Impurity Scattering and Gap Structure in the Anisotropic Superconductor UPt$_3$

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Abstract

The thermal conductivity, $\kappa$, of the heavy fermion superconductor UPt$_3$ was measured down to $T_c/10$. The absence of a linear term in the temperature dependence as $T\to 0$ strongly suggests there are no zero energy quasiparticle excitations, in contradiction with the gapless behaviour often inferred from specific heat. A non-vanishing anisotropy ratio $\kappa_c/\kappa_b$ as $T\to 0$ establishes a new property of the gap structure: the presence of nodes along the c-axis. Furthermore, recent calculations by Fledderjohann and Hirschfeld show these cannot be point nodes with a linear k-dependence.

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One of the central endeavours in the field of unconventional superconductivity is the precise determination of the gap structure. Numerous efforts are currently being made to establish whether or not a d-wave gap is realized in the cuprate superconductors. In the heavy fermion superconductor UPt$_3$, a number of candidate states are still possible after 10 years of investigation. Any state with a line node in the basal plane of the hexagonal crystal structure, of which there are more than ten, is compatible with most of the existing data [1,2]. In particular, any such state is thought to be compatible with the compelling anisotropy of transverse sound attenuation [3]. A related issue of interest is the impact of impurity scattering on the low temperature properties of such superconducting states. The appearance of gapless behaviour, i.e. a residual density of quasiparticle excitations at T=0, even for small concentrations of non-magnetic impurities is under active investigation in the high T$_c$ cuprates. In heavy fermions, no detailed study has been made of this issue – even though the specific heat of UPt$_3$, for example, has often been held as indicative of a residual normal fluid in this compound – nor have there been quantitative comparisons with existing calculations.

The detailed nodal structure of the gap and the effect of impurity scattering will bear directly on the various scenarios for the multicomponent phase diagram of UPt$_3$ [2]. Both properties can only be studied at very low temperatures, typically in the region of T$_c$/10. In a previous paper [4], we showed how the thermal conductivity is a privileged probe of quasiparticles in UPt$_3$. In particular, it is sensitive to the anisotropy of the gap structure. Motivated by the recent calculations of Fledderjohann and Hirschfeld [5], we have now measured the thermal conductivity of UPt$_3$ down to T$_c$/10 and report two new facts: 1) there is no trace of a residual normal fluid at T=0 (at the 1% level) and 2) the gap must go to zero along the c-axis, in addition to the well-established line node in the basal plane. A comparison with the results of Fledderjohann and Hirschfeld further suggests that the gap cannot vanish linearly at a point, thereby weighing against the E$_{1g}$ representation for the superconducting order parameter of UPt$_3$ [2].

The details of the experiment and the crystal characteristics are described in Ref. [4].
The thermal conductivity $\kappa(T)$ of UPt$_3$ was measured for a heat current along each of the two high-symmetry directions of the hexagonal lattice ($J//b$-axis and $J//c$-axis), as a function of temperature down to $T_c/10 \approx 50$ mK. The low-temperature results are shown in Fig. 1, plotted as $\kappa/T$ vs $T$, while the overall behaviour is shown in the inset. The normal state behaviour below $T_c^+=0.5$ K was obtained by applying a magnetic field of $3 \ T > H_{c2}(0)$ (dashed lines). In this paper, we will only be concerned with the low temperature-low field phase B, which exists below $T_c^-=0.44$ K [2].

