Twofold Spontaneous Symmetry Breaking in the Heavy-Fermion Superconductor UPt_3

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The field-orientation dependent thermal conductivity of the heavy-fermion superconductor UPt_3 was measured down to very low temperatures and under magnetic fields throughout the distinct superconducting phases: B and C phases. In the C phase, a striking twofold oscillation of the thermal conductivity within the basal plane is resolved reflecting the superconducting gap structure with a line of node along the a axis. Moreover, we find an abrupt vanishing of the oscillation across a transition to the B phase, as a clear indication of a change of gap symmetries. We also identify extra two line nodes below and above the equator in both B and C phases. From these results together with the symmetry consideration, the gap function of UPt_3 is determined as a E_{1u} representation characterized by a combination of two line nodes at the tropics and point nodes at the poles.

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Understanding unconventional superconductivity, in which electron pairs are formed without a phonon, has been a challenge. Part of the problem in uncovering the mechanism is that little is known about the pairing symmetry. The heavy-fermion superconductor UPt_3 is one of the examples whose pairing symmetry is as yet to be clarified. The most intriguing feature of this material is the existence of a multiple phase diagram; UPt_3 undergoes a double superconducting transition at the upper critical temperature $T_c^+ \sim 540$ mK into the A phase and at the lower critical temperature $T_c^- \sim 490$ mK into the B phase [1]. In addition, the third (C) phase is stabilized at low temperatures under high magnetic fields [2,3]. A crucial role of a weak antiferromagnetic order below $T_N \sim 5$ K for the phase multiplicity is indicated by the pressure studies [4]. Power law dependence of the thermodynamic and transport quantities reveals the presence of nodes in the superconducting gap [5–9]. Moreover, a possibility of an odd-parity pairing is suggested from the nuclear magnetic resonance studies of the Knight shift [10] and is supported theoretically [11] by eliminating the singlet even parity scenario.

Extensive theoretical effort has been devoted to explaining these disparate experimental results [11–13]. Among them, the E_{2u} scenario with a line node in the basal plane and point nodes along the c axis has been regarded as one of the promising candidates [11,14]. Several experimental results, such as the anisotropy of the thermal conductivity [15] and the ultrasonic attenuation [16] as well as the recent small-angle neutron scattering [17] and the Josephson tunnel junction [18], have been claimed to be compatible with this model. On the other hand, there exist some controversies in explaining d vector with two spin directions in the B phase [10] and a tetracritical point in the superconducting phase diagram [2]. In addition, an existence of the spontaneous internal field due to the broken time-reversal symmetry is controversial [19,20]. Moreover, to date no experiments directly probing the nodal directions associated with the E_{2u} model have been performed. The pairing symmetry of UPt_3, therefore, remains unclear.

One of the most conclusive ways to identify the pairing symmetry is to elucidate the gap structure by the thermal conductivity measurements with rotating magnetic fields relative to the crystal axes deep inside the superconducting state. This technique has been successful in probing the nodal gap structure of several unconventional superconductors by virtue of its directional nature and sensitivity to the delocalized quasiparticles [21]. In this Letter, we present the angular dependence of the thermal conductivity of UPt_3 revealing the spontaneous rotation symmetry lowering, namely, the unusual gap structure with a lower rotational symmetry than the crystal structure.

High quality single crystal of UPt_3 with the high residual resistivity ratio of 800 was grown by the Czochralski pulling method in a tetra-arc furnace [22]. We measured the thermal conductivity along the hexagonal c axis (heat current $q \parallel c$). The sample has a bar shape with a length of 3 mm along the c axis. A cross section is a parallelogram shape with edge lengths of 0.4×0.42 mm^2. The length of 0.4 mm is parallel to the a axis, and a longer (shorter) diagonal points around the azimuthal angle $\phi \sim -30(+60)^\circ$ measured from the a axis, respectively. We used the RuO_2 thermometers mounted within the ac plane.

