Lifshitz transition underlying the metamagnetic transition of UPt$_3$

To cite this article: A McCollam et al 2021 J. Phys.: Condens. Matter 33 075804

View the article online for updates and enhancements.
Lifshitz transition underlying the metamagnetic transition of UPt$_3$

A McCollam$^1$, Mingxuan Fu$^2$ and S R Julian$^{3,*}$

$^1$ High Field Magnet Laboratory (HFML-EMFL), Radboud University, Toernooiveld 7, 6525 ED Nijmegen, The Netherlands
$^2$ Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan
$^3$ Department of Physics, University of Toronto, Toronto, M5S 1A7, Canada

E-mail: sjulian@physics.utoronto.ca

Received 18 August 2020, revised 19 October 2020
Accepted for publication 3 November 2020
Published 23 November 2020

Abstract
Comparing quantum oscillation measurements, dc magnetoresistance measurements, and Fermi surfaces obtained from LDA calculations, we argue that the metamagnetic transition of UPt$_3$, which occurs at an applied field $H_M \sim 20$ T, coincides with a Lifshitz transition at which an open orbit on the band 2 hole-like Fermi surface becomes closed for one spin direction. At low field, proximity of the Fermi energy to this particular van Hove singularity may have implications for the superconducting pairing potential of UPt$_3$. In our picture the magnetization comes from non-linear spin-splitting of the heavy fermion bands. In support of this, we show that the non-linear field dependence of a particular quantum oscillation frequency can be fitted by assuming that the corresponding extremal Fermi surface area is proportional to the magnetization. In addition, below $H_M$, we find in our LDA calculations a new, non-central orbit on band 1, whose non-linear behaviour explains a field-dependent frequency recently observed in magnetoacoustic quantum oscillation measurements.

Keywords: heavy fermion, metamagnetism, Lifshitz transition, strongly correlated electrons

(Some figures may appear in colour only in the online journal)

1. Introduction
An interesting question in the physics of heavy fermion metals is the extent to which conventional band theory can be applied to the heavy quasiparticle bands that result from Kondo coupling of local $f$-moments to the light conduction electron bands. This question is particularly relevant when considering the magnetization, $M$, induced in a paramagnetic heavy fermion metal by an external magnetic field, $H$. Does the magnetism result from simple paramagnetic splitting of the heavy fermion bands, or does the character of the bands change in a more fundamental way, for example by the disappearance of $f$-electrons from the Fermi volume?

In heavy fermion metals, $M(H)$ is generally non-linear. The most extreme non-linear behaviour is seen at metamagnetic transitions, at which $M(H)$ suddenly rises over a narrow range of $H$ (for a recent review see reference [1]).

The conventional band theory of metamagnetism goes back to Wohlfarth and Rhodes [2], who argued that a metal which nearly satisfies the Stoner criterion at $H = 0$ can be pushed over the edge into ferromagnetism if, with increasing $H$, the density of states at the Fermi energy $E_F$ increases as the energy bands undergo spin-splitting. A typical scenario is that a van Hove singularity, located near $E_F$, crosses the Fermi energy as the bands become spin polarized. van Hove singularities crossing $E_F$ are associated with Lifshitz transitions—changes in the Fermi surface topology such as the appearance of a new Fermi surface, or the merging of existing Fermi surfaces [3].
triple, \[24\], and the linear coefficient of specific heat is about 1.5 times [25, 26], their respective low-field values. In this paper, using previous quantum oscillation results supplemented by new measurements of dc and quantum oscillatory magnetoresistance, we argue that not only does field-induced splitting of heavy fermion bands explain the 20 T metamagnetic transition in the heavy fermion system UPt3, but also that it is possible to identify which of several possible Lifshitz transitions, found near \(E_F\) in the LDA band structure, drives the metamagnetic transition. UPt3 is a nearly-ferromagnetic heavy fermion system, and it provided the first definitive example of a superconductor with multiple superconducting phases [19–21], which is unambiguous evidence of non-BCS superconductivity. Information about which bands are responsible for the proximity to ferromagnetism is thus of interest beyond the theory of heavy fermion metamagnetism.

For \(H\) applied in the \(a\)–\(b\) plane the magnetization of UPt3 initially grows roughly linearly, reaching about 0.35 \(\mu_B\) per U at 18 T. It then rises much more rapidly through the metamagnetic transition that is centred on \(H_M \sim 20\) T, reaching \(\sim 0.6\) \(\mu_B/U\) by 22 T. Above 22 T, \(M(H)\) again rises more slowly, at a roughly constant rate [22, 23]. (This behaviour is illustrated in figure 4(a)). At \(H_M\) the many-body properties are enhanced: the \(T^2\) coefficient of resistivity is roughly three times, [24], and the linear coefficient of specific heat is about 1.5 times [25, 26], their respective low-field values.

