

Determination of the gap structure in UPt₃ by thermal conductivity

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(Received 19 April 1995)

The thermal conductivity κ of the heavy-fermion superconductor UPt₃ was measured down to $T_c/10$, along the b and c axis of a single crystal. The anisotropy ratio κ_c/κ_b as $T \rightarrow 0$ is shown to be a powerful new probe of the gap structure in this compound. Our observation of a nonvanishing value for this ratio establishes a property of the gap in the B phase: the presence of nodes along the c axis. Furthermore, recent calculations by Fledderjohann and Hirschfeld strongly suggest these cannot be point nodes with a linear k dependence.

One of the central endeavors 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 UPt₃, 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 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 behavior, i.e., a residual density of quasiparticle excitations at $T=0$, 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 UPt₃, for example, has often been held as indicative of a residual normal fluid, nor have there been quantitative comparisons with existing calculations.

The detailed nodal structure of the gap and the effect of impurity scattering can best 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 UPt₃. 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 UPt₃ 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.

The details of the experiment and the crystal characteristics are given in Ref. 4. The thermal conductivity $\kappa(T)$ of UPt₃ was measured for a heat current along the b axis and the c axis, down to $T=T_c/10 \approx 50$ mK. The low-temperature results are shown in Fig. 1, plotted as κ/T vs T , while the overall behavior is shown in the inset. The error bars are less than the size of the data points, namely 5% at 50 mK and 1% at 150 mK, the main source of uncertainty being the calibration of the two thermometers on the sample. The normal state behaviour below $T_c^+=0.5$ K, called κ_N , was obtained by applying a magnetic field of $3 T > H_{c2}(0)$ (dashed lines). In this paper, we are only concerned with phase B (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. 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, and Λ_{ph} is the phonon mean free path. One then assumes that Λ_{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 (Ref. 6) and v_{ph} (Ref. 7) is 1880 (1440) m s⁻¹ for the b (c) axis.⁶ Therefore, the maximum $\kappa_{ph} = 85$ (67) T³ mW K⁻¹ cm⁻¹ for the b (c) axis. For $T < 150$ mK, this represents at most 15% of the measured κ_b and 6% of κ_c . Of course, scattering by quasiparticles will strongly decrease Λ_{ph} from this maximum possible value. Hence, κ_{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 $\kappa_{ph} \ll \kappa_e$, this is less likely to have been the case in previous

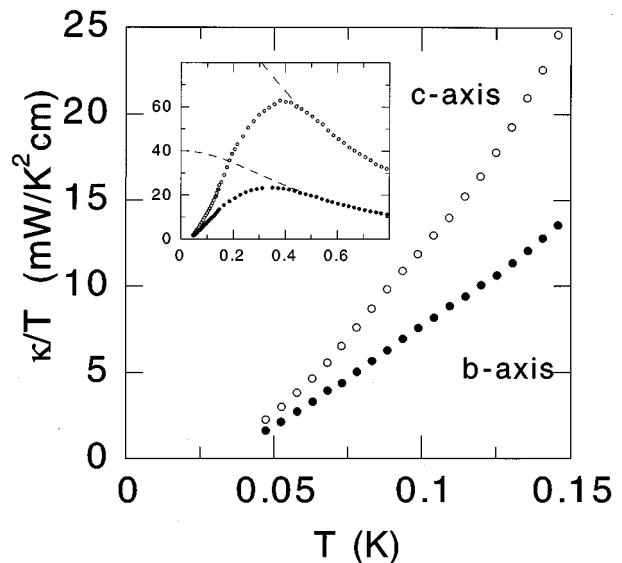


FIG. 1. Low-temperature thermal conductivity of UPt₃, divided by temperature, for a heat current along the c axis (open circles) and the b axis (solid circles). Inset: κ/T up to 0.8 K. The normal state behavior (κ_N), obtained by applying a field above H_{c2} , is also shown (dashed lines).

