Superconducting Gap Structure of Spin-Triplet Superconductor Sr_2RuO_4 Studied by Thermal Conductivity

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To clarify the superconducting gap structure of the spin-triplet superconductor Sr_2RuO_4, the in-plane thermal conductivity has been measured as a function of relative orientations of the thermal flow, the crystal axes, and a magnetic field rotating within the 2D RuO_2 planes. The in-plane variation of the thermal conductivity is incompatible with any model with line nodes vertical to the 2D planes and indicates the existence of horizontal nodes. These results place strong constraints on models that attempt to explain the mechanism of the triplet superconductivity.

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Since its discovery in 1994 [1], the superconducting properties of Sr_2RuO_4 has been attracting considerable interest. A remarkable feature which characterizes this system is the spin-triplet pairing state with **d**-vector perpendicular to the conducting plane, which has been confirmed by ^{17}O NMR Knight shift measurements [2]. Moreover, muon spin rotation (μSR) experiments suggest that the time reversal symmetry is broken in the superconducting state [3]. Up to now, the spin-triplet pairing state is identified only in superfluid ^{3}He, heavy fermion UPt_3 [4], and organic $(TMTSF)_2PF_6$ [5], though it is probably also realized in the recently discovered UGe_2 [6]. At an early stage, the gap symmetry of Sr_2RuO_4 was discussed in analogy with ^{3}He in which Cooper pairs are formed by the ferromagnetic spin fluctuation. Then the pairing state with the isotropic gap in the plane, $\mathbf{d}(\mathbf{k}) = \Delta_0\hat{\mathbf{z}}(k_x + ik_y)$, has been proposed as being likely to be realized [7,8].

However, recent experiments have revealed that the situation is not so simple. Neutron inelastic scattering experiments have shown the existence of strong incommensurate antiferromagnetic correlations and no sizable ferromagnetic spin fluctuation [9]. Furthermore, the specific heat C_p and NMR relaxation rate T_1^{-1} on very high quality compounds exhibit the power-law dependence of $C_p \propto T^2$ [10] and $T_1^{-1} \propto T^3$ [11] at low temperatures, indicating the presence of nodal lines in the superconducting gap. These results have motivated theorists to propose new models [12–15]. Most of them predict the line nodes which are vertical to the 2D planes. However, the detailed structure of the gap function, especially the direction of the nodes, is an unresolved issue. Since the superconducting gap function is closely related to the pairing interaction, its clarification is crucial for understanding the pairing mechanism.

A powerful tool for probing the anisotropic gap structure is the thermal conductivity κ because it is a *directional* probe, sensitive to the orientation relative to the thermal flow, the magnetic field, and nodal directions of the order parameter [16–19]. In fact, a clear fourfold modulation of κ with an in-plane magnetic field which reflects the angular position of nodes of $d_{x^2-y^2}$ symmetry has been observed in $YBa_2Cu_3O_{7-\delta}$, demonstrating that the thermal conductivity can be a relevant probe of the superconducting gap structure [20,21]. Although previous attempts have been made to measure the thermal conductivity in Sr_2RuO_4, the experimental resolution was not good enough to identify the nodal directions [22]. In this Letter, we performed a high precision measurement of the in-plane thermal conductivity as a function of angle between the thermal current **q** and the magnetic field **H** rotating within the RuO_2 plane.

Single crystals were grown by the floating-zone method. The thermal conductivity was measured by a steady state method. In the present measurements, it is very important to rotate **H** within the RuO_2 planes with high accuracy because a slight field misalignment produces a large effect on κ due to the large anisotropy. For this purpose, we constructed a system with two superconducting magnets generating **H** in two mutually orthogonal directions and a ^{3}He cryostat equipped on a mechanical rotating stage with a minimum step of $1/500°$ at the top of the Dewar. Computer controlling two magnets and the rotating stage, we were able to rotate **H** continuously within the RuO_2 planes with a misalignment less than 0.015° from the plane. Precise in-plane alignment of $\mathbf{H} \parallel a$ axis was achieved by measuring the in-plane anisotropy of H_{c2} [23].