The main features of the band structure of UPt3 are well known from previous quantum oscillation, magnetoresistance, and LDA band structure studies [24, 28–31]. The Fermi surface is, unfortunately, rather complicated, arising from five bands that cross the Fermi energy. The Fermi surfaces corresponding to bands 1 to 3 are illustrated in figure 1(a). Bands 1 and 2 produce hole-like Fermi surfaces that are centred on the \(A\)-point of the Brillouin zone. Band 3 is electron-like and has a large closed surface, centred on the \(\Gamma\)-point which is at the centre of the Brillouin zone, and six smaller \(K\)-centred ellipsoids, which in figure 1(a) are joined to the central surface (in some band-structure calculations the \(K\)-centred ellipsoids are disconnected from the central surface, e.g. [30, 31]). Bands 4 and 5 are electron-like and centred on \(\Gamma\). At fields \(H < H_M\) evidence for all of these bands has been found in quantum oscillation and magnetoresistance studies, and in the case of bands 1, 2 and 3, the agreement between experiment and theory is very good, with the exception of the predicted \(K\)-centred ellipsoids of band 3 for which evidence is tenuous at best, and if they do exist they are not joined to the \(\Gamma\)-centred surface of band 3. Above \(H_M\) the situation is less clear: taking into account spin-splitting, up to ten bands now cross the Fermi energy, some of which produce Fermi surfaces with multiple extremal orbits. This produces a forest of low-frequency quantum oscillations, and disentangling them is challenging [32].

The earliest low-temperature magnetoresistance measurements in UPt3 were thought to show evidence of a major reconstruction of the Fermi surface at the metamagnetic transition [24]. The most likely scenario for such a reconstruction is localization of some or all of the uranium 5\(f\)-electrons. Even at zero field anywhere from zero to three 5\(f\) electrons per uranium can be itinerant. Non-magnetic UPd3 is at one extreme, with two 5\(f\)-electrons in a localized singlet state below \(E_F\), while the next lowest 5\(f\) state is above \(E_F\) (i.e. the uranium ion is tetravalent) [33]. In UPt3, at the other extreme, all three 5\(f\) electrons occupy bands that cross \(E_F\). As a function of magnetic field, it is conceivable that one or more of these 5\(f\)-electrons becomes localized, with a sudden shrinking of the overall Fermi volume, which would be expected to change not only the size but also the topology of some Fermi surfaces.

In a subsequent study of the quantum oscillatory magnetoresistance, however, we showed that the large \(\Gamma\)-centred surface of band 3 survives the metamagnetic transition, and that heavy fermions also survive above \(H_M\) [32]. The frequency spectra from the smaller Fermi surface orbits were too complex to be followed across \(H_M\) but, very recently, magnetoacoustic quantum oscillation measurements as a function of field-angle found that several of the smaller Fermi surfaces survive above \(H_M\) with little change [34]. Moreover, we show below...
that an open orbit on band 2 survives above $H_M$, at least for one spin direction; and, finally, we track a particularly strong low-frequency quantum oscillation across $H_M$, as the corresponding Fermi surface becomes spin-polarized, apparently smoothly.

Taken together, these results tell us that the Fermi surface of UPt$_3$ does not undergo a major change—such as would result from localization of some or all of the U $f$-electrons—at $H_M$. Thus, a spin-polarized band picture is likely to be correct, and it is reasonable to look for a Lifshitz transition on one of the bands as the source of the metamagnetic transition. Due to the complexity of the Fermi surface, identifying such a Lifshitz transition at $H_M$ could be a major challenge. In recent work Holleis et al [34] suggest that the metamagnetic transition arises from a Lifshitz transition on the band 1 Fermi surface. For $H \parallel a$, they observe a strong quantum oscillation with a frequency of about 0.7 kT well below $H_M$. As $H_M$ is approached from below the frequency increases rapidly, while above $H_M$ the oscillation is absent. The field dependence below $H_M$ indicates that the corresponding extremal orbit is shrinking as $H_M$ is approached from below. They ascribe this frequency to band 1, and thus hypothesize that a Lifshitz transition at which the majority-spin branch of band 1 disappears is responsible for the metamagnetic transition.

Below, we point out that if the LDA band structure is to be believed, the Lifshitz transition identified by Holleis et al would lead to a decrease, rather than an increase, in the density of states, so it would not drive a metamagnetic transition. We also present new magnetoresistance measurements that extend our earlier work. We find a curious feature in the density of states, so it would not drive a metamagnetic transition at which the majority-spin branch of band 1 disappears is responsible for the metamagnetic transition.

The purpose of our band-structure calculations is to find features that might be linked to the metamagnetic transition. Specifically, we assume in the following that the bands are rigidly spin split as $H$ is increased through $H_M$, and we look for anomalies in the density of states that could drive the metamagnetic transition.

In figure 2(a) we show the spaghetti plot for the five bands that cross $E_F$, while figures 2(b) and (c) show their corresponding densities of states. In our naive band-magnetism picture, under a magnetic field each band splits into a minority-spin branch that moves up in energy, and a majority-spin branch that moves down, and consequently each density of states curve splits in two identical parts that shift up(down) relative to $E_F$ corresponding to the minority(majority)-spin bands.

In figure 2(c), which zooms in on the region around the Fermi energy, we have labeled van Hove singularities in the density of states arising from extrema in the band structure that lie closest to $E_F$. For four of them, $b$, $c$, $d$ and $d'$, are also indicated in the spaghetti plot, figure 2(a). (The fifth, $a$, is not indicated there because it is not located on a high-symmetry line in $k$-space).