measurements on polycrystalline UPt_3 , where κ_c was 4–6 times smaller, 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, sizable nonlinear contributions to $C(T)$ exist at low temperature. In UPt_3 , the quasiparticle contribution, although large, is overwhelmed below 100 mK by a huge upturn in C/T of ill-understood origin.⁸ Second, even if a residual linear term is extracted reliably, it cannot automatically be attributed entirely to fermion excitations. For UPt_3 , the standard approach has been to extrapolate down from the roughly linear behavior of C/T observed above about 100 mK,^{1,2,8} assuming it to persist down to $T=0$. This procedure yields an intercept at $T=0$, called γ_0 , which typically ranges from 10 to 40% of the normal state value γ_N .^{1,8} In our crystal, $\gamma_0=16\% \gamma_N$.⁹

The observation of a finite κ/T at $T \rightarrow 0$ would be a direct indication of zero-energy quasiparticle excitations. From Fig. 1, it is clear that a smooth extension of the κ/T data to $T=0$ leads to a negligible intercept. More quantitatively, at $T=0.1T_c^-$, $\kappa/\kappa_N=2$ (4%) for $\mathbf{J} \parallel \mathbf{c}(\mathbf{b})$, or 3% on average, whereas an extrapolated C/T for the same crystal gives $\gamma(T=0.1T_c)/\gamma_N=30\%$. This order-of-magnitude discrepancy, combined with the strong sample dependence of γ_0 , certainly suggests that such a γ_0 is of extrinsic origin. To settle this point, however, a detailed comparison with theory is needed.

The impact of impurity scattering on unconventional gap structures was treated by several authors in the mid 1980s (see Refs. 1, 10 and 11 and references therein). Within a weak-coupling BCS theory, Hirschfeld *et al.*¹⁰ 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 κ/T at $T=0$. Recent calculations of this kind were performed by Fledderjohann and Hirschfeld⁵ 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 Γ_0 and the scattering phase shift δ_0 .⁵ The assumption of isotropic scattering is supported by the fact that both elastic and inelastic processes give the same anisotropy in normal state transport, i.e., $\kappa_c/\kappa_b=2.8$ and $\sigma_c/\sigma_b=2.7$ from $T=0.1$ K to 0.8 K, which can therefore be attributed to an anisotropy in the Fermi velocity.^{4,5} From de Haas–van Alphen (dHvA) measurements,¹² the Fermi surface is known to be made of several sheets, and a single ellipsoid is certainly an oversimplification. However,

TABLE I. The gap structure of uniaxial states allowed by hexagonal symmetry (for strong spin-orbit coupling and ellipsoidal Fermi surface), and the limiting value of the anisotropy ratio κ_c/κ_b expected as $T \rightarrow 0$, in the absence of gapless behavior (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. Only those odd-parity states with $\mathbf{d} \parallel \mathbf{c}$ are listed (see Ref. 14).

Gap	Nodes	States	κ_c/κ_b
S wave	None	$A_{1g} (\Delta_b < \Delta_c)$	0
Axial I	LP	$E_{1u}(1,i)$	∞
Axial II	QP	$E_{2g}(1,i)$	∞
Polar	Line	A_{1u}	0
Hybrid I	Line + LP	$E_{1g}(1,i)$	0
Hybrid II	Line + QP	$E_{2u}(1,i)$	1

the usual Dingle plot analysis yields fairly uniform scattering rates, with $\tau = 1/\Gamma_{\text{dHvA}} = 2-4 \times 10^{-11}$ s, in crystals of a quality comparable to ours.¹² In temperature, this corresponds to $\Gamma_{\text{dHvA}} = 0.2-0.4 T_c$. The scattering rate Γ_0 appropriate for transport will be smaller than Γ_{dHvA} by a factor which depends on the type of scattering. A value of $\Gamma_0 = 0.1T_c$ seems reasonable, corresponding to $l_0 = v_F/\Gamma_0 = 400$ nm.¹² A separate estimate, obtained from the shear viscosity at T_c ,¹¹ gives $\Gamma(T_c) = \Gamma_0[\rho(T_c)/\rho_0] = 0.22T_c$, so that again $\Gamma_0 \approx 0.1T_c$. Of course, these estimates of Γ_0 could be off by a factor 2 or so, but hopefully not by much more. As for δ_0 , Fledderjohann and Hirschfeld assumed the unitary limit of $\pi/2$; theoretical arguments for such a limit can be found in Refs. 10 and 11, and in references therein.