To apply the magnetic fields with high accuracy relative to the crystal axes, we used a system with two superconducting magnets generating the fields in two mutually orthogonal directions. The magnets are installed in a Dewar seating on a mechanical rotating stage, enabling the continuous rotation of the magnetic fields.

First, we begin with demonstrating that the thermal conductivity (κ) well probes the superconducting quasiparticle (QP) structures from its temperature (T) and magnetic field (H) dependences. From now on, the hexagonal $[\bar{1}2\bar{1}0]$, $[\bar{1}010]$, and [0001] axes are denoted as the a, b, and c axes, respectively. The inset of Fig. 1 shows the T dependence of $\kappa(T)/T$ under zero field and 3 T along the b axis. With decreasing T, the zero-field $\kappa(T)/T$ shows a steep increase up to $\sim$0.3 K without apparent anomalies at T_c^+ and T_c^-. On further cooling, $\kappa(T)/T$ considerably decreases due to a reduction of the QP densities, and takes an extremely small value at the lowest $T \sim T_c^+/20$, consistent with the previous measurements [8]. In the normal state (3 T), $\kappa(T)/T$ appears to continuously increase down to the lowest T. The dashed line denotes $\kappa(T)/T$ obtained from the normal-state resistivity $\rho(T)$ using the Wiedemann-Franz law, $\kappa(T)/T = L_0/\rho(T)$ (L_0: the Lorentz number). Importantly, we confirm that $\kappa(T)/T$ is close to $L_0/\rho(T)$ at low temperature $T < 100$ mK, indicating the dominant electronic contribution in the heat transport. In this T range, the H dependence of the thermal conductivity $\kappa(H)/T$ at 55 mK shows a remarkable H-linear dependence at low fields for both c and b directions (the main panel of Fig. 1) in contradiction to the field-insensitive behavior of fully gapped superconductors except in the vicinity of H_{c2} [23]. The pronounced increase

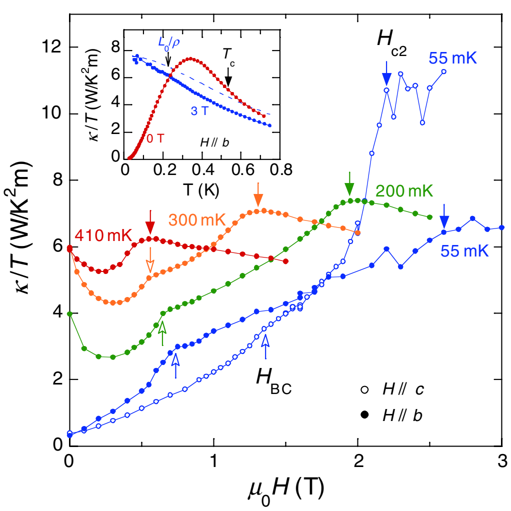

FIG. 1 (color online). Magnetic field dependence of the thermal conductivity $\kappa(H)/T$ along the c and b axes at various temperatures. The open and closed arrows represent the $B \to C$ transitions H_{BC} and the upper critical fields H_{c2}, respectively. Inset: temperature dependence of $\kappa(T)/T$ under zero field and at 3 T for $H \parallel b$. The dashed line shows $\kappa/T = L_0/\rho$ (L_0: the Lorentz number) obtained from the normal-state resistivity ρ at 3 T using the Wiedemann-Franz law.

of $\kappa(H)/T$ at low fields indicates that the heat conduction is dominated by the quasiparticle density of states (DOS) in the T region of $T \sim 50$ mK.

In addition, we find distinct anomalies associated with a transition from the B to C phase at H_{BC} (open arrows). The fact that the BC transition manifests by a sharp change of the slope implies a suppression of one of the degenerate order parameter components in the B phase. This behavior can be more clearly resolved for the b axis. The determined H_{BC} together with H_{c2} denoted by the solid arrows are summarized in Fig. 3(d) for $H \parallel b$. We also note that a striking anisotropy is found in $\kappa(H)/T$ at 55 mK near H_{c2}: κ/T for $H \parallel c$ shows a rapid increase just below H_{c2}, while the one for $H \parallel b$ linearly increases up to H_{c2}, as similarly observed in Sr_2RuO_4 [24]. A search of the relevance of this behavior to the odd-parity superconductivity is a fascinating issue to be addressed.

