Anisotropy of Heat Conduction in the Heavy Fermion Superconductor UPt_3

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We report on the first measurement of the anisotropy of heat conduction in a heavy fermion superconductor, performed on a single crystal of UPt_3 with a current parallel and perpendicular to the hexagonal axis. Beyond the temperature-independent anisotropy of the normal state, a clear additional anisotropy develops in the superconducting state. This direct measure of gap anisotropy places precise constraints on the possible states for the two zero-field phases. An axial gap is excluded for both, and a comparison with existing calculations favors a d-wave gap for the low-temperature phase.

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The occurrence of superconductivity in strongly correlated $5f$ electron systems such as the heavy fermion compounds UBe_{13} ($T_c = 0.9$ K), URu_2Si_2 ($T_c = 1.2$ K), and UPt_3 ($T_c = 0.5$ K) has intrigued researchers for a decade. The first indication that these systems cannot be described by the standard theory (with an s-wave order parameter and a finite gap at all points on the Fermi surface) came from the nonexponential temperature dependence of the specific heat, NMR relaxation rate, London penetration depth, ultrasound attenuation, and other properties below T_c. Although this points to the presence of nodes in the superconducting gap, little more can be stated with confidence about the precise gap symmetry and position of nodes [1]. For a more direct access to the anisotropy of the gap, a combination of a directional probe and single crystals must be adopted. Over the years, several attempts have been made to use this more powerful approach. As yet, they have all been inconclusive [2], with one exception. Shivaram *et al.* [3] measured the attenuation of transverse sound propagating in the hexagonal basal plane of UPt_3 for polarizations in and out of the plane ($\mathbf{e}//\mathbf{c}$ and $\mathbf{e}\perp\mathbf{c}$). They found a clear difference developing below T_c, with a linear temperature dependence for $\mathbf{e}\perp\mathbf{c}$ and a higher power law dependence for $\mathbf{e}//\mathbf{c}$. Their results have led to the conclusion that the gap structure of UPt_3 has a line of nodes in the basal plane (see Ref. [1]).

Since then, three distinct superconducting phases were discovered in UPt_3, with two transitions in zero magnetic field and ambient pressure at $T_c^+ = 0.5$ K (into phase A) and $T_c^- = 0.44$ K (into phase B), and a field-induced phase C [1,2]. Most theoretical models for the multicomponent phase diagram are based on a coupling between the superconducting order parameter and the antiferromagnetic moment which appears below $T_N = 5$ K (see Refs. [4–6], and references therein). By breaking the symmetry of the lattice, the coupling is thought to lift a degeneracy of the order parameter and thus produce two transitions. These models assign to each phase a certain superconducting state, and hence to each a specific gap structure. Theories based on a two-dimensional (2D) representation [4,5] assign to phase B either an "axial" gap (with point nodes along the c axis), or a "hybrid" gap (with, in addition, a line of nodes in the basal plane). The theory of Machida *et al.* [6] based on a 1D representation requires either a hybrid gap or a "polar" gap (with only a line of nodes at $k_z = 0$).

In order to provide a firm assessment of gap structure, we have used heat conduction as a probe of gap anisotropy in UPt_3. For the first time in a heavy fermion superconductor, two independent components of the thermal conductivity tensor, κ_{xx} and κ_{zz} ($\mathbf{x}//\mathbf{a},\mathbf{b}$ and $\mathbf{z}//\mathbf{c}$), were measured on the same single crystal. The results show an unambiguous additional anisotropy below T_c which excludes an axial gap and favors a hybrid gap for the low-temperature phase B.

Previous studies of thermal conductivity in UPt_3 all arrive at the same basic fact (see Refs. [7] and [8], and references therein): from $T = 0$, $\kappa(T)$ increases with temperature, an order of magnitude faster than in the standard BCS theory. Quasiparticles are thermally excited much more easily than would be possible for a gap without nodes. To go beyond this general fact, a study of heat transport along different directions of a single crystal is necessary. Behnia *et al.* investigated heat conduction along both the b and the c axes of UPt_3 [8], but the fact that two different crystals (with significantly different impurity levels) were used precluded any firm conclusion about the intrinsic anisotropy of κ.

