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DIRECT OBSERVATION OF HEAVY QUASIPARTICLES IN UPt₃ VIA THE dHvA EFFECT

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We present the results of an investigation of the de Haas-van Alphen (dHvA) effect in the heavy fermion superconductor UPt₃. 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 UPt₃, 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

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

where $\alpha$ and $\phi$ 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 $\tilde{M}$ is associated with each extremal cross-sectional area $\mathcal{A}$ of the Fermi surface lying in a plane normal to $H$. The fundamental component of $\tilde{M}$ parallel to $H$ for a given area $\mathcal{A}$ is in general still given by eq (1), where $k_0$, $m^*$ and $l_0$ are now appropriate averages associated with the orbit around $\mathcal{A}$. Specifically, $k_0$ is the average Fermi wavevector defined by $\mathcal{A} = \pi k_0^2$, $m^*$ 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-
particle 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 $\mathcal{A}$ of the Fermi surface, the corresponding dHvA frequency $F = \frac{h c \mathcal{A}}{2 \pi e}$ 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 $\Gamma K$ 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 $\Gamma A$ (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 $H$ parallel to the $a$-axis. The signal is detected by a low frequency and low amplitude field modulation technique.](image-url)
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.

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 25me for the lowest dHvA frequencies, to 50me for the intermediate frequencies.

Table 1

<table>
<thead>
<tr>
<th>F(MG)</th>
<th>k_o(Å⁻¹)</th>
<th>m*(me)</th>
<th>l_o(10⁻⁹ Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4 8(3)</td>
<td>0 121(3)</td>
<td>25(4)</td>
<td>1 0(2)</td>
</tr>
<tr>
<td>6 1(3)</td>
<td>0 136(3)</td>
<td>–</td>
<td>≥ 1</td>
</tr>
<tr>
<td>0 8(4)</td>
<td>0 156(3)</td>
<td>40(7)</td>
<td>≥ 1</td>
</tr>
<tr>
<td>14 2(3)</td>
<td>0 208(3)</td>
<td>50(8)</td>
<td>≥ 1</td>
</tr>
<tr>
<td>21 4(3)</td>
<td>0 255(2)</td>
<td>60(8)</td>
<td>1 5(4)</td>
</tr>
<tr>
<td>59 5(5)</td>
<td>0 425(3)</td>
<td>90(15)</td>
<td>2 2(5)</td>
</tr>
</tbody>
</table>

(IA = 0.641 Å⁻¹)
Fig 5 The first Brillouin zone of the hexagonal close packed structure

quencies, and up to 90\textit{m}_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 \( \tau_0 \) and a scattering (Dingle) temperature \( T_D \) for the quasiparticles can be defined by [8]

\[ \hbar/\tau_0 = 2\pi k_B T_D = \hbar v_0/l_0 = \hbar^2 k_0/m^* l_0 \]  

(2)

Table 2 Comparison of band calculated [17] and experimental values of the average wavevector \( k_0 \) (Fermi surface area \( \Delta = \pi k_0^2 \)) 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]

<table>
<thead>
<tr>
<th>Sheet</th>
<th>( k_0 (\text{Å}^{-1}) )</th>
<th>Masses (m\textit{e})</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \Gamma1 ) (e)</td>
<td>0.072 0.12</td>
<td>1.3 25</td>
</tr>
<tr>
<td>AL5 (h)</td>
<td>0.10 0.14</td>
<td>0.7 –</td>
</tr>
<tr>
<td>L4 (e)</td>
<td>0.12 0.16</td>
<td>1.0 40</td>
</tr>
<tr>
<td>K3’ (e)</td>
<td>0.19 0.21</td>
<td>2.1 50</td>
</tr>
<tr>
<td>( \Gamma2 ) (e)</td>
<td>0.22 0.25</td>
<td>2.5 60</td>
</tr>
<tr>
<td>( \Gamma3 ) (e)</td>
<td>0.37 0.43</td>
<td>4.2 90</td>
</tr>
<tr>
<td>L4 (h)</td>
<td>0.38 –</td>
<td>6.2 –</td>
</tr>
</tbody>
</table>

\* The identification of calculated with experimental values is tentative

Because \( T_D \) is inversely proportional to \( m^* \) its magnitude is unusually small in UPt\textsubscript{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 \( \Gamma \) (1, 2 and 3), closed electron surfaces centered on \( K \) (3’), two nested toroidal hole surfaces about \( \Gamma A \) (4 and 5), and toroidal surfaces about \( LH \) (4) linking neighbouring zones. Because UPt\textsubscript{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 \( \Gamma ALM \) 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 \textit{number} of extremal areas predicted and the corresponding \textit{range} of \( k_0 \) agree generally with the dHvA results of table 1. On the other hand, the measured masses are \textit{in all cases} much larger, typically of the order of 20 times larger, than the calculated masses. This
The 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 \( \text{Å}^{-1} \), and cyclotron masses ranging from 25 to 90\( m_e \). The largest area has an average radius equal to 67% of the distance from \( \Gamma \) 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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References