Next, to shed light on the nodal topology in the superconducting phases, we concentrate on the angular dependence of κ. The most significant effect on the thermal transport for nodal superconductors in the mixed state comes from the Doppler shift of the QP energy spectrum, $E(\boldsymbol{p}) \to E(\boldsymbol{p}) - \boldsymbol{v}_s \cdot \boldsymbol{p}$, in the circulating supercurrent flow $\boldsymbol{v}_s$. This effect becomes important at such positions where the gap becomes smaller than the Doppler shift term ($\Delta < \boldsymbol{v}_s \cdot \boldsymbol{p}$). The maximal magnitude of the Doppler shift strongly depends on the angle between the node direction and H, giving rise to the oscillation of the DOS. Consequently, κ attains the maximum (minimum) value when H is directed to the antinodal (nodal) directions [25,26]. Figure 2 shows $\kappa(\phi)$ normalized by the normal-state value κ_n as a function of the azimuthal angle ϕ at 50 mK ($\sim T_c^+/10$) at $|\mu_0 H| =$ (a) 3.0, (b) 1.0, and (c) 0.5 T, respectively. The experimental error increases with increasing the field because the very large thermal conductivity of UPt_3 under high field at low temperature makes generation of the thermal gradient difficult. The data are taken in rotating H after field cooling at $\phi = -70°$, and κ_n is measured at 50 mK above H_{c2} for $H \parallel b$. In the normal state (3.0 T) and the B phase (0.5 T), we find no ϕ dependence within experimental error.

By contrast, what is remarkable is that $\kappa(\phi)$ exhibits a distinct twofold oscillation with a minimum at $\phi = 0°$ in the C phase (1.0 T). The open circles are obtained under field cooling condition at each angle. The data obtained by different procedures of field cooling coincide well with each other, indicating a negligibly small effect of the vortex pinning, contrary to the previous experiments [17,27]. Strikingly, since the twofold symmetry is lower than the hexagonal crystal structure, the in-plane anisotropy of the Fermi surface [28] and H_{c2} [29] is immediately ruled out as the origin. As shown by the solid lines, $\kappa(\phi)$ can be decomposed into two terms: $\kappa(\phi) = \kappa_0 + \kappa_{2\phi}$, where κ_0 is a ϕ-independent term and $\kappa_{2\phi} = C_{2\phi} \cos 2\phi$ is a twofold component. Figure 2(e) shows the amplitude of the twofold component $|C_{2\phi}/\kappa_n|$ as a function of H/H_{c2},