Previous studies have found compelling agreement between quantum oscillation measurements and the Fermi surfaces derived from the LDA calculations. There is, therefore, good reason to trust that the van Hove singularities of figure 2 exist near $E_F$. Their precise position relative to $E_F$ is known to be uncertain by as much as $\sim 1$ mRy, because this is the amount by which bands needed to be shifted to get good agreement between the calculated Fermi surface and quantum oscillation measurements [28, 29]. Moreover, the shifts are not all in the same direction: some bands need to
Figure 2. (a) The band structure for the five bands that cross \(E_F\) in our LDA calculation, along high-symmetry directions in \(k\)-space. Magenta arrows indicate van Hove singularities. The corresponding density of states from bands 1 to 5 is shown in (b), while (c) zooms in on fine structure near \(E_F\). In (c), the letters \(\text{a} \) to \(\text{d}'\) refer to density of states anomalies associated with the van Hove singularities of (a), and the Lifshitz transitions shown in figure 3. (The van Hove singularity corresponding to the \(\text{a}\) density of states anomaly is not shown in figure (a) because it is not located on the \(k\)-space paths shown there).

be moved up in energy, and others down, in order to correctly predict the size and shape of extremal orbits. A similar level of uncertainty is evident upon comparing different calculations. For example, the bandstructure calculation of Harima [30] places \(E_F\) below the density of states feature \(\text{a}\) in figure 2(c), while our calculation and that of Norman [29] place it above (by more than 1 mRy in our case). Similarly, only in some calculations are the \(K\)-centred ellipsoids and the \(\Gamma\)-centred ellipsoid of band 3 connected to each other, which requires that density-of-states feature \(\text{b}\) lies below \(E_F\) (as in our calculation); in other calculations [29, 30] \(\text{b}\) is above \(E_F\), and the \(K\)-centred ellipsoids are separate.

Thus, we cannot use the calculated band structure to say which of these van Hove singularities actually lies closest to \(E_F\), and we must appeal to Fermi surface measurements to try to answer this question.

The Lifshitz transitions that correspond to van Hove singularities \(\text{a} \) to \(\text{d}'\) are illustrated in figure 3, and they each have a distinctive effect on the Fermi surface. The top row in figure 3 shows the Lifshitz transition that corresponds to van Hove singularity \(\text{a}\) of figure 2(c). Across the transition, narrow tubes form, linking the arms of the band 2 'octopus' Fermi surface. The arrow in the middle column of figure 3 indicates the direction in which the Lifshitz transition has to proceed in order to increase the density of states, as required to drive a metamagnetic transition. In this case, the low-field Fermi surface would have to be as pictured on the right, and at \(H = H_M\) the minority-spin Fermi surface would undergo a Lifshitz transition to become like the Fermi surface pictured on the left.

At the next Lifshitz transition, \(\text{b}\), the large \(\Gamma\)-centred ellipsoid of band 3 links up with the six small \(K\)-centred ellipsoids. As indicated by the double arrow, the low field Fermi-surface would have to be the left-hand one, and at \(H = H_M\) the majority-spin Fermi surface would change to become like that on the right.

Figure 3. Lifshitz transitions on bands 1 to 3, corresponding to the van Hove singularities indicated by the letters \(\text{a} \)–\(\text{d}'\) on figure 2(c). In each row, the left/right pictures show the relevant Fermi surface when the van Hove singularity is above/below \(E_F\). The double arrows indicate in which direction the Lifshitz transition causes the density of states to increase. Only in the second row is the Brillouin zone centred on the \(\Gamma\) point; all of the others, involving bands 1 and 2, are centred on \(A\).
At the third Lifshitz transition, c, holes form in the band 1 Fermi surface along the $A$ to $H$ line, as shown in figure 3(c) and again the majority spin Fermi surface would change shape from the left to the right.

At Lifshitz transition d these holes in band 1 expand into a ring that cuts the band 1 Fermi surface into disconnected pieces. This is a rather broad feature in the density of states that actually covers two Lifshitz transitions, one at which the necks that join the central part to the outer ring disappear, and the other when the outer torus breaks up into separate islands. At this transition the low-field Fermi surface would have to be the one on the right, and at $H_M$ the minority-spin Fermi surface would change to the shape on the left.

Finally, d' is a Lifshitz transition of the majority-spin Fermi surface of band 2 in which it acquires holes, as shown in the bottom rows of figures 3 and 9. (Note that we have used the d, d' notation because, in the non-symmorphic $P6_3/mmc$ crystal structure, which is only very weakly departed from, bands 1 and 2 are degenerate along the line $A - L$ on the Brillouin zone and these two Lifshitz transitions then coincide. Even with the symmorphic crystal structure they should occur very close together.)

In the following two sections, using quantum oscillations and dc-magnetoresistance data, we are able to discard all but Lifshitz transition d' as the cause of the metamagnetic transition, and we provide strong evidence from the dc-magnetoresistance that Lifshitz transition d' indeed occurs at $H_M$.

4. Quantum oscillations

We now compare the Lifshitz transition scenarios of the previous section with quantum oscillation measurements. We use old quantum oscillation data [28, 30–32], the recent results of Holleis et al [34], plus our new magnetoresistance measurements.

We begin with Lifshitz transition a. A priori, this is the most promising candidate for driving the metamagnetic transition, because it gives the biggest and sharpest jump in the density of states. As noted above, in this scenario, $E_F$ at zero field would have to be located close-to but above density of states feature a in figure 2, and at $H_M$ the minority-spin Fermi surface would suddenly acquire narrow tubes that link the arms of the band 2 Fermi surface. If this were to happen, a new low-frequency quantum oscillation would appear at $H_M$. There are no quantum oscillation studies where such a low frequency suddenly appears at $H_M$. This statement requires some qualification, however, because there is a very strong low-frequency quantum oscillation that has been observed in both the oscillatory magnetoresistance [31, 32] and magnetoacoustic quantum oscillation measurements [34]. It is particularly strong for some field directions, for example when the field is around 5° from the b-axis (e.g. the blue line in figure 4(b)). At these angles, strong low-frequency oscillations are seen both above and below $H_M$, but the frequency obtained from Fourier analysis is quite different above and below $H_M$, being ~0.15 kT below 12 T, and ~0.4 kT at 30 T. It is important in the context of this discussion to establish whether the $H < H_M$ and $H > H_M$ oscillations represent two different Fermi surfaces, or the same Fermi surface that changes size across $H_M$. The latter has previously been suggested in reference [34].