In order to use heat conduction as a measure of electronic transport one must ensure that phonons do not contribute significantly, i.e. that $\kappa_{ph}<<\kappa_e$ in the total conductivity $\kappa=\kappa_{ph}+\kappa_e$. The safest estimate of the maximum possible phonon contribution is obtained by using the formula $\kappa_{ph}=C_{ph}v_{ph}\Lambda_{ph}/3$, where $C_{ph}=\beta T^3$ is the low temperature phonon specific heat, $v_{ph}$ is the average sound velocity in the direction of the heat current and $\Lambda_{ph}$ is the phonon mean free path. One then assumes that $\Lambda_{ph}$ takes on its maximum value, namely the size of the crystal, equal to 0.7 mm [4]. From published data, $\beta=20$ J K$^{-4}$ m$^{-3}$ [6], in agreement with the known sound velocities [6]. The correct average velocity $v_{ph}$ [7] is 1880 m s$^{-1}$ for acoustic waves travelling along the b-axis and 1440 for the c-axis [6]. The maximum heat conduction by phonons in our crystal is therefore $\kappa_{ph}=85$ (67) T$^3$ mW K$^{-1}$cm$^{-1}$ for the b (c) axis. For $T<150$ mK, this represents at most 15% of the measured $\kappa_b$ and 6% of $\kappa_c$. Of course, scattering by quasiparticles, which are present in sufficient numbers to account for more than 85% of $\kappa$, will decrease $\Lambda_{ph}$ from this maximum possible value. This decrease will occur very rapidly as the temperature is increased, with $\Lambda_{ph}$ reaching a mere 5 $\mu$m at $T=T_c$ [4]. Therefore, $\kappa_{ph}$ is certainly less than the upper bounds of 15% and 6%, and can thus be neglected. We point out that while in our crystal the electronic mean free path is long enough to ensure that $\kappa_{ph}<<\kappa_e$, this is less likely to have been the case in previous measurements on polycrystalline UPt$_3$, where $\kappa_e$ was 4 to 6 times smaller [1], not to mention measurements on other heavy fermion compounds [1]. As a result, we believe this is the first time heat transport by heavy fermion quasiparticles is reliably measured down to $T_c/10$. This allows us to examine the possibility of a residual normal fluid at $T=0$ and it provides
us with a new and powerful probe of the gap structure.

Ideally, the question of a residual normal fluid should be answered by low-temperature measurements of the specific heat. Unfortunately, this has proven difficult both in UPt$_3$ and in the high T$_c$ cuprates, for two reasons. First, a number of sizeable non-linear contributions to C(T) exist at low temperature. In UPt$_3$, the quasiparticle contribution, although large, is completely overwhelmed below 100 mK by a huge upturn in C/T of ill-understood origin [8]. Secondly, even if a residual linear term is extracted reliably, it cannot automatically be attributed entirely to fermion excitations, for other mechanisms also lead to a linear specific heat. For UPt$_3$, the standard approach has been to extrapolate down from the roughly linear behaviour of C/T observed above about 100 or 150 mK [1,2,8], assuming it to persist down to T=0. This procedure yields an intercept at T=0, called $\gamma_0$, which typically ranges from 10 to 40% of the normal state value $\gamma_N$ in high quality samples [1,8]. In our crystal, $\gamma_0=16\%\gamma_N$ [9].

In recent years, several theories have been proposed for this extrapolated $\gamma_0$, taken to be an intrinsic property of the quasiparticle fluid in UPt$_3$ [1,10,11]. In light of the strong sample dependence, we are more inclined to regard this as an extrinsic property. For unambiguous evidence about a possible residual normal fluid at T=0, we therefore argue that one must turn to a more reliable measurement, such as heat conduction, free from the problems mentioned above. The observation of a finite $\kappa/T$ at T→0, such as seen in the high T$_c$ compound YBa$_2$Cu$_3$O$_7$, would be a direct indication of zero-energy quasiparticle excitations. It is clear from Fig.1 that a smooth extension of the $\kappa/T$ data to T=0 leads to a negligible intercept. Of course, strictly speaking, an abrupt flattening off of both curves below 40 mK cannot be excluded; in this case the largest linear term would represent 2% (4%) of its normal state value at T=0, equal to $L_o/\rho_o$ [4], for $J//c$ ($J//b$). We therefore conclude that, in the absence of any real sign of a residual linear term in our data, there is no evidence for gapless excitations in UPt$_3$.

More quantitatively, we note that at our lowest temperature T=0.1 T$_c$, $\kappa/\kappa_N=3\%$ (on average), whereas an extrapolated C/T for the same crystal gives $\gamma(T=0.1T_c)/\gamma_N=30\%$. 
Could it be that the mean free path due to impurity scattering in the superconducting state is 10 times smaller than in the normal state? One of the few measurements of the quasiparticle mean free path below $T_c$ comes from the degradation of a heat current by vortices. The increase in the thermal resistivity $1/\kappa$ with increasing magnetic field just above $H_{c1}$ is proportional to the density of vortices $B/\Phi_0$, the carrier mean free path $l_0$ and the vortex effective diameter $D$. From their measurement of this effect on a crystal of a quality similar to ours, Behnia et al. [12] obtained $l_0D \simeq 3000 \text{ nm}^2$ at $T=0.2$ K. If $D$ is taken to be the coherence length, then $D \simeq \xi(T = 0.2 K) \simeq \xi_0 \simeq 10 \text{ nm}$ [1,6], and $l_0 \simeq 300 \text{ nm}$. As we will see below, this is very close to the mean free path we estimate for the normal state. Therefore, it appears unlikely that the relaxation time in the superconducting state is suppressed by an order of magnitude.