The thermal conductivity was measured using the steady-state technique with two RuO_2 thermometers. A single crystal of cylindrical shape (20 mm length, 6 mm diam) was grown by Czochralski pulling in ultrahigh vacuum. Two sections (of length 2 mm) separated by 10 mm were cut out and then annealed at 1200 °C for six days. Their specific heat was measured between 0.15 and 1.0 K and found to be identical (within 1%), thus confirming homogeneity. The two well-separated transition jumps are complete at $T_c^+ = 0.50$ K and $T_c^- = 0.44$ K, with respective widths of 25 and 20 mK. Out of one section, two small and adjacent rectangular pieces were spark cut, each with dimensions $2.0 \times 0.7 \times 0.7$ mm^3. The length of one is parallel to the c axis and that of the other to the b axis.

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Their high purity is attested to by the low residual resistivity: for $\mathbf{J}//\mathbf{c}$, $\rho_0 = 0.23$ $\mu\Omega$ cm, the second lowest value reported for a bulk crystal.

The two components of the thermal conductivity, κ_b and κ_c, are displayed in Fig. 1 as a function of temperature. The normal state behavior obeys $\kappa_N(T)/T = (a + bT^2)^{-1}$, with $a = 0.25$ (0.09) m K^2 W^{-1} and $b = 1.0$ (0.37) m W^{-1} for $\mathbf{J}//\mathbf{b}$ ($\mathbf{J}//\mathbf{c}$). Quasiparticle scattering gives rise to the T^2 term, which grows to be as large as the elastic impurity component at T_c (i.e., $bT_c^2 = a$). From Fig. 2, where κ/T is normalized at $T_c = 0.5$ K, it is seen that the anisotropy is *independent of temperature* above T_c, and $\kappa_c/\kappa_b = 2.8$ from 0.5 to 0.8 K within 2%.

The anisotropy in the electrical resistivity was *also* found to be independent of temperature. The resistivity obeys $\rho(T) = \rho_0 + AT^2$ perfectly below 0.8 K, with $\rho_0 = 0.61$ (0.23) $\mu\Omega$ cm and $A = 1.60$ (0.59) $\mu\Omega$ cm K^{-2} for $\mathbf{J}//\mathbf{b}$ ($\mathbf{J}//\mathbf{c}$). The anisotropy in both elastic and inelastic components is virtually the same: 2.65 and 2.71, respectively, and $\rho_b/\rho_c = 2.7$ to within 1% from 0.5 to 0.8 K. This means that the Lorenz number $L = \kappa\rho/T$, although strongly temperature dependent, is nearly isotropic. By applying a field of 3 T [i.e., above $H_{c2}(0)$] and correcting for the known magnetoresistance, κ_N was measured down to 0.1 K, to give $L(0.1\text{ K}) = 0.99L_0$, as expected from the Wiedemann-Franz law (i.e., $\rho_0/a = L_0 = 2.44 \times 10^{-8}$ W Ω K^{-2}), while $L(0.8\text{ K}) = 0.75L_0$. These numbers suggest that the phonon contribution, κ_{ph}, to the total thermal conductivity, $\kappa = \kappa_e + \kappa_{ph}$, is very small in the normal state. Indeed, with nearly isotropic sound velocities [9], a κ_{ph} greater than 2% of κ_b at 0.5 K would show up as a detectable change in anisotropy at 0.8 K (Fig. 2). This implies that $\kappa_{ph}(T_c) < 0.2$ mW K^{-1} cm^{-1}. In fact, the phonon conductivity is well below this upper bound. Franse *et al.* [7] measured $\kappa(T)$ at temperatures where κ_{ph} becomes sizable. From their data, one can extract $\kappa_{ph} \approx 0.5\kappa = 4$ mW K^{-1} cm^{-1} at 5 K. The T^2 dependence due to electron scattering (the dominant process below 10 K) then yields $\kappa_{ph}(T_c) = 0.04$ mW K^{-1} cm^{-1}, namely 0.4% of our measured $\kappa_b(T_c)$.