The inset of Fig. 1(a) shows the T dependence of κ/T in zero field. Since the electrical resistivity is very small, which is an order of 0.1 $\mu\Omega \cdot$ cm, the electron contribution well dominates over the phonon contribution [24]. At T_c κ/T shows a kink. At low T, κ/T decreases almost linearly with decreasing T with finite residual values at $T = 0$. The residual κ decreases with increasing T_c and is very small in the crystal with highest $T_c (= 1.45$ K). These T^2 dependence and residual κ/T are consistent with the presence of the line nodes [10,11].

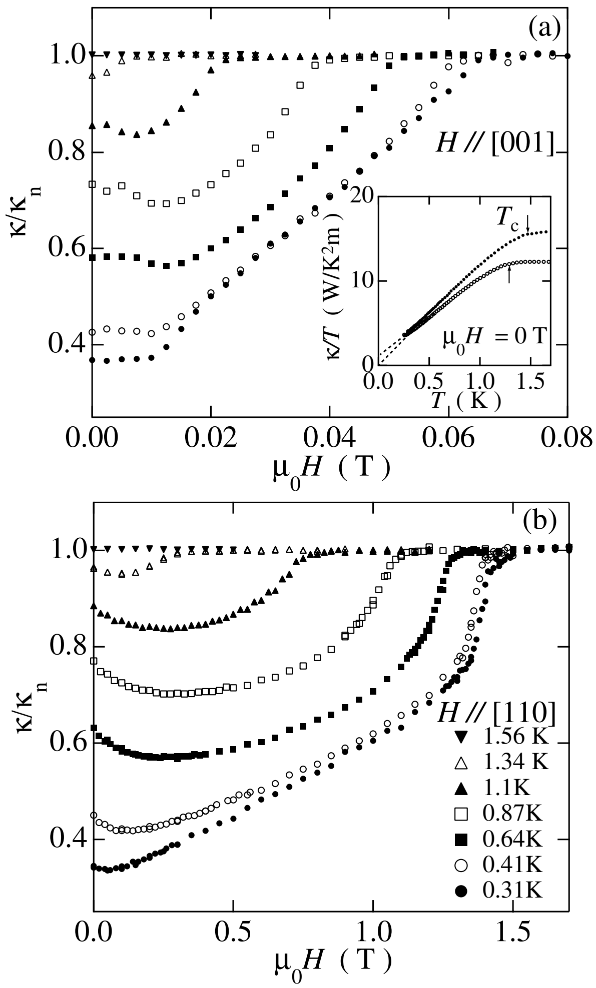

FIG. 1. Field dependence of the thermal conductivity of Sr_2RuO_4 ($T_c = 1.45$ K) in (a) perpendicular field $\mathbf{H} \perp ab$ and (b) parallel field $\mathbf{H} \parallel [110]$. The thermal current $\mathbf{q}$ is applied along the [110] direction. In perpendicular field, κ is H independent below the lower critical field. Inset: T dependence of κ/T in zero field for two crystals with different T_c ($T_c = 1.45$ and 1.32 K).

Figures 1(a) and 1(b) show the H dependence of κ for the sample with $T_c = 1.45$ K in perpendicular ($\mathbf{H} \perp ab$ plane) and parallel ($\mathbf{H} \parallel ab$ plane) fields, respectively. In both orientations, κ increases with H after the initial decrease at low fields. The consequent minimum is much less pronounced at lower temperatures. At low T, κ increases linearly with H. We note that the H-linear dependence of κ is observed only in the very clean crystals with $T_c >$ 1.3 K, and κ increases with an upward curvature in samples with lower T_c. In parallel field, κ rises very rapidly as H approaches H_{c2} and attains its normal value with a large slope ($d\kappa/dH$), while κ in the perpendicular field remains linear in H up to H_{c2}. The understanding of the heat transport for superconductors with nodes has largely progressed during the past few years [17,25,26]. There, in contrast to the classical superconductors, the heat transport is dominated by contributions from delocalized quasiparticle states rather than the bound state associated with vortex cores. The most remarkable effect on the thermal transport is the Doppler shift of the quasiparticle energy spectrum [$\varepsilon(\mathbf{p}) \rightarrow \varepsilon(\mathbf{p}) - \mathbf{v}_s \cdot \mathbf{p}$] in the circulating supercurrent flow $\mathbf{v}_s$ [27]. This effect becomes important at such positions where the local energy gap becomes smaller than the Doppler shift term ($\Delta < \mathbf{v}_s \cdot \mathbf{p}$), which can be realized in the case of superconductors with nodes. In the presence of line nodes where the density of states (DOS) of electrons $N(\varepsilon)$ has a linear energy dependence [$N(\varepsilon) \propto \varepsilon$], $N(H)$ increases in proportion to $\sqrt{H}$. While the Doppler shift enhances the DOS [27], it also leads to a suppression of both the impurity scattering time and Andreev scattering time off the vortices [21,26]. This suppression can exceed the parallel rise in $N(\varepsilon)$ at high temperature and low field, which results in the nonmonotonic field dependence of $\kappa(H)$.