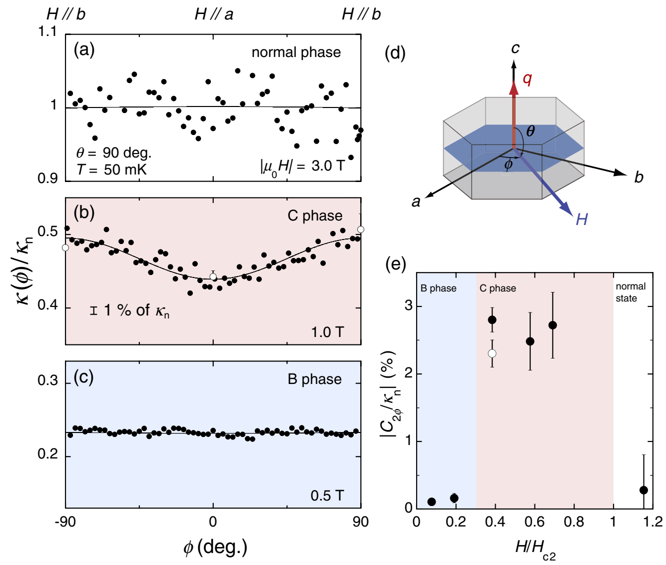

FIG. 2 (color online). Angular variation of the thermal conductivity $\kappa(\phi)$ normalized by κ_n at 50 mK as a function of the azimuthal angle ϕ for $|\mu_0 H| =$ (a) 3.0, (b) 1.0, and (c) 0.5 T, respectively. $\kappa(\phi)$ is measured with rotating H within the ab plane (the polar angle $\theta = 90°$) as schematically shown in (d), where ϕ and θ are measured from the a and c axes, respectively, and q is injected along the c axis. The solid lines show the twofold component in $\kappa(\phi)/\kappa_n$. The open circles represent $\kappa(\phi)/\kappa_n$ at 1 T obtained under the field cooling condition at every angle. (e) Field variation of the twofold amplitude $|C_{2\phi}/\kappa_n|$ at 50 mK at $\theta = 90°$ (solid circles) and 63° (open circle), respectively.

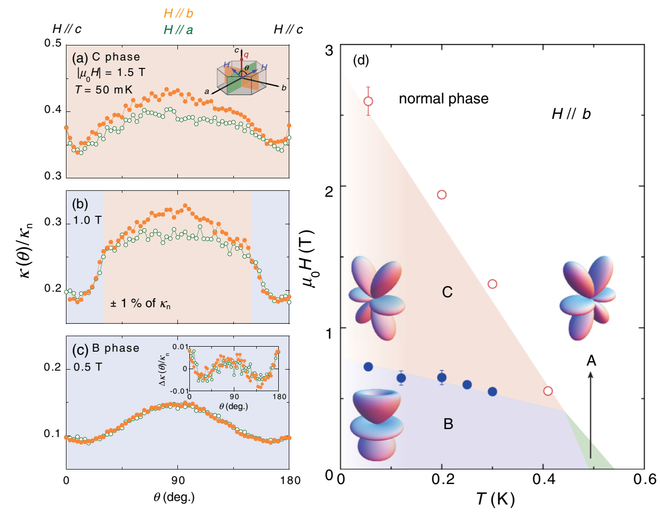

FIG. 3 (color online). Angular variation of the thermal conductivity $\kappa(\theta)$ normalized by κ_n at 50 mK as a function of the polar angle θ for $|\mu_0 H| =$ (a) 1.5, (b) 1.0, and (c) 0.5 T, respectively. The $\kappa(\theta)/\kappa_n$ curves measured by rotating H [inset of (a)] within the ac plane (open circles) and the bc plane (closed circles) are simultaneously plotted. Inset of (c): $\Delta\kappa(\theta)/\kappa_n \equiv (\kappa(\theta) - \kappa_0 - \kappa_{2\theta})/\kappa_n$ vs θ plot at 50 mK at 0.5 T, where κ_0 is a θ-independent term and $\kappa_{2\theta} = C_{2\theta}\cos 2\theta$ is a twofold component. (d) The phase diagram of UPt_3 with the three superconducting phases, labeled A, B, and C, for $H \parallel b$. The open and closed circles represent H_{BC} and H_{c2}, respectively, deduced from the present measurements. The schematic shapes of the gap symmetries for each phase are shown.

where $H_{c2} = 2.6$ T for $H \parallel b$. It can be clearly seen that $|C_{2\phi}/\kappa_n|$ suddenly appears to be finite $\sim 3\%$ in the C phase, implying a change of the gap symmetries across the BC transition that is of second order. We note that $|C_{2\phi}/\kappa_n|$ obtained by rotating H conically around the c axis at fixed $\theta = 63°$ is same order of magnitude with the values at $\theta = 90°$ as denoted by an open circle in Fig. 2(e).