The calculated κ_b ,⁵ for two values of Γ_0 (0.1 and $0.01T_c$) is reproduced in Fig. 2, along with our data. The data are plotted as κ/κ_N vs T/T_c , where $\kappa_N = T/(a + bT^2)$ (Ref. 4) and $T_c = T_c^- = 0.44$ K. Given that $b/a = 4.0 \text{ K}^{-2}$,⁴ κ_N deviates from linear behavior (elastic scattering) by only

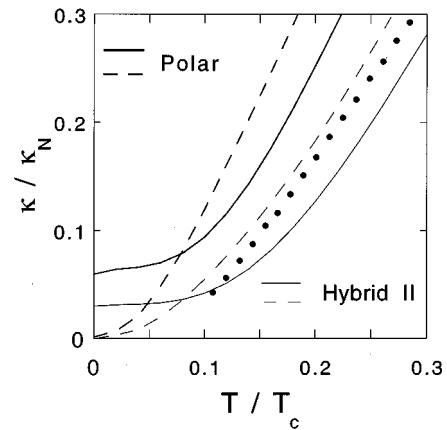


FIG. 2. Thermal conductivity along the b axis, normalized by the normal state conductivity $\kappa_N(T)$, for temperatures below $0.3T_c$, where $T_c = T_c^- = 0.44$ K. The data (points) are compared with calculations for two uniaxial gaps with line nodes (polar and hybrid II) and for two values of the impurity scattering rate Γ_0 , $0.1T_c$ (solid lines) and $0.01T_c$ (dashed lines) (after Ref. 5).

7% at $T=0.3T_c$. This means that inelastic scattering can safely be neglected below that temperature. The calculations of Fig. 2 assume elastic scattering only, so that $\kappa_N(T) = T[\kappa(T_c)/T_c]$. 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 Fig. 2 reveals that the rapid increase in κ/κ_N with temperature, 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 UPt₃ (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 behavior is more compatible with calculations based on a smaller Γ_0 , such as $0.01T_c$. Until the real Fermi surface is used both in the calculations and in the estimates of a transport Γ_0 , related directly to the measured ρ_0 , it is difficult to make firm conclusions from this comparison. Nevertheless, it does seem as though a very 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. Finally, we stress that although the calculation for a hybrid-II gap with $\Gamma_0 = 0.01T_c$ can account reasonably well for the observed $\kappa_b(T)$ at low temperatures, it also does for the hybrid-I gap, so that one cannot discriminate between the two types of gap on this basis alone.

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. 4 we showed how the ratio κ_c/κ_b is a direct probe of the anisotropy of the gap in UPt₃, 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 $\mathbf{v} \parallel \mathbf{b}$. 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 $T \rightarrow 0$ that a measurement of κ_c/κ_b becomes particularly useful. Indeed at $T \ll T_c$, 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.

Our results for κ_c/κ_b below $0.3T_c$ are shown in Fig. 3, normalized at $T_c = T_c^-$. The striking finding is that *the ratio does not go to zero as $T \rightarrow 0$* . Instead, it extrapolates to a large finite value (between 0.4 and 0.5). This definitively excludes a polar gap, which gives $\kappa_c/\kappa_b \rightarrow 0$ as $T \rightarrow 0$, as a result of the clear difference between excitation of quasiparticles with $\mathbf{v} \parallel \mathbf{c}$ (across a finite gap) and with $\mathbf{v} \parallel \mathbf{b}$ (in the