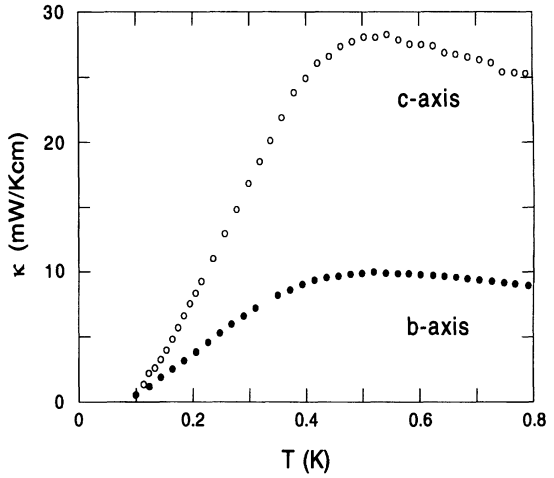

FIG. 1. Thermal conductivity of UPt_3 for a heat current along two directions of the hexagonal crystal lattice: $\mathbf{J}//\mathbf{c}$ (open circles) and $\mathbf{J}//\mathbf{b}$ (solid circles).

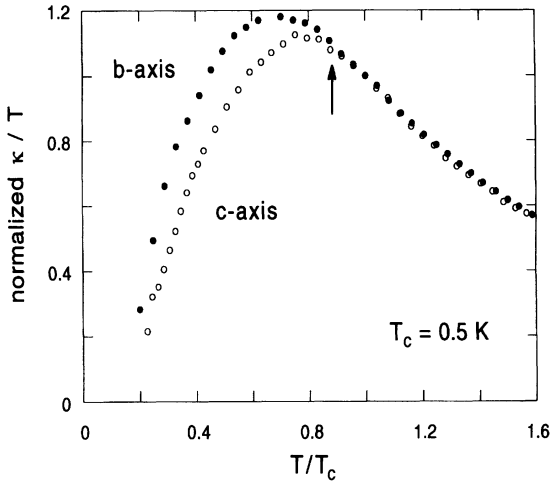

FIG. 2 Thermal conductivity divided by temperature vs reduced temperature for the two directions of heat current. κ_b and κ_c are both normalized to unity at $T = 0.5$ K. Note the additional anisotropy appearing below T_c^- (arrow).

The normal state of UPt_3 is often characterized as a Fermi liquid: there is a Fermi surface and, the specific heat is linear in temperature ($C/T = \gamma$) below 1.5 K. The coefficient γ is enormous, due to the huge renormalized masses. It is interesting to ask whether the electrons responsible for the large γ are also the carriers of heat. From de Haas–van Alphen measurements on crystals of similar quality [10], the Fermi velocity and mean free path of electrons on the largest surface (which accounts for most of the density of states) are $v_F = 5500$ m/s and $l_0 = 220$ nm. With $\gamma = 0.422$ J K^{-2} mol^{-1} [9], the thermal conductivity of quasiparticles is $\kappa_e/T = \frac{1}{3}\gamma v_F l_0 = 4.0$ W K^{-2} m^{-1} at $T < 0.1$ K. The measured value (in the normal state) is $\kappa_N/T = L_0/\rho_0 = 4.0$ W K^{-2} m^{-1}, for $\mathbf{J}//\mathbf{b}$. Although this perfect agreement is fortuitous, it is a confirmation that the Fermi liquid picture of heavy and itinerant quasiparticles is quantitatively consistent in UPt_3.