In the appendix we provide a detailed demonstration that this is almost certainly the same Fermi surface that has expanded non-linearly as the system becomes magnetically polarized. Figure 4 compares the measured quantum oscillatory magnetoresistance with a calculated quantum oscillation (blue and red lines in figure 4(b)). The calculated oscillation has the form $A(B)\sin(F(B)/B + \phi)$ with the quantum oscillation frequency varying as $F(B) = F(0) + aM(B)$. That is, we assume that the field dependent part of the oscillation frequency is proportional to the magnetization. In the Onsager formula $F$ is related to the extremal area $A$ of a Fermi surface by $F = \hbar A/2\pi e$, so we are in effect assuming that the field-dependent spin-splitting of the extremal orbit is proportional to the magnetization. In the data, which was taken with the applied field 4° from the b-axis (blue line in figure 4(b)) the strong low-frequency oscillation is most clearly observed below 13 T and above 21 T, where it can be seen that the calculation agrees well with the data. The most telling agreement is a peculiar turning point in the oscillation near 23 T, marked by the yellow arrow.

Note that the frequency shown on the left-hand axis of figure 4(a) is not what one would measure from the usual Fourier analysis of quantum oscillations. Rather, as explained in the appendix, the frequency obtained from Fourier analysis is the so-called back-projected frequency, and figure 10(c) shows that this agrees well with the 0.15 kT and 0.4 kT values.

![Figure 4. Continuous, non-linear expansion of a Fermi surface across $H_M$.](image-url)

(a) $F(B)$ (T) (b) $F(B)$ (T)
strongly field-dependent quantum oscillation frequency which
starts, below 14 T, at about 0.7 kT, and then increases rapidly as
HM is approached from below, disappearing permanently just
before HM is reached (similar to the blue line in figure 5(c),
which is a result of the simulation described below). They sug-

gest that this oscillation comes from the band 1 Fermi surface,
and interpret this behaviour as meaning that the metamagnetic
transition is caused by a Lifshitz transition in which one spin
component of the band 1 Fermi surface disappears.

The assignment of the 0.7 kT oscillation to band 1 was
not obvious. Previous authors have assigned only one quan-
tum oscillation to band 1: the so-called \( \delta \) orbit, which has a
frequency of \(~1.2–1.4\) kT for fields in the basal plane [28].
Although they do not address this point directly, Holleis et al
[34] seem to feel that this is a mis-assignment, and instead
assign the \( \delta \) frequency to the

\[
F(B) - B\delta F/\delta B
\]

\(1/\mu\), H (T⁻¹)

\[
F(\Delta E_F)
\]

\[\mu = \frac{\mu_0}{2}\pi e \frac{\partial A(B)}{\partial B},\]

where \( A \) is the extremal area of the Fermi surface
as a function of field, while the blue curve is the
"back-projected" frequency, \( F_{\text{proj}}(B) = F(B) - B\delta F/\delta B \). These are plotted
vs \(1/\mu, H\) for comparison with figure 7 of reference [34].

Figure 5. (a) is a plot of the calculated dHvA frequency arising from the
non-central orbit of band 1, as the band shifts down in energy
relative to \( E_F \); \( \Delta E_F = 0 \) is where our LDA calculation places the
Fermi energy. The three Fermi surface pictures above the plot show the
evolution of the topology of band 1 as it shifts in energy. The
non-central (dark blue) orbit abruptly disappears at Lifshitz
transition \( d \) when band 1 has shifted down by slightly more than
0.7 mRy; (b) To simulate the field dependence of the non-central
orbit’s frequency, we assume that the shift of the band energy
follows the magnetization curve used in figure 4(c), tuning the
proportionality so that the Lifshitz transition occurs at about 20 T.
(c) The red curve is the resulting ’Onsager’ frequency
\( F(B) = \hbar A(B)/2\pi e \), where \( A \) is the extremal area of the Fermi
surface) as a function of field, while the blue curve is the
‘back-projected’ frequency,
\( F_{\text{proj}}(B) = F(B) - B\delta F/\delta B \). These are plotted
vs \(1/\mu, H\) for comparison with figure 7 of reference [34].

In addition to lending support to our contention that Lifshitz
transition \( a \) does not occur at \( H_M \), because a new low frequency
oscillation does not suddenly appear at \( H_M \), the success of this
modelling of \( F(B) \) further supports our spin-polarized-band
picture of the metamagnetism of UPt₃.