To further elucidate the gap symmetry, we present the polar angle (θ) dependence of κ in Fig. 3, showing $\kappa(\theta)/\kappa_n$ measured by rotating H within the ac plane (open circles) and the bc plane (closed circles) at 50 mK at $|\mu_0 H| =$ (a) 1.5, (b) 1.0, and (c) 0.5 T. Here, κ_n is measured at 50 mK above H_{c2} for $H \parallel c$. The dominant twofold oscillation is found in all the fields with maxima at $\theta = 90°$ as previously reported [30], which could be attributed to, such as the Fermi surface and/or the gap topology or the difference in transport with H parallel to and normal to the heat current q. Regardless of the origin, the fact that $\kappa(\theta)/\kappa_n$ is maximized at $\theta = 90°$ excludes an artificial origin of the in-plane twofold oscillation in the C phase due to a misalignment of H relative to q. We thus conclude that the in-plane twofold symmetry in the C phase is a consequence of the node.

In the B phase (0.5 T), the two different scanning procedures within the ac and bc planes well converge with each other, consistent with the ϕ independence of κ. In addition, we find extra two minima around $\theta \sim 20°$ and 160°. By plotting $\Delta\kappa(\theta)/\kappa_n \equiv (\kappa(\theta) - \kappa_0 - \kappa_{2\theta})/\kappa_n$ vs θ after the subtraction of a θ-independent term κ_0 and a twofold component $\kappa_{2\theta} = C_{2\theta}\cos 2\theta$ originating from the out-of-plane twofold anisotropy of the Fermi surface and/or magnetothermal resistance, the minima become clearly visible around 35° and 145° [Fig. 3(c), inset]. This double-minimum structure is also found in the C phase [Fig. 3(a)]. We infer that these minima are derived from the two horizontal line nodes at the tropics as discussed below. In contrast to the B phase, the two scanning results do not coincide in the C phase [Fig. 3(a)]; the difference is diminished at the poles and maximized at $\theta = 90°$, being consistent with the in-plane twofold symmetry. Moreover, a significant appearance of the twofold symmetry across the BC transition can be seen at 1.0 T [Fig. 3(b)], in which one experiences the BC (CB) transition twice by varying θ because of the anisotropy of H_{BC}. Indeed, the transitions occur at $\theta \sim 30°$ and 150° taking distinct kinks. Remarkably, the difference between the two scanning procedures becomes finite upon entering the C phase, providing the compelling evidence for the twofold symmetry of the gap structure in the C phase. Moreover, the fact that $|C_{2\phi}/\kappa_n|$ takes same order of the magnitude at $\theta = 90°$ and 63° is in favor of a line node along the a axis rather than the point nodes in the basal plane. Notably, although a mechanism which fixes domains is a puzzle, the in-plane twofold symmetry of $\kappa(\phi)$ indicates a single superconducting domain.

We discuss the order parameter symmetry of UPt_3 within the triplet category. The present experiments indicate (i) the line node along the a axis in the C phase, (ii) the absence of in-plane gap anisotropy in the B phase, and (iii) the two horizontal line nodes at the tropics in both B and C phases. Taking into account all these results and the d-vector configurations assigned by the Knight shift [10], the order parameter is determined with a form of $(k_a\hat{b} + k_b\hat{c})(5k_c^2 - 1)$ for the B phase, where $\hat{b}$ and $\hat{c}$ are unit vectors of the hexagonal axes representing the directions of d vectors. This state belongs to two-dimensional E_{1u} representation with the f-wave character, the so-called planar state in triplet pairing in the D_{6h} hexagonal symmetry, and to degenerate E_u state for the recent claimed D_{3d} trigonal symmetry [31,32]. The gap structure consists of the two horizontal line nodes at the tropics ($k_c = \pm 1/\sqrt{5}$, $\theta \sim 63°$ and $117°$) and the point nodes at the poles ($k_a = k_b = 0$). Note that although the locations of the horizontal line nodes estimated by assuming a spherical Fermi surface do not agree with the observation ($\theta \sim 35°$ and $145°$), they could be changed by considering the realistic Fermi surface [28].