Our main result is the additional anisotropy which develops in the superconducting state, as seen in Fig. 2. The large, temperature-independent anisotropy of the normal state is considerably reduced at low temperature (Fig. 1). This is made evident in Fig. 3, where the ratio $\kappa_c(T)/\kappa_b(T)$ is plotted as a function of temperature. *Such a change can only be due to gap anisotropy.* A conventional "s-wave" gap $\Delta(\mathbf{k})$, although finite everywhere on the Fermi surface, can be smaller for certain $\mathbf{k}$ directions and thus lead to an anisotropy in the electronic thermal conductivity κ_e below T_c. This is what happens in zinc [11], where $\Delta(\mathbf{k})$ is smaller for $\mathbf{k}$ in the

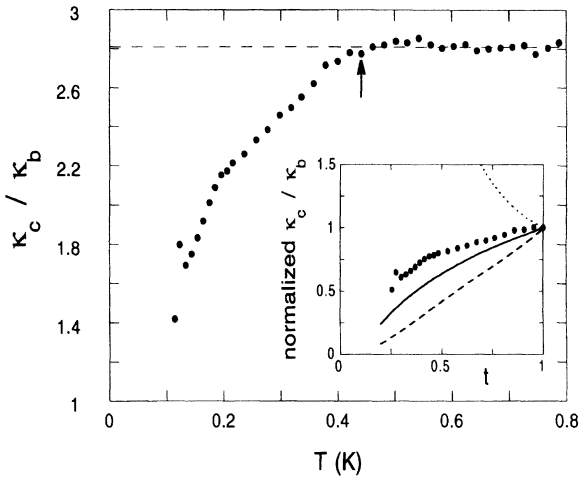

FIG. 3. Anisotropy ratio vs temperature. The constant anisotropy of the normal state drops, below T_c^- (arrow) excluding the possibility of an axial gap for phase B. Inset: κ_c/κ_b normalized at T_c^- vs $t = T/T_c^-$; data (circles), and theoretical curves for three gap structures: axial (dotted), polar (dashed), and d wave (solid) (after Ref. [16]).

hexagonal basal plane. An unconventional gap, however, which actually vanishes for certain **k** directions, will not only lead to an anisotropy in κ_e, governed by the distribution of nodes, but will cause κ_e to rise with temperature much more rapidly, reflecting the ease with which quasiparticles with **k** vectors near the nodes can be thermally excited. And indeed, it is the sheer magnitude of κ_e in UPt_3 at low temperature which distinguishes it from conventional superconductors. From standard BCS theory [12], κ_e/κ_N—the electronic thermal conductivity calculated for impurity scattering only, normalized by the normal state conductivity—increases very slowly at first and reaches only 4% of its value at T_c by $T = T_c/4$. This is what is observed in pure Al [12], and similarly for other pure superconductors with low T_c/θ_D: 3% in Nb [13], 2%–3% in Sn [14], and 1%–2% in Zn and Cd [11]. On the other hand, in UPt_3, κ/κ_N has already risen to 27% of its normal state value at $T_c/4$ (**J**//**b**). More than any power law dependence, this order-of-magnitude difference in the rate of thermal excitation of quasiparticles is compelling evidence for nodes in the gap. Of course, this is reflected in other properties; for the electronic specific heat, $C_{es}/\gamma T_c = 1/40$ at $T_c/4$ in standard superconductors [12], while it equals 1/8 in UPt_3 [1].