Moving on to the band 1 Lifshitz transitions \( c \) and \( d \), Holleis
et al [34] recently reported an interesting observation of a
strongly field-dependent quantum oscillation frequency which
was not obvious. Previous authors have assigned only one quan-
tum oscillation to band 1: the so-called \( \delta \) orbit, which has a
frequency of \(~1.2–1.4\) kT for fields in the basal plane [28].
Although they do not address this point directly, Holleis et al
[34] seem to feel that this is a mis-assignment, and instead
assign the \(~1.2\) kT frequency to the \( K \)-centred pockets of
band 3, but this seems very unlikely given the excellent agree-
ment between the predicted and observed angle dependence of
the \( \delta \) frequency as the field is rotated away from the basal plane
[28]. Applying the SKEAF extremal-area-finding routine [38]
to our LDA band structure, however, turns up a second, non-
central orbit on band 1 with a frequency close to 0.7 kT, which
has not previously been identified. The original \( \delta \) orbit and
this new orbit are indicated by the red and blue lines on the
band 1 Fermi surfaces in figure 5(a), which also shows how
this new frequency changes as band 1 moves down in energy
relative to \( E_F \). As shown by the dark blue points in figure 5(a),
the frequency depends quasi-linearly on \( E \sim E_F \), until it sud-
denly disappears at \( \Delta E_F \sim 0.72\) mRy as the band 1 Fermi
surface undergoes the \( d \) Lifshitz transition. The frequency
will, however, vary non-linearly as function of \( B \), because
\( E \sim E_F \) is non-linear in \( B \) (figure 5(b)). The resulting non-
linear behaviour of \( F \) (red line in figure 5(c)) can be analyzed to
obtain the ‘back-projected’ frequency \( F(B) - B\delta F/\delta B \) (blue
line in figure 5(c)), and this agrees quite well with figure 7 of

Figure 6. Main figure: some typical curves of resistivity, \( R \), vs
applied field, \( H \), at various angles of the field between the \( a \) axis (0°)
and the \( b \) axis (30°). The inset shows resistance vs field-angle at 8 T
(dashed line, taken from our previous study [31]) and at 25 T (this
study), at \( T < 100 \) mK. The sharp minimum at the \( a \)-axis (0°)
arises from an open orbit on band 2 of the Fermi surface, shown as the
blue line in figure 1(b).
Holleis et al. This agreement is strong evidence that $\varepsilon_F$ is initially located below, but near, Lifshitz transition $d$ on band 1. This in turn rules out Lifshitz transition $c$, which requires that $\varepsilon_F$ be far from $d$. It also supports the contention of Holleis et al. that a Lifshitz transition $d$ of band 1 occurs near, or even at, $H_M$, although it does not correspond to their picture of a complete disappearance of the band 1 majority-spin Fermi surface at $H_M$.

Lifshitz transition $d$ does not, however, produce a metamagnetic transition within this scenario for the simple reason that the density of states feature is not only rather weak but it also goes in the wrong direction—the density of states decreases when the band 1 majority spin Fermi surface undergoes this Lifshitz transition, whereas metamagnetism results from an increase in the density of states with increasing field.

Rather, we feel that the importance of the result of Holleis et al. [34] is that it establishes not only that $d$ occurs close to $H_M$, but also that $d'$ occurs close to $H_M$. As noted above, bands 1 and 2 are nearly degenerate along the $A$–$L$ line of the Brillouin zone (indeed they would be exactly degenerate if the structure were the non-symmorphic $P6_3/mmc$), so $d$ and $d'$ must occur close together in field. Thus if $d$ occurs near $H_M$, so must $d'$. We argue below, based on new dc-magnetoresistance results, that $d'$ indeed coincides with $H_M$.

5. Non-oscillatory magnetoresistance

Figure 6 shows typical results of our measurements of the resistance $R$ vs field $\mu_s H$ between 0 and 33 T. At most angles the magnetoresistance is large. Typical behaviour (except for $H||a$) is that the resistance rises quasi-linearly between the upper critical field for superconductivity (near 3 T) and somewhere between 8 and 11 T. This is followed by a broad local maximum, after which $R(H)$ decreases up to $H_M \sim 20$ T, above which it again rises rapidly.

The inset of figure 6 shows $R$ vs angle at 25 T, at $\sim 55$ mK. The overall shape is similar to that at 8 T (dashed line). The magnetoresistance minima at $H||a$ and $H||b$ are interpreted as arising from open orbits on band 2 [24, 31], which are illustrated in figure 1(b). UPt$_3$ is a compensated metal, and for intermediate angles, the quasiparticles on all Fermi surfaces undergo cyclotron motion under the influence of the applied field, which in the limit $\omega_c \tau \gg 1$, where $\omega_c$ is the cyclotron frequency and $\tau$ is the scattering time, produces a large magnetoresistance. When $H$ is parallel to the $a(b)$ axis, however, quasiparticles on the blue(red) lines in figure 1(b) pass from one Brillouin zone to the next, without a sign-change in their $c$-axis velocity, hence they ‘short-circuit’ the $\omega_c \tau$ magnetoresistance of quasiparticles on the rest of the Fermi surface, producing a much lower magnetoresistance. (A similar phenomenon is well known in copper [39], where it arises from open-orbits across the ‘necks’ of the Fermi surface.) What the inset of figure 6 demonstrates is that, at least for one spin direction, the general shape of band 2 is unchanged above $H_M$, because the open orbits are still present.

We have not yet eliminated Lifshitz transition $b$ from contention, but figure 6 allows us to do that. If Lifshitz transition $b$ were to occur at $H_M$, a new open orbit would form which would depress the magnetoresistance for $H || b$. Figure 6 shows that, if anything, the $H || b$ magnetoresistance is larger for $H > H_M$ than for $H < H_M$.