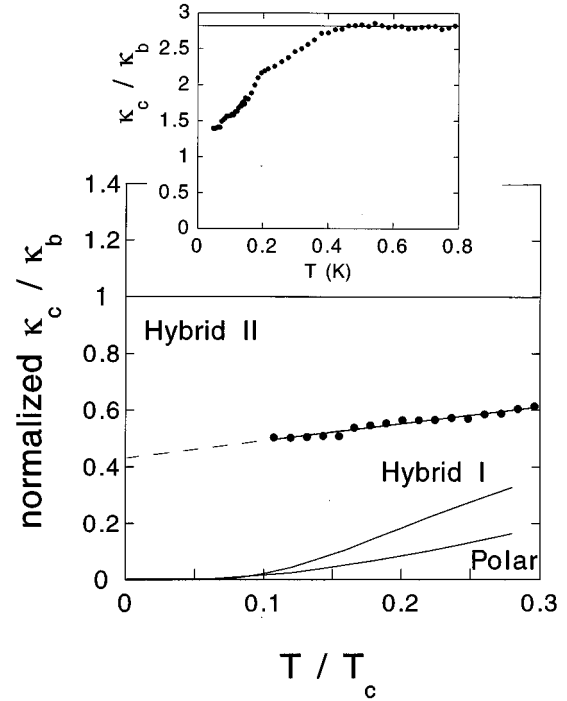


FIG. 3. Low-temperature behavior of the anisotropy ratio κ_c/κ_b vs reduced temperature and normalized to 1 at T_c . The data (points) are compared with calculations for gap structures with a line node in the basal plane, with $\Gamma_0 = 0.01T_c$ (after Ref. 5). The dashed line is a linear fit to the data below $0.3T_c$. Inset: unnormalized data up to 0.8 K.

vicinity of a line node). We conclude that the gap of the UPt₃ phase B must have nodes along the *c* axis. With the possible exception of recent studies of point-contact spectroscopy,¹³ no previous experiment could discriminate between a polar and a hybrid gap.¹

A comparison with the calculations of Fledderjohann and Hirschfeld⁵ allows us to go further and gain insight into the specific nodal structure near $k_x = k_y = 0$. Their results on κ_c/κ_b for the three uniaxial gaps with line nodes are shown in Fig. 3. In the absence of gapless behavior (e.g., when $\Gamma_0 = 0.01T_c$), they find that $\kappa_c/\kappa_b \rightarrow 0$ as $T \rightarrow 0$ 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 κ_c/κ_b as $T \rightarrow 0$. Specifically, such a gap on a single ellipsoidal Fermi surface leads to no change in the anisotropy below T_c .⁵ Although this perfect isotropy will not hold for the real Fermi surface,⁵ the finite limit is certainly expected to.

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 favor a gap of the hybrid-II type, being the only uniaxial gap with the correct limiting behavior of κ_c/κ_b (see Table I). Of course, states with nonuniaxial symmetry should also be considered in future calculations. In the context of the 2D theory for the multicomponent phase diagram of UPt₃ (see Refs. 2

and 14, 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.¹⁴ Note, however, that the real gap structure associated with the above states is expected to be more complicated than the simple gaps considered so far. Calculations with the real Fermi surface are needed to determine the effect of additional structure in the gap for the various representations.

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 behavior and the nodal structure of the gap function. In the absence of any detectable linear term in κ as $T \rightarrow 0$, we find no clear evidence for an intrinsic gapless regime. A comparison with calculations based on resonant impurity scattering indicate that the elastic scattering rate in our crystal is small (less than $T_c/10$). The unusual observation of a finite value for the anisotropy ratio κ_c/κ_b as $T \rightarrow 0$ 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.

Note added. After this paper was written, extensive calculations including the real Fermi surface of UPt_3 were reported by Norman and Hirschfeld.¹⁵ Their results show that both E_{1g} (hybrid-I) and E_{2u} (hybrid-II) gaps can account for $\kappa(T)$ if Γ_0 is chosen to be less than $0.1T_c$. However, a large value of the anisotropy ratio (around half of the normal state value) as $T \rightarrow 0$ is seen to favor the E_{2u} gap, at least in the limit of low scattering rates (lower than $0.1T_c$).

We are grateful to A. Fledderjohann and P. J. Hirschfeld for extensive discussions and for allowing us to reproduce their calculations here. This work was funded by NSERC of Canada and FCAR of Québec. L.T. acknowledges the support of the Canadian Institute for Advanced Research and the A.P. Sloan Foundation.

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