The above arguments assume that κ is mostly electronic below T_c ($\kappa_{ph} \ll \kappa$). Let us verify this. We have shown how κ_{ph} at T_c is approximately 0.4% of the measured $\kappa_b(T_c)$. In the superconducting state, the number of electrons that scatter phonons drops. In UPt_3, the rate of decrease is much slower than in BCS superconductors, and from the specific heat one can argue that the number has dropped 5 times less at $T_c/4$. In Nb [13], $\kappa_{ph}(T_c/4) = 12\kappa_{ph}(T_c)$, so that in UPt_3 one might roughly expect $\kappa_{ph}(T_c/4) = 12/5\kappa_{ph}(T_c) = 0.1\ \mathrm{mW\,K^{-1}\,cm^{-1}}$, which is 5% (8%) of the measured κ_c (κ_b) at $T_c/4 = 125$ mK. The proportion will rapidly get smaller at higher temperatures. Admittedly, this is a rough estimate of κ_{ph} in the superconducting state, but greater accuracy requires a full-fledged calculation. The essential point is that even a 20%–30% contribution from phonons at $T_c/4$, which drops to less than 1% at T_c, would have little influence on our analysis and conclusions. For example, if $\kappa_{ph} = 0.3\kappa_b$ at 125 mK, the *electronic* κ_c/κ_b would equal 1.65, instead of the measured 1.5 (Fig. 3), a 10% correction which decreases rapidly. It is because phonons can be neglected that the thermal conductivity is a privileged probe of quasiparticle excitation in this superconductor.

Now that we can exclude a conventional s-wave gap, the position of the nodes is the question of interest. A careful distinction must be made between phase A (from T_c^- to T_c^+) and phase B (below T_c^-). Inspection of the data (see Fig. 2) reveals little change in either κ_b or κ_c with the appearance of phase A. This unusual insensitivity is suggestive of a nodal structure isotropic in the *b-c plane* (such as a line in that plane). In contrast, the anisotropy κ_c/κ_b starts to drop with the onset of phase B at T_c^- (Figs. 2 and 3). This may be an indication of a different nodal structure in the gap of phases A and B. That κ_c/κ_b *drops* allows us to discard unambiguously the possibility of an axial gap for phase B. This immediately eliminates two of the four candidate representations for the order parameter in the "2D theory" [4,5] for the phase diagram of UPt_3. Indeed, the $(1, i)$ state attributed to phase B has an axial gap in both the E_{1u} and E_{2g} representations. On the other hand, it has a hybrid gap in the E_{1g} and E_{2u}. In order to determine what gap structures are truly compatible with our results, detailed calculations are essential. Assuming resonant scattering of quasiparticles by impurities, Hirschfeld, Wölfle, and Einzel [15] and Arfi, Bahlouli, and Pethick [16] calculated $\kappa_b(T)$ and $\kappa_c(T)$ for three simple gap structures: $\Delta(\hat{\mathbf{k}}) = \Delta_{max}\cos\theta$ (polar), $\Delta_{max}e^{i\phi}\sin\theta$ (axial), and $2\Delta_{max}e^{i\phi}\sin\theta\cos\theta$ ("d wave"). A direct comparison with their calculations is not strictly possible, mainly because of their neglect of inelastic processes. Since that may not have much of an effect on the anisotropy, we begin by comparing our data with the ratio κ_c/κ_b obtained from the results of Arfi, Bahlouli, and Pethick [16]. This is shown in the inset of Fig. 3, for the three gap structures. The axial gap can obviously be discarded. The polar gap yields much too rapid a drop, and unless electron scattering and Fermi surface effects prove to have a dramatic impact on the anisotropy, this would seem to disqualify all but one of the candidate states in the "1D theory" [6]. Although the data is 30% more isotropic than the d-wave prediction, the comparison argues convincingly for a hybrid nodal structure. Whether it is this particular gap, associated with the $k_z(k_x + ik_y)$ state of the E_{1g} representation [4,5], which is ultimately confirmed, or the hybrid gap of the A_{2u} [6] or E_{2u} [5], with their

different **k** dependence, can be established only by further calculations. The temperature dependence of κ_c/κ_b is a new and potentially decisive element in the current debate over 1D vs 2D and E_{1g} vs E_{2u} scenarios [5,6].