It has been shown that in the superconductors with line nodes, κ increases in proportion to H in the "superclean regime" where the condition, $\frac{\Gamma}{\Delta} \ll \frac{H}{H_{c2}}$ is satisfied. Here Γ is the pair breaking parameter estimated from the Abrikosov-Gorkov equation $\Psi(1/2 + \Gamma/2\pi T_c) - \Psi(1/2) = \ln(T_{c0}/T_c)$, where Ψ is a digamma function and T_{c0} is the transition temperature in the absence of the pair breaking. Assuming $T_{c0} = 1.50$ K and $\Delta = 1.76T_c$, Γ/Δ is estimated to be 0.025 (0.067) for $T_c = 1.45$ K ($T_c = 1.37$ K), showing that our field range is well inside the superclean regime, except at very low fields smaller than 400 Oe (1000 Oe). Thus the H-linear dependence of $\kappa(H)$ observed in very clean crystals is consistent with the κ of superconductors with line nodes. The steep increase of κ in the vicinity of H_{c2} in the parallel field is also observed in pure Nb [28]. When the vortices are close enough near H_{c2}, tunneling of the quasiparticle excitations from core-to-core becomes possible, which leads to a large enhancement of the quasiparticle mean-free path and κ. The absence of a steep increase in the perpendicular field may be related to the difference of the vortex core structure. We note that a similar behavior is observed in UPt_3, in which the steep increase of κ is present in $\mathbf{H} \parallel c$ while is absent in $\mathbf{H} \parallel b$ [29].

We now move on to the angular variation of the thermal conductivity in the parallel field. Figures 2(a) and 2(b) depict $\kappa(H, \theta)$ as a function of $\theta = (\mathbf{q}, \mathbf{H})$. No hysteresis of κ related to the pinning of the vortices was observed in rotating θ. In all data, $\kappa(H, \theta)$ can be decomposed into three terms with different symmetries: $\kappa(H, \theta) = \kappa_0(H) + \kappa_{2\theta}(H) + \kappa_{4\theta}(H)$, where κ_0 is θ independent; $\kappa_{2\theta}(H) = C_{2\theta}(H)\cos 2\theta$ is a term with twofold symmetry; $\kappa_{4\theta}(H) = C_{4\theta}(H)\cos 4\theta$ with fourfold symmetry with respect to the in-plane rotation. These angular variations disappeared above H_{c2}. Figures 3(a)–3(d) show $\kappa_{4\theta}/\kappa_n$ after the subtraction of the κ_0 and $\kappa_{2\theta}$ terms from κ.

The sign and magnitude of $C_{2\theta}$ and $C_{4\theta}$ provide important information on the gap structure. The term $\kappa_{2\theta}$ appears as a result of the difference of the effective DOS for quasiparticles traveling parallel to the vortex and for quasiparticles moving in the perpendicular direction. In the