The whole question of the impact of impurity scattering on unconventional gap structures, and vice versa, was treated by several authors in the mid-80s (see Refs.[1,13,14] and references therein). Within a weak-coupling BCS theory, Hirschfeld et al. [13] showed that a self-consistent treatment of impurity scattering for a gap with line nodes can lead to a residual density of quasiparticle states, showing up as a finite $C/T$ and $\kappa/T$ at $T=0$. Recent calculations of this kind were performed by Flederjohann and Hirschfeld [5] for three uniaxial gap structures, each with a line node in the basal plane: 1) a polar gap (with no other nodes) and 2) two hybrid gaps (with in addition a point node at each pole, i.e. along the c-axis). One of the hybrid gaps, which we call hybrid-I, vanishes linearly in $k$ at the point nodes, while the other, called hybrid-II, vanishes quadratically. Some of the states allowed by hexagonal symmetry to which these gaps correspond are listed in Table I. The calculations so far assume a single ellipsoidal Fermi surface and s-wave scattering, and they require two input parameters, the impurity scattering rate $\Gamma_0$ and the scattering phase shift $\delta_0$ [5]. Much information about the nature of the scattering can be obtained from the normal state properties. From the fact that the anisotropy of transport (both heat and charge) is independent of temperature, i.e. that $\kappa_c/\kappa_b=2.8$ and $\sigma_c/\sigma_b=2.7$ all the way from $T=0.1$ K (in the normal state) where elastic scattering dominates to $T=0.8$ K where inelastic electron-
electron scattering dominates [4], it is very likely that the anisotropy of 2.7-2.8 is due to the Fermi velocity, and both impurity-electron collisions and electron-electron collisions should be well-described by isotropic (s-wave) scattering [5]. From de Haas-van Alphen measurements (dHvA) [15], the Fermi surface is known to be made of several sheets, and a single ellipsoid is certainly an oversimplification. However, the usual Dingle plot analysis yields fairly uniform scattering rates, with $\tau = 1/\Gamma_{dHvA} = 2 - 4 \times 10^{-11}$ sec, in crystals of a quality comparable to ours [15]. In temperature, this corresponds to $\Gamma_{dHvA} = 0.2 - 0.4 \, T_c$. The scattering rate $\Gamma_0$ appropriate for transport, which is less sensitive to small-angle scattering and dephasing, will be smaller than $\Gamma_{dHvA}$ by a factor which depends on the type of scattering.

A value of $\Gamma_0 = 0.1 \, T_c$ seems quite reasonable, corresponding to $l_0 = v_F/\Gamma_0 = 400$ nm [15]. A separate estimate, obtained from the shear viscosity at $T_c$ [14], gives $\Gamma(T_c) = \Gamma_0(\rho(T_c)/\rho_0) = 0.22 \, T_c$, so that again $\Gamma_0 \approx 0.1 \, T_c$. Finally, Fledderjohann and Hirschfeld took $\delta_0 = \pi/2$, the unitary limit, seeing as weaker scattering leads to sharp disagreement with observed properties [5]. Theoretical arguments for such a limit can be found in Refs. [13] and [14], and in references therein.

The results of the calculations for $\kappa_b$ [5], for two values of $\Gamma_0$ (0.1 and 0.01 $T_c$) are reproduced in Fig.2, alongside our data, as a function of reduced temperature $T/T_c$. The data are plotted as $\kappa/\kappa_N = \kappa(H=0)/\kappa(H>H_{c2})$, where $\kappa_N = T/(a+bT^2)$ [4] and $T_c = T^-_{c_2} = 0.44$ K. Given that $b/a = 0.25 \, K^{-2}$ [4], $\kappa_N$ deviates from linear behaviour (elastic scattering) by only 7% at $T=0.3T_c$. This means that inelastic scattering can safely be neglected in the calculations below that temperature, shown therefore as $\kappa/T$ normalized to 1 at $T_c$. Only the curves for a polar and a hybrid-II gap structures are shown; the corresponding curves for the hybrid-I gap lie in between [5]. Inspection of both data and calculated curves reveals that the rapid increase in $\kappa/\kappa_N$ with temperature at such low $T/T_c$ – an order of magnitude faster than in a conventional superconductor [4] – is well reproduced by the calculations; this is a convincing confirmation of a line node in the basal plane of the gap structure of UPt$_3$ (phase B). However, the data show less curvature than either of the curves with $\Gamma_0/T_c = 0.1$ and, indeed, will not smoothly extrapolate to any significant intercept at $T=0,$
such as expected from the theory. In this sense, the observed behaviour is more compatible
with calculations based on a much smaller $\Gamma_0$, say 0.01 $T_c$, with a temperature dependence
close to that of the polar gap, although with a magnitude closer to the hybrid-II gap. Until
the real Fermi surface is used both in the calculations and in the estimates of a transport
$\Gamma_0$, related directly to the measured $\rho_0$, it is difficult to make firm conclusions from this
comparison. Nevertheless, it does seem as though an unreasonably low scattering rate is
needed to keep the number of zero energy quasiparticle excitations obtained in the current
self-consistent calculations at the low level observed in the experiment.