In figure 7 we show the magnetoresistance for $H || a$ compared with that just one degree away. In addition to the obvious difference in the magnitude of the magnetoresistance, the inset, by focussing on the metamagnetic transition, reveals a more subtle difference: the resistance with $H \perp a = 1^\circ$ changes smoothly across $H_M$, while that for $H || a$ has a cusp, or sudden change of slope, near $\mu_s H = 20.5$ T.

In figure 8 we compare all of our resistance curves in the metamagnetic transition region. While many of the curves show distinctive features at or near $H_M$, the $H || a$ curve is the only one with such a sharp change of slope. The $\mu_s H || b$ curve looks, at first glance, as though it may also have a cusp, but
Figure 8. Resistance vs magnetic field from the $b$-axis (30$^\circ$) to the $a$-axis (0$^\circ$). At all angles except 0$^\circ$ the magnetoresistance changes smoothly through the 20 T metamagnetic transition. For 0$^\circ$ there is a cusp at 20.5 T, which we interpret as evidence of a Lifshitz transition that interrupts the open orbit on the majority-spin Fermi surface of band 2.

Careful inspection of figure 7(b) shows that the slope is continuous, with perhaps a field-dependent quantum oscillation superposed on top of a gradual change of slope.

Moreover, from figure 7 we can see that the resistance for $H \parallel a$ (i.e. 0$^\circ$) rises by a factor of nearly two upon crossing $H_M$. A natural interpretation of the $H \parallel a$ magnetoresistance is that the open orbit has been interrupted at $H_M$ for one spin direction. This is consistent with Lifshitz transition $d'$.

Figure 9. Spin-splitting of the calculated band 2 Fermi surface. The top row is at zero field, where the majority and minority-spin Fermi surfaces are identical, both having open orbits (blue lines). For fields just below $H_M$ (middle row), both the minority (right) and majority (left) Fermi surfaces still have open orbits, but as $H_M$ is crossed the open orbit of the majority-spin Fermi surface (bottom left figure) changes into two closed orbits.

6. Discussion and conclusions

Above we showed that the LDA band structure of UPt$_3$ has several Lifshitz transitions that are very close to $E_F$. We argued that a crude picture, in which the bands split rigidly under the application of an external field, is consistent with magnetoresistance and quantum oscillation measurements. While uncertainties within the LDA calculation mean that any one of the Lifshitz transitions could produce the 20 T metamagnetic transition of UPt$_3$, comparison with old and new quantum oscillation studies allowed us to eliminate all but two of the Lifshitz transitions from consideration.

Moreover, one of the remaining transitions, the band 1 Lifshitz transition $d$, which seems to occur very close to $H_M$, is not consistent with metamagnetism because the density of states changes in the wrong direction. In the course of this discussion we identified a new non-central orbit on band 1. The remaining transition, $d'$, while not directly observed in quantum oscillation measurements, should occur in conjunction with $d$, and moreover it shows up in our magnetoresistance measurements, which suggests that for one spin direction an open orbit on band 2 is interrupted at $H_M$, as predicted for the $d'$ Lifshitz transition. We thus feel that it is a firm conclusion that Lifshitz
transition $d'$ causes the metamagnetic transition that occurs at $H_M$.

Since their introduction Lifshitz transitions have been classified according to the strength of the singularity that they produce in the density of states [3]. For conventional Lifshitz transitions in three-dimensional metals, of the ‘neck-forming’ or ‘new-surface-appearing’ type, there is a one-sided square-root singularity in the density of states. Recently, new categories have been recognized for Lifshitz transitions at high symmetry points of quasi-two-dimensional materials [5, 40] that produce more singularities in the density of states. In our (three-dimensional) case, the van Hove singularity is centred on the Brillouin zone boundary but it is not otherwise at a high symmetry point. Nevertheless the density of states anomaly is not simple. Figure 9 makes it appear that the van Hove singularity is of the straightforward ‘neck-forming’ type. Fitting the LDA band-structure around the singularity, however, gives the dispersion as

$$
\delta \varepsilon(k) = \frac{\hbar^2 k_x^2}{2m_\parallel} + \frac{\hbar^2 k_y^2}{2m_\parallel} + \frac{\hbar^2 k_z^2}{2m_z} - a_4 k^4_z,
$$

where the masses and $a_4$ are all positive constants. The positive $k_x^2$ and negative $k_y^2$ terms combine to make the dispersion relation quite flat in the $k_z$ direction near the singularity, enhancing the density of states anomaly. Moreover, this combination means that, although the approach to the Lifshitz transition looks like a simple ‘neck-forming’ transition will occur, as the critical field is approached the transition takes place in two stages: a new pocket first forms at the Brillouin zone boundary, and then it quickly grows in the $k_z$ direction to form the neck that pierces the band 2 Fermi surface.

The classification of the Lifshitz transition would be more exotic if the crystal structure were the non-symmorphic $P6_3/mmc$ structure, because then Lifshitz transitions $d$ and $d'$ would coincide. The $d$ transition is a ‘neck-breaking’ transition (with no appreciable fourth-order term in $k$ in the dispersion relation) with the density of states anomaly on the low-field side of $H_M$, while the $d'$ transition has its density of states anomaly on the high-field side of $H_M$. Thus, for this crystal structure there would be density of states anomalies on both sides of $H_M$. Even in the observed symmorphic crystal structure, the splitting of these transitions may be very weak, so that these transitions nearly coincide.

