Searching for the in-plane anisotropy of the specific heat of UPt$_3$ in rotating fields

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Abstract. In order to identify the gap structure of a spin-triplet superconductor UPt$_3$, we performed field-angle-resolved specific heat ($C_\phi$) measurements in a magnetic field rotated around the [0001] axis. Although various theories predict fourfold symmetry of the gap in the superconducting C phase, we observed no angular variation in $C_\phi$ in a temperature range down to 88 mK ($T_c^0$: 17 T$_c$). This absence of the variation in $C_\phi$ implies that the field-angle-dependent quasiparticle excitations, reflecting the gap anisotropy, are averaged out in the bulk of UPt$_3$.

1. Introduction
The heavy-fermion superconductor UPt$_3$ has attracted great interest because of its unusual superconducting (SC) properties such as the multiple SC phases (Fig. 1(a)) [1]. NMR Knight shift measurements suggested that the spin state of the Cooper pair is triplet in UPt$_3$ [2] and various experiments indicated the presence of line nodes in the SC gap [1]. On theoretical grounds, various multi-component order parameters have been proposed to explain the multiple SC phases. [1, 3, 4]. For instance, in the $E_{2u}$ ($E_{1g}$) scenario with the spin-triplet (spin-singlet) pairing, the three distinct SC phases can be explained by introducing a symmetry-breaking field which lifts the degeneracy of the SC order parameters $k_z(k_x^2 - k_y^2)$ and $2k_xk_yk_z$ ($k_xk_y$ and $k_z$). Accordingly, for the the $E_{2u}$ ($E_{1g}$) scenario, the gap function has fourfold (twofold) symmetry in the A and C phases with vertical line nodes, which are absent in the B phase whose gap function becomes $|k_z|(k_x^2 + k_y^2)$ with axial symmetry. Schematic gap structures in the B and C phases for the $E_{2u}$ model are depicted in Figs. 1(b) and 1(c), respectively. Nevertheless, the origin of the symmetry-breaking field is still controversial. It could possibly be related to short-range antiferromagnetic fluctuations developing below 5 K [5] and/or distortions in the crystal structure [6].

On experimental grounds, however, the SC gap structures of UPt$_3$ have not been well resolved yet. A recent junction experiment has reported that vertical nodes exist in the A phase at 45 degrees from the $a$ axis in the $ab$ plane whereas they are absent in the B phase [7]. The gap structure in the C phase has not yet been examined by the directional probe. In the present study, we have performed field-angle-resolved specific heat measurements on UPt$_3$ in order to explore the gap structure in the low-temperature B and C phases. This technique is a powerful tool to determine nodal directions of
bulk superconductors [8], and has recently been applied to CeCoIn$_5$ to conclude the $d_{x^2-y^2}$ symmetry; in a magnetic field $H$ rotated around the [0001] axis, a fourfold oscillation of the specific heat has been observed with maxima occurring along the [100] directions [9] as predicted in the low-$T$ and low-$H$ “Doppler shift” regime [10].

2. Experimental

The single crystal of UPt$_3$ used in the present study (54.2 mg weight) was grown by the Czochralski pulling method. The sample was shaped into a disk with the flat surfaces parallel to the (0001) plane, as shown in Fig. 1(d). The specific heat was measured by a standard quasi-adiabatic heat-pulse technique as well as a relaxation method in a dilution refrigerator (Oxford Kelvinox AST Minisorb). Magnetic fields were generated in the $x$-$z$ plane using a vector magnet composed of a vertical solenoid (3 T) and horizontal split-pair coils (5 T). The vector magnetic field was then rotated around the $z$ axis of the specimen by rotating the refrigerator with a stepper motor mounted at the top of a magnet Dewar. For each field angle, the specific-heat value was determined by the average of ten successive measurements. In all the data presented, a nuclear Schottky contribution ($C_n$) by the Zeeman splitting of $^{195}$Pt nuclei ($I = 1/2$) was subtracted. It was confirmed that the addenda contribution is always less than 4% of the sample specific heat and is invariant in rotating fields.

3. Results and Discussion

Figure 2 compares the temperature dependence of $C/T$ measured by the quasi-adiabatic (open circles) and the relaxation (dots) methods. The $C/T$ data measured by the two methods well coincide each other, and are also in agreement with the results of the previous reports [11]. These facts guarantee the high quality of our sample ($T_c = 0.52$ K) and ensure the reliability of our experimental setups. As can be seen in the figure, a rapid increase of $C/T$ was observed below about 0.1 K even in zero field. This Schottky-like behavior was previously reported to originate from an antiferromagnetic long-range ordering at 20 mK [12, 13].