By lifting the double degeneracy, the order parameter for the C phase is given by $k_b\hat{c}(5k_c^2 - 1)$ for $H \parallel ab$ and $k_b\hat{a}(5k_c^2 - 1)$ for $H \parallel c$, respectively. In the same manner, the $k_a\hat{b}(5k_c^2 - 1)$ state is readily assigned for the A phase [33]. The schematic shapes of the gap symmetries in the three phases are shown in Fig. 3(d). We emphasize that this state is compatible not only with the hybrid gap state indicated by the several experiments [15,16], in the sense that the line and point nodes simultaneously exist, but also with some experimental results for which the E_{2u} model [11] has failed to describe, i.e., the absence of the internal magnetic field [19,34], d vector with two spin directions for the B phase [10], and the tetracritical point in the phase diagram [2]. On the other hand, a theoretical verification of the temperature dependence of the thermal conductivity (the inset of Fig. 1) and the transverse ultrasound [16] in the framework of our proposed representation remains to be resolved as future work.

To further strengthen our identification, in particular, on the existence of the horizontal line nodes on the tropics, we calculate the angle-resolved DOS by solving the Eilenberger equation [35] for several possible gap functions. Since κ is dominated by the DOS at $T \sim 50$ mK as mentioned above, we compare here putative three gap functions in the C phase relative to the data in Fig. 4 where $\kappa(\theta)/\kappa_n$ measured at 50 mK and the DOS differences along the vertical nodal and antinodal θ scannings are depicted. The double peak structure characteristic in E_{2u} and E_{1g} whose origin comes from the horizontal node on the equator is not supported by the data that are consistent with the present E_{1u} with the horizontal nodes on the tropics. In view of the Doppler shift idea mentioned above the QPs in the horizontal node on the equator contribute more when the field direction is away from $\theta = 90°$.

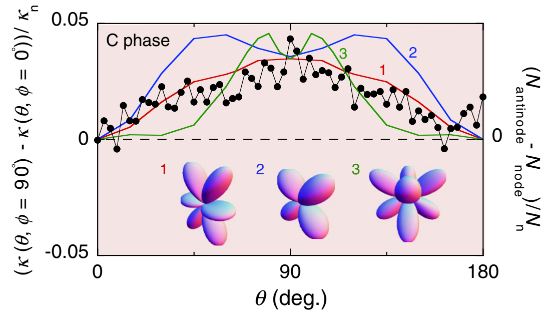

FIG. 4 (color online). θ dependence of the thermal conductivity difference obtained by subtracting $\kappa(\theta, \phi = 0°)$ from $\kappa(\theta, \phi = 90°)$ denoted by the open and closed circles, respectively, in Fig. 3(a). Right axis: the density of states difference normalized at $\theta = 90°$ (arbitrary scale) along the vertical nodal and antinodal scannings for three possible gap functions in the C phase: 1. The present $E_{1u}(k_b(5k_c^2 - 1))$, 2. $E_{1g}(k_bk_c)$, 3. $E_{2u}(k_ak_bk_c)$. Those gap structures are sketched in the inset.

In summary, we find striking twofold oscillations in angle-resolved thermal conductivity measurements at low temperatures in a strongly correlated heavy-fermion superconductor UPt_3. This spontaneous symmetry lowering, which is the lowest possible rotational symmetry breaking in hexagonal crystal fortuitously and effectively narrows down the possible symmetry classes and leads us to identify the pairing symmetry for each phase in the multiple phase diagram. We conclude that the realized pairing function is E_{1u} with the f-wave character, i.e., the so-called planar state in the triplet pairing. This state is analogous to the B phase in superfluid ^{3}He, and obviously bears the Majorana zero mode at a surface [36,37]. Thus it is worth exploring further to understand this interesting material as a new platform for topological physics.

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[32] The previously proposed E_{1u} with the p-wave character such as $k_a + ik_b$ is simply inappropriate because it yields no line node in the B phase.

[33] There remains a possibility of sign reversal of the in-plane twofold amplitude taking place at even lower temperature ($T < 50$ mK), leading to a change of the nodal direction from along the a axis to the b axis for the C phase.

[34] If the time-reversal symmetry is broken, the gap function for the B phase should be $(k_a\hat{b} + ik_b\hat{c})(5k_c^2 - 1)$ characterized by four vertical line nodes. This gap structure conflicts with our experimental results.

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