Aside from its relevance to the metamagnetism of UPt3, the superconducting pairing interaction may depend on which van Hove singularities are closest to $E_F$. Recently, the pairing interaction was estimated for UPt3 using magnetic fluctuations derived from an effective Hamiltonian based on the LDA band-structure [36]. At least in a naive Lindhard susceptibility calculation, the distance between a density-of-states peak and $E_F$ is important because of the energy denominator. Our picture of the metamagnetic transition in UPt3 shows that the band 1 and band 2 van Hove singularities are the closest to $E_F$ at zero field, while the van Hove singularities $a$ to $e$ are further away. It may be interesting to see how the pairing fluctuations are affected by the position of these van Hove singularities relative to $E_F$, and in particular whether the strongest pairing fluctuations might be related to these particular van Hove singularities.

**Acknowledgments**

We are grateful to Andreas Hermann, Andrew Huxley and Hiroaki Ikeda, for helpful discussions. SRJ is grateful to NSERC for funding (RGPIN-2019-06446), and to Andrew Huxley and the University of Edinburgh for hosting a sabbatical visit. This work was supported by HFML-RU/NWO, member of the European Magnetic Field Laboratory (EMFL).

**Appendix. Tracking a low frequency quantum oscillation across $H_M$**

Here we provide the analysis behind figure 4 in the main body of the paper, in which a model is fitted to the quantum oscillations from a small extremal orbit as the orbit grows in size, with increasing magnetic field, across the metamagnetic transition.

Figure 10(a) shows raw magnetoresistance data, taken at 55 mK, with the external field $4^\circ$ from the $b$-axis in the $a$--$b$ plane, while panel (b) shows the oscillatory signal obtained by subtraction of cubic backgrounds. We have subtracted different backgrounds below and above $H_M \sim 20$ T, hence the break in the data at 20 T.

Below about 13 T and above 21 T there is a single dominant low frequency quantum oscillation, but the frequency is quite different above and below $H_M$: Fourier analysis gives a value of around 0.15 kT at 12 T vs 0.40 kT at 30 T.

Between about 13 T and 20 T the magnetoresistance is dominated by an even larger and lower-frequency oscillation, whose origin is not understood.

The question we must address is whether the low frequency quantum oscillation below 12 T comes from the same Fermi surface as the one above 21 T. They seem always to appear together at the same field-angles, which argues for them being from the same Fermi surface. Moreover the change in frequency might plausibly be explained as arising from non-linear spin-polarization of the Fermi surface, consistent with the picture of approximately rigid spin-splitting of the zero-field Fermi surface, and this was indeed suggested by Holleis et al [34], but they did not attempt a quantitative analysis.

Past studies of the quantum oscillations across a metamagnetic transition have focused on the back-projected frequency (see e.g. [41–44]), which is related to the textbook ‘Onsager’ frequency $F(B) = \hbar A(B)/2\pi e$, where $A(B)$ is the extremal area, by

$$
\sin(2\pi F(B)/B + \phi) \simeq
$$

$$
\sin(2\pi[F(B_c) + (B - B_c)F'(B_c)]/B + \phi) \simeq \sin(2\pi F_{obs}(B)/B + F'(B_c) + \phi),
$$

where $F'(B_c) = \partial F(B)/\partial B|_{B_c}$, and the so-called back-projected frequency, that is found when quantum oscillations...
from the measured $M$ vs $\sin(2\pi b)$ Oscillatory magnetoresistance, obtained by subtracting separate oscillations with a zero-crossing of the back-projected frequency. (blue line). The yellow arrow connects the turning point in the Comparison of the fitted oscillatory signal (red line) and the data large amount even across one oscillation, as explained below.

indeed around 23 T the oscillation frequency changes by a $F_{\text{simulated}}$ centred on a field $B_c$ are analyzed, is $F_{\text{obs}}(B_c) \equiv F(B_c) - B_c F'(B_c)$ [45]. If $F_{\text{obs}}(B_c)$ is changing slowly enough as a function of field, then Fourier analysis of short field-intervals of data can be used. ‘Slowly enough’ means having several oscillations with nearly the same frequency in each field interval. This is not the case with the oscillation in figure 10, indeed around 23 T the oscillation frequency changes by a large amount even across one oscillation, as explained below.

For this reason we attempted to fit the oscillations with a simulated $F(B)$, by assuming that the Fermi surface extremal area that produces $F(B)$ grows in proportion to the magnetization, $M(B)$. We digitized the magnetization data of reference [23] (blue circles in figure 10(c)), and then fitted $M(B)$ using a simple phenomenological interpolation formula consisting of three regions where $M(B)$ is linear in $B$ with smooth crossovers between these regions (this fit is the red line, right-hand axis, in figure 10(c)). We then assumed that $F(B)$ takes the form $F(B) = F(0) + aM(B)$, and adjusted $F(0)$ and $a$ to optimize agreement with the experimental quantum oscillation signal below 13 T and above 21 T. The resulting $F(B)$ is the red line/left-hand axis in figure 10(c). In figure 10(d) (figure 4(b) of the paper) the red line is the fit of $A(B) \sin(2\pi F(B)/B + \phi)$ to the oscillatory signal (blue line). It should be emphasized that we are focused on the frequency, not the amplitude, so in judging the fit the positions of maxima, minima and zero-crossings are the criterion, not the amplitude of the oscillations. (For $A(B)$ we used a simple quadratic dependence on $B$).

