cases comparable, within experimental error, to the ratio of the measured and band calculated linear coefficient of the heat capacity [13–17]

Sheet	k_0(Å^{-1})		Masses (m_e)	
	Calc *	Exp	Calc *	Exp
Γ1 (e)	0 072	0 12	1 3	25
AL5 (h)	0 10	0 14	0 7	–
L4 (e)	0 12	0 16	1 0	40
K3′ (e)	0 19	0 21	2 1	50
Γ2 (e)	0 22	0 25	2 5	60
Γ3 (e)	0 37	0 43	4 2	90
L4 (h)	0 38	–	6 2	–

* The identification of calculated with experimental values is tentative

Because T_D is inversely proportional to m^* its magnitude is unusually small in UPt_3 (see the caption of fig 4)

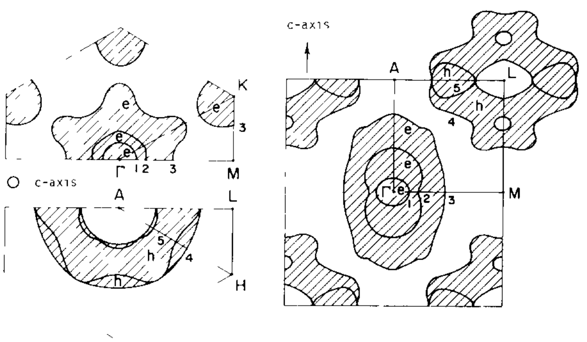
It should be stressed that the effective mean free path appearing in the dHvA amplitude reflects the effect of all scattering processes and of phase cancellation due to inhomogeneities, and is thus normally much smaller than the corresponding mean free path derived from the electrical resistivity

4. Discussion of the results

In this section we discuss the relationship of our dHvA results to conventional energy band models based on the local spin density approximation for the exchange correlation potential

All calculations of this kind predict very similar band models [13–18], and the results of Oguchi and Freeman for the Fermi surface are presented in fig 6 for illustration Their predicted Fermi surface consists of three closed electron surfaces centred on Γ (1, 2 and 3), closed electron surfaces centered on K (3′), two nested toroidal hole surfaces about ΓA (4 and 5), and toroidal surfaces about LH (4) linking neighbouring zones Because UPt_3 is a compensated metal the total volumes enclosed by the electron surfaces and the hole surfaces are equal Surfaces 3 and 4 involve the largest number of carriers and dominate the overall density of states at the Fermi level

The extremal cross-sectional areas in planes normal to the a-axis, in the central ΓALM plane for sheets 1–5 and in a non-central plane for sheet 3′ (fig 6), predicted by the Fermi surface model are given in table 2 (in terms of k_0), together with the calculated cyclotron masses Also in table 2, along with the calculated k_0 and m^* for each orbit, a possible and tentative identification with the observed results is given for ease of comparison It is seen that the *number* of extremal areas predicted and the corresponding *range* of k_0 agree generally with the dHvA results of table 1 On the other hand, the measured masses are *in all cases* much larger, typically of the order of 20 times larger, than the calculated masses This



(a) (b)

Fig 6 Principal sections through the Fermi surface of UPt_3 predicted by the band structure model of Oguchi and Freeman [15], (a) ΓKM, AHL, (b) ΓALM The Fermi surface consists of three closed electron (e) surfaces centred on Γ (1, 2 and 3), closed electron (e) surfaces centred on K (3′), two nested toroidal hole (h) surfaces about ΓA (4 and 5), and toroidal hole (h) surfaces about LH (4)

mass enhancement ratio is comparable to that of the linear coefficient of the heat capacity [13–17]

We note that although the ranges of k_0 calculated and observed are comparable, a unique identification of the detailed Fermi surface topology must await further measurement, as a function of crystal orientation

5. Conclusions

Cyclotron orbits have been observed which correspond to extremal areas of the Fermi surface with average radii ranging from 0 12 to 0 43 $Å^{-1}$, and cycltron masses ranging from 25 to $90 m_e$ The largest area has an average radius equal to 67% of the distance from Γ to A in the first Brillouin zone (fig 5), and it is characterised by the highest cyclotron mass observed so far in any metal

The effective mean free path of the carriers, as inferred from the field dependence of the dHvA amplitude, is between 1000–2200 Å for the various orbits

The experimental Fermi surface areas are not inconsistent with recent local density band calculations [13–18] The observed cyclotron masses are, however, in all cases much greater than predicted by the local density band models, by a factor which is, within the experimental error, of the same order of magnitude as the ratio of the experimental to the band calculated linear coefficient of the heat capacity

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