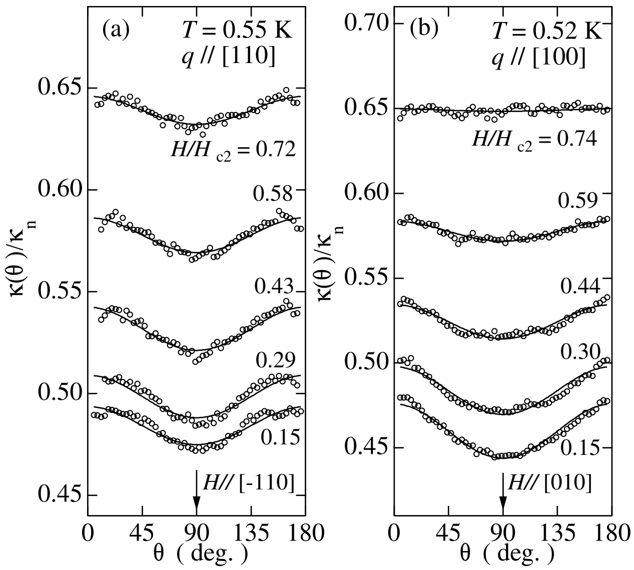

FIG. 2. (a) Angular variation of $\kappa(\theta)/\kappa_n$ for Sr_2RuO_4 ($T_c = 1.45$ K). **q** is applied to the [110] direction. (b) Same data for the sample with $T_c = 1.37$ K. **q** is applied to the [100] direction. The solid lines show the twofold component in $\kappa(\theta)/\kappa_n$.

particle lifetime from the Andreev scattering off the vortex lattice, which has the same symmetry as the gap function [20,21,26]. This effect is important at very low fields, where κ decreases with H. In addition to the fourfold symmetry associated with vertical nodes, there is another contribution to the $\kappa_{4\theta}$ term originating from the in-plane anisotropy of H_{c2} [23], which may arise from the tetragonal band structure inherent to the Sr_2RuO_4 crystal. We will discuss this effect later.

presence of vertical nodes, the term $\kappa_{4\theta}$ appears as a result of two effects. The first effect is the DOS oscillation associated with the rotating **H** within the plane. This effect arises from the fact that DOS depends sensitively on the angle between **H** and the direction of nodes of order parameter, because the quasiparticles contribute to the DOS when their Doppler-shifted energy exceeds the local energy gap. In this case, κ attains the maximum value when **H** is directed to the antinodal directions and becomes minimum when **H** is directed along the nodal directions [18,19]. The second effect is the quasi-

The most important subject is, "Is the observed $\kappa_{4\theta}$ a consequence of the vertical line nodes"? Before analyzing the data, we list the various proposed gap functions [13]:

(1) Type I: vertical nodes at $(\pm\pi, 0)$ and $(0, \pm\pi)$ [12,15]; $\mathbf{d}(\mathbf{k}) = \Delta_0\hat{\mathbf{z}}(\sin k_x + i\sin k_y)$ and $\mathbf{d}(\mathbf{k}) = \Delta_0\hat{\mathbf{z}}k_xk_y(k_x + ik_y)$.

(2) Type II: vertical nodes at $(\pm\pi, \pm\pi)$ [15,30]; $\mathbf{d}(\mathbf{k}) = \Delta_0\hat{\mathbf{z}}(k_x^2 - k_y^2)(k_x + ik_y)$.

(3) Type III: horizontal nodes [14,15]; $\mathbf{d}(\mathbf{k}) = \Delta_0\hat{\mathbf{z}}(k_x + ik_y)(\cos ck_z + \alpha)$ with $\alpha \leq 1$ (c is the interlayer distance) and $\mathbf{d}(\mathbf{k}) = \Delta_0\hat{\mathbf{z}}k_z(k_x + ik_y)^2$.

As shown in Figs. 3(a)–3(c), $\kappa_{4\theta}$ shows minimum at $\mathbf{H} \parallel [110]$. Therefore, *this result immediately excludes the type I symmetry*, in which $\kappa_{4\theta}$ should exhibit a maximum at $\mathbf{H} \parallel [110]$. We next discuss the amplitude of $\kappa_{4\theta}$. Figure 4 depicts the H dependence of $|C_{2\theta}|$ and $|C_{4\theta}|$. In the vicinity of H_{c2} where κ increases steeply, $|C_{4\theta}|/\kappa_n$ is of the order of several percent [see Fig. 3(a)]. However, $|C_{4\theta}|/\kappa_n$ decreases rapidly and is about 0.2%–0.3% at lower field, where κ increases linearly with H [see Figs. 3(b) and 3(c)]. At very low field where κ decreases with H, no discernible fourfold oscillation is observed within the resolution of $|C_{4\theta}|/\kappa_n < 0.1\%$ [see Fig. 3(d)].