In Fig. 4 we compare the separate $\kappa_b(T)$ and $\kappa_c(T)$ data with the calculations of Ref. [16] for the d-wave gap, assuming resonant impurity scattering in the unitarity limit (phase shift of $\pi/2$). Both the data (circles) and the calculated curves (lines) are plotted as $[\kappa(T)/T]/[\kappa(T_c)/T_c]$. T_c is taken to be $T_c^- = 0.44$ K, and the comparison applies to phase B. Because of inelastic scattering, it can only be qualitative. Nonetheless, the overall temperature dependence is not far off. The fact that the data lies above the theoretical curves is due to the neglect of inelastic scattering. As a lowest order treatment, we "take out" the scattering time by normalizing with $\kappa_N(T)$. As seen in the inset of Fig. 4, the d-wave calculation gives a correct overall temperature dependence for κ/κ_N, albeit with an exaggerated anisotropy.

We conclude that phase B of UPt_3 most probably has a hybrid gap, the anisotropy of which was directly revealed. The only other direct measure of gap anisotropy, the early transverse ultrasound study of Shivaram *et al.* [3], is consistent with our findings for phase B. Given more realistic calculations, our results can lead to a greater precision in the identification of the gap, with the prospect of further discriminating between the allowed representations for the order parameter.

A distinguishing feature of a gap with nodes is the possibility of a finite linear term in $\kappa(T)$ at low temperatures even for a small amount of impurities [15]. Although there is no indication of this in our data, a residual κ/T could still be present and show up below $T_c^-/5$. If it exists, it is at most 20% of the value at T_c^-, or 10% of κ_N/T at $T = 0$.

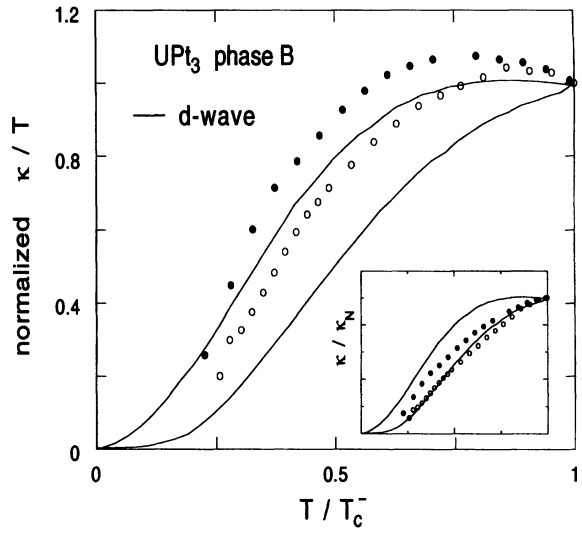

FIG. 4. Thermal conductivity of phase B vs reduced temperature T/T_c^-, for **J**//**c** (open circles) and **J**//**b** (solid circles). The data are plotted as κ/T and normalized at $T_c^- = 0.44$ K. The solid lines are calculations by Arfi, Bahlouli, and Pethick [16], for a d-wave gap. Inset: the same calculated curves, compared with $\kappa(T)/\kappa_N(T)$ (circles).

In summary, for the first time the thermal conductivity was used to establish the presence and help determine the position of nodes in the gap structure of a superconductor. From the drop in anisotropy observed upon entering the superconducting phase B of UPt_3, the possibility of an axial gap associated with that phase is definitively excluded. Instead, a comparison with existing calculations strongly favors a hybrid gap, with both a line of nodes in the basal plane and point nodes along the c axis. There appears to be negligible b-c anisotropy in the gap structure of phase A. There is no evidence of a residual linear term at $T = 0$ which would indicate a finite density of zero-energy quasiparticle excitations. Further calculations, including quasiparticle-quasiparticle scattering and the real Fermi surface, applied to all potential gap structures would allow a critical assessment of current theories for the phase diagram.

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