The anisotropy of electronic heat conduction has long been known to be a useful probe
of gap anisotropy, even in $s$-wave superconductors. In Ref.\cite{4} we showed how the ratio $\kappa_c/\kappa_b$
is a direct probe of the anisotropy of the gap in UPt$_3$, insofar as it is constant above $T_c$ and
starts falling immediately below $T_c$ (see inset of Fig.3). The fact that it decreases rather
than increases suggests there are more thermally excited quasiparticles with velocities along
the b-axis than along the c-axis. This could either result from a finite gap being larger
along c than along b (anisotropic $s$-wave gap), from the presence of nodes in the gap along
the b-axis in the absence of any along the c-axis (polar gap), or from the presence of nodes
along both axes (hybrid gap) provided the nodal structure is such that more quasiparticles
have $\mathbf{v}||\mathbf{b}$. This, for example, would exclude an axial gap (with only point nodes along
the c-axis) \cite{4}. Note, however, that the current discussion relies on the assumption that
the scattering rate does not change with temperature in some unexpected way below $T_c$.
To be free from such ambiguity, the analysis must be done in a regime where the strong
electron-electron scattering is not important, namely below 150 mK or so. Moreover, it is
in the limit of $T\rightarrow0$ that a measurement of $\kappa_c/\kappa_b$ becomes particularly useful. Indeed at
$T<<T_c$, the regions of the gap very close to the nodes dominate the thermal properties and
a measurement of heat conduction can then shed light on the detailed structure of the gap
in the vicinity of both high-symmetry directions. These are the main justifications for the
present low temperature study.

Our results for $\kappa_c/\kappa_b$ below 0.3 $T_c$ are shown in Fig.3, normalized at $T_c=\tilde{T}_c$ (i.e. divided
by 2.8, the constant value of the normal state anisotropy. The striking finding is that the ratio does not go to zero as $T \to 0$. This definitively excludes not only an axial gap, but also a polar gap. Indeed, the latter will give $\kappa_c/\kappa_b \to 0$ as $T \to 0$, as a result of the clear difference between excitation of quasiparticles with $v//c$ (across a finite gap) and with $v//b$ (in the vicinity of a line node). We conclude that the gap of UPt$_3$-phase B must have nodes along the c-axis. No previous experiment imposed such a requirement on the gap structure of this material, except perhaps the V-shaped gap features observed in recent studies of point-contact spectroscopy [16]. Indeed, all other measured properties are so far compatible with a polar gap [1].

A comparison with the calculations of Fledderjohann and Hirschfeld [5] allows us to go further and gain insight into the specific nodal structure near $k_x=k_y=0$. Their results on $\kappa_c/\kappa_b$ for the three uniaxial gaps with line nodes (polar, hybrid-I and hybrid-II) are shown in Fig.3. In the absence of gapless behaviour (i.e. for $\Gamma_0 = 0.01 \ T_c$), they find that $\kappa_c/\kappa_b \to 0$ as $T \to 0$ not only for the polar gap, as expected, but also for the hybrid-I gap. In essence, a gap vanishing at a point node with linear k-dependence does not cause as many quasiparticles to be excited thermally as a gap vanishing along a line. Remarkably, these authors found this not to be true for a point node with quadratic k-dependence, and the hybrid-II gap yields a finite value for $\kappa_c/\kappa_b$ as $T \to 0$. More specifically, such a gap on a spherical or ellipsoidal Fermi surface leads to no change in the anisotropy of heat conduction with respect to the normal state [5]. Note that the perfect isotropy will not be preserved for the real Fermi surface [5].