The correspondence between the calculation and the measurement in figure 10(d) is surprisingly good, including the unusual turning point in the oscillation near 23 T (marked with a yellow arrow). This turning point corresponds to a zero-crossing from negative to positive of the back-projected frequency (green line in figure 4(c)). It is in this region, around 23 T, that the back-projected frequency $F_{\text{obs}}(B)$ changes by a large amount during one period of the oscillation, a situation that can only be dealt with by direct fitting of the signal.

There are two regions where there are significant differences between the data (blue line in figure 10(d)) and the fit (red line in figure 10(d)). The first, above about 30 T, where the back-projected frequency is evidently lower than our fit predicts, may be due to our assumption that the Fermi surface cross-sectional area is linearly proportional to the magnetization. We adopted this model because it is simple and it works quite well, but at these high fields even a slight non-linearity in the magnetization, or in the $F(M)$ vs $M$ relationship, would be enough to produce the observed deviation, and so we do not regard this as a failure of the model. In the second region, between about 13 and 21 T, it seemed clear that our model would not fit, so we set any contribution to $\chi^2$ to zero in this field range. The behaviour of the magnetoresistance in this field range is not understood at this time. We have considered three scenarios, but given the complexity of the Fermi surface there may be other possibilities. Firstly, it can be seen in figure 10(c) that there is a second zero-crossing of the back-projected frequency between 15 and 16 T, close to where the largest deviation from the calculated line occurs, so at least some of this signal may arise from the same oscillation we are fitting, but with the amplitude and detailed field dependence being different from our simple model. Secondly, the Fermi surface that produces our low frequency oscillation in figure 10 must have a minority-spin branch. If this Fermi surface splits symmetrically, then the minority-spin surface would shrink to zero at approximately 12.5 T. It may be, however, that it does not split symmetrically, so the Lifshitz transition may take place above 13 T causing anomalies in the magnetoresistance. Finally, it has been seen in figure 5 that the band 1 Fermi surface undergoes some rather complicated reconstructions as a function of field, leading up to the final Lifshitz transition at which the central disk separates completely from the outer ring, and these too could cause anomalies in the magnetoresistance. There may be some hope, in the future, of disentangling

Figure 10. (a) Raw resistivity measured at 26° (4’ from b) at 55 mK. (b) Oscillatory magnetoresistance, obtained by subtracting separate cubic backgrounds from the raw $H < H_M$ and $H > H_M$ data. The yellow arrow indicates an unusual turning point in the oscillation. (c) The red line (left-hand axis) shows the field-dependent quantum oscillation frequency $F(B)$ obtained by assuming that $F(B) = F(0) + aM(B)$ where $F(0)$ and $a$ are fitting parameters and $M(B)$ is taken from the measured $M$ vs $\mu$, $H$ of Sugiyama et al (blue circles and right-hand axis) [23]. $F(0)$ and $a$ were obtained by fitting $A(H) \sin(2\pi F(B)/B + \phi)$ to the blue line in (b), excluding the region between 13 and 21 T, where other features dominate the oscillatory signal. $A(H)$ is taken to be a simple quadratic function of $H$, and is used only to make the visual comparison easier; the goal was to fit the frequency, not the amplitude. Also shown in (c) is $F_{\text{obs}}(B)$ (green line), the so-called back-projected frequency that corresponds to the instantaneous observed frequency of the oscillations. (d) Comparison of the fitted oscillatory signal (red line) and the data (blue line). The yellow arrow connects the turning point in the oscillations with a zero-crossing of the back-projected frequency.
this behaviour through careful study of the temperature dependence of the magnetoresistance, or other properties, between 13 and 21 T, but at present this is an unsolved problem.

Overall, however, as well as showing that a new low frequency oscillation does not appear suddenly at $H_M$ (in contrast to the expectation if Lifshitz transition a were to occur at $H_M$), success of this fit in the key regions below 13 T and above 21 T lends further support to a spin-polarized-band picture of the metamagnetism of UPt$_3$.

**References**

[10] Pfau H et al 2013 Interplay between kondo suppression and lifshitz transitions in YbRh$_2$Si$_2$ at high magnetic fields Phys. Rev. Lett. 110 256403
[18] Joynt R and Taillefer L 2002 The superconducting phases of UPt$_3$Rev. Mod. Phys. 74 235
[22] Sugiyma K et al 1999 Metamagnetic transition in UPt$_3$ studied by high-field magnetization and de Haas–van Alphen experiments Phys. Rev. B 60 9248
[38] Pippard A B 1989 Magnetoresistance in Metals (Cambridge: Cambridge University Press)
[40] Chandrasekaran A, Shtyk A, Betouras J J and Chamon C 2020
Catastrophe theory classification of fermi surface topological
transitions in two dimensions Phys. Rev. Res. 2 013355

The nature of elementary excitations below and above the
metamagnetic transition in CeRu2Si2 Phys. B 206–207 29

R and Onuki Y 1996 dHvA effect study of metamagnetic
transition in CeRu2Si2II—the state above the metamagnetic

[43] Aoki H, Takashita M, Kimura N, Terashima T. Uji S,
Matsumoto T and Onuki Y 2001 New features of the meta-
magnetic transition in CeRu2Si2 from the dhva effect under
high pressure J. Phys. Soc. Japan 70 774

phase of Sr3Ru2O7 Phys. Rev. Lett. 103 176401

[45] van Ruitenbeek J M, Verhoef W A, Mattocks P G, Dixon A E,
van Deursen A P J and de Vroomen A R 1982 A de Haas–van
Alphen study of the field dependence of the Fermi surface in