Figure 3 shows the field-strength dependence of $C/T$ at 88 mK for $H \parallel ab$. As shown in the inset of Fig. 3, $C/T$ is linear to the square root of $H$ in the wide field range from 0 T to nearly $H_{c2}$. This $\sqrt{H}$ behavior is much clearer than the previously reported one obtained at 150 mK [14], and strongly suggests that Doppler-shifted quasi-particle excitations around the line nodes are predominant in the field variation of $C/T$ at 88 mK.
In Fig. 4, we present the results of the specific heat measurements in magnetic fields rotated around the [0001] axis. The field direction is defined by the angles $\theta$ and $\phi$, where $\theta$ is the polar angle of $H$ with respect to the crystalline [0001] axis and $\phi$ denotes the azimuthal angle within the (0001) plane measured from the crystalline [1210] axis, as depicted in Fig. 1(e). We investigated the specific heat as a function of $\phi$, $C_\phi$, under various conditions ($T$, $H$, and $\theta$) as summarized in Fig. 1(a) by symbols. It is very important to examine the dependence of $C_\phi$ because the gap functions are considered to possess a horizontal line node. If the order parameter of UPt$_3$ in the C phase is $k_xk_yk_z$ or $k_xk_z$, the anisotropy of $C_\phi$ is expected to become largest at $\theta \approx 60^\circ$ rather than $\theta = 90^\circ$ because the magnetic field is always in the nodal direction for $\theta = 90^\circ$ in the presence of the horizontal line node. Therefore, we investigated $C_\phi$ with $\theta = 60^\circ$ (conical rotation of $H$) as well as $\theta = 90^\circ$. Nevertheless, in a wide temperature range between 88 and 200 mK, no angular variation was detected in $C_\phi$ for both $\theta = 60^\circ$ and $\theta = 90^\circ$ within the limit of our experimental resolution (0.1% of the electronic specific heat) in any field strengths below $H_{c2}$. This is in striking contrast to the observation of a fourfold oscillation of $C_\phi$ with an amplitude of larger than 1% in a $d_{x^2-y^2}$-wave superconductor CeCoIn$_5$ [9].

To evaluate the $\theta$ dependence of the fourfold oscillation amplitude, we compared $C(\phi = 0)$ with $C(\phi = 45)$ at various $\theta$ and $H$ at 88 mK. The difference $\Delta C(\theta) = C(\theta, \phi = 0) - C(\theta, \phi = 45)$ normalized by $C(\theta = 90, \phi = 45)$ is plotted in Fig. 5. For all field strengths studied, $\Delta C(\theta)$ stays nearly zero, indicating that the fourfold oscillation in $C_\phi$ is absent at any $\theta$.

Considering the results of a number of studies on the gap structure of UPt$_3$, it is not reasonable to conclude from the present results that the gap of UPt$_3$ is isotropic around the [0001] axis in the C phase as well as the B phase. As a possible origin of the absence of the anisotropy in $C_\phi$, we suggest that the superconducting order parameter forms a domain structure. The order parameters with the fourfold or twofold symmetry around the [0001] axis would have three domain states in the hexagonal crystal structure. If these domains are equally populated, then the in-plane anisotropy of the specific heat might be averaged out in the bulk. It is also possible that the lowest measurement temperature of 88 mK is not sufficiently low enough to detect the gap anisotropy through $C_\phi$. 

![Figure 2](image_url)  
*Figure 2. Temperature dependence of the nuclear-subtracted $C/T$ at (a) 0 T and (b) 1 T for $H \parallel ab$. Open (closed) symbols are the data measured using the quasi-adiabatic (relaxation) method.*

![Figure 3](image_url)  
*Figure 3. Field dependence of the nuclear-subtracted $C/T$ at 88 mK for $H \parallel ab$. Inset shows the $\sqrt{H}$-dependence. The dashed lines represent fits of the data using the function $a \sqrt{H} + b$.***
4. Conclusion
We measured the specific heat of UPt$_3$ in rotating magnetic fields around the [0001] axis. It was found that the specific heat is invariant in the rotating fields of any strength below $H_{c2}$ at temperatures between 88 and 200 mK. This result indicates that the change in the density of states as a function of in-plane field angle is tiny in the bulk. Although the anisotropy of the gap structure has not been detected, we proposed a possibility that the gap anisotropy is smeared out in the bulk by the presence of domain structures of the symmetry-breaking fields or by thermal excitations of quasi-particles in the present measurement.

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