We conclude that the gap structure of phase B is unlikely to be of the hybrid-I type, as has been widely believed over the past few years [2]. Instead, our low temperature results favour a gap of the hybrid-II type, being the only uniaxial gap with the correct limiting behaviour of $\kappa_c/\kappa_b$ (see Table I). Of course, states with non-uniaxial symmetry should also be considered in future calculations. In the context of the 2D theory for the multicomponent phase diagram of UPt$_3$ (see Refs. [2] and [17], and references therein), this appears to disqualify the (1,i) state of the $E_{1g}$, $E_{2g}$ and $E_{1u}$ representations as possible candidates for
phase B, and leave only the (1,i) state of the $E_{2u}$ representation with $d$-vector parallel to the $c$-axis [17].

In summary, measurements of the thermal conductivity of UPt$_3$ down to $T_c/10$ have shed light on two important aspects of unconventional superconductivity: the possibility of a gapless behaviour and the nodal structure of the gap function. In the absence of any detectable linear term in $\kappa$ as $T \to 0$, we find no evidence for an intrinsic gapless regime, such as previously inferred from the specific heat. A comparison with calculations based on resonant impurity scattering and reasonable assumptions for the scattering strength and anisotropy leads to a significant discrepancy in the limiting value of $\kappa_b/T$. The unusual observation of a finite value for the anisotropy ratio $\kappa_c/\kappa_b$ as $T \to 0$ leads to new information about the gap structure of UPt$_3$ in phase B: the gap vanishes along the $c$-axis, and it does so with a special $k$-dependence, not compatible with a linear point node but probably so with a quadratic point node. This places the most stringent constraints so far on the possible superconducting states, and hence on the various theoretical models, for UPt$_3$.

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FIGURES

FIG. 1. Low-temperature thermal conductivity of UPt$_3$, divided by temperature, for a heat current along the c-axis (open circles) and the b-axis (solid circles). Inset: $\kappa/T$ up to 0.8 K. The normal state behaviour, obtained by applying a field above $H_{c2}$, is also shown (dashed lines).

FIG. 2. Thermal conductivity along the b-axis below 0.3 T$_c$, where T$_c$=T$_c^-$=0.44 K. Upper frame: data plotted as $\kappa/\kappa_N$, where $\kappa_N$=T(a+bT$^2$)$^{-1}$ (see text). Lower frame: $\kappa/T$ calculated for two uniaxial gaps with line nodes (polar and hybrid-II) and for two values of the impurity scattering rate $\Gamma_0$, 0.1 T$_c$ (solid lines) and 0.01 T$_c$ (dashed lines) (after Ref.[5]).

FIG. 3. Low-temperature behaviour of the anisotropy ratio $\kappa_c/\kappa_b$, plotted as a function of reduced temperature and normalized to unity at T$_c$. The data (points) are compared with calculations for three gap structures with a line node in the basal plane, with $\Gamma_0$=0.01 T$_c$ (after Ref.[5]). The dashed line is a linear fit to the data below 0.3 T$_c$. Inset: unnormalized data up to 0.8 K.
TABLE I. The gap structure of uniaxial states allowed by hexagonal symmetry (for strong spin-orbit coupling), and the limiting value of the anisotropy ratio $\kappa_c/\kappa_b$ expected as $T \to 0$, in the absence of gapless behaviour (see Ref. [5]). The nodal structures include a gap going to zero at a point along the $c$-axis, either with a linear (LP) or a quadratic (QP) $k$-dependence, and along a line in the basal plane (line node). Only odd-parity states with $d//c$ are listed (see Ref. [17]).

| Gap          | Nodal structure | Superconducting states         | $\kappa_c/\kappa_b$ as $T \to 0$ |
|--------------|-----------------|--------------------------------|-----------------------------------|
| Anisotropic s-wave | none            | $A_{1g} (\Delta_b < \Delta_c)$ | 0                                 |
| Axial I      | LP              | $E_{1u} (1, i)$                 | $\infty$                         |
| Axial II     | QP              | $E_{2g} (1, i)$                 | $\infty$                         |
| Polar        | line node       | $A_{1u}$                       | 0                                 |
| Hybrid I     | line node + LP  | $E_{1g} (1, i)$                 | 0                                 |
| Hybrid II    | line node + QP  | $E_{2u} (1, i)$                 | 1                                 |