Recently, the amplitudes of $\kappa_{4\theta}$ for various symmetries with vertical nodes have been calculated at the field range

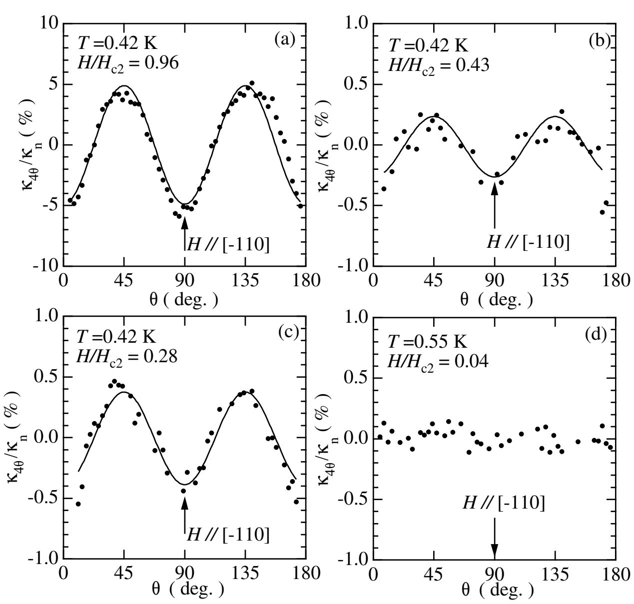

FIG. 3. (a)-(d) The fourfold symmetry $\kappa_{4\theta}/\kappa_n$ at several fields. **q** is applied to the [110] direction.

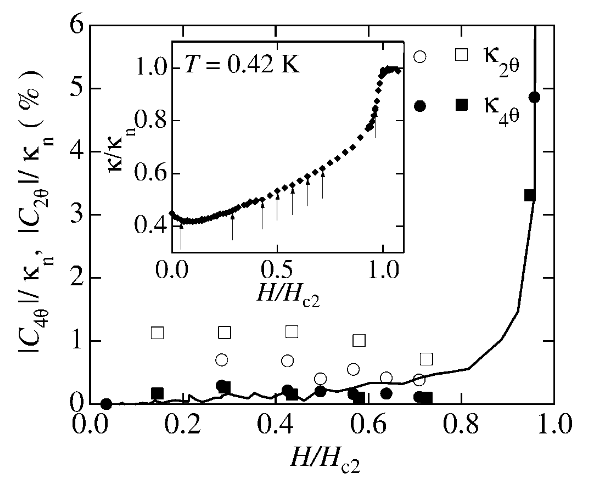

FIG. 4. The amplitude of twofold and fourfold symmetry as a function of H/H_{c2}. The filled circles and squares indicate $|C_{4\theta}|/\kappa_n$ at $T = 0.42$ and 0.55 K, respectively. The open circles and squares indicate $|C_{2\theta}|/\kappa_n$ at $T = 0.42$ and 0.55 K, respectively. The solid line represents $|C_{4\theta}|/\kappa_n$ calculated from the fourfold symmetry of H_{c2}. Inset: H dependence of κ. The arrows indicate the points at which we measured $C_{2\theta}$ and $C_{4\theta}$.

where κ obeys an H-linear dependence [15]. We will examine our result in accordance with Ref. [15]. For both type I and II symmetries, $|C_{4\theta}|/\kappa_n$ is expected to be about 6% at low field. Apparently, the observed $|C_{4\theta}|/\kappa_n \lesssim 0.3\%$ at low fields are less than 1/20 of the prediction for type I and II symmetries. Thus *it is very unlikely that the observed fourfold symmetry is an indication of vertical line nodes.* We then consider the in-plane anisotropy of H_{c2} as an origin of $\kappa_{4\theta}$. In our crystal, we find that H_{c2} is well expressed as $H_{c2}(\phi)/H_{c2}(0) = 1 + A\cos 4\phi$ with $A = -0.013$, where ϕ is the angle between **H** and the a axis. In Fig. 4, we plot $|C_{4\theta}| = |A|Hd\kappa(H)/dH$ calculated from the in-plane anisotropy of H_{c2} with no fitting parameter. The calculation reproduces the data, indicating that the fourfold symmetry of κ is indeed mainly due to the in-plane anisotropy of H_{c2}.

We next discuss $\kappa_{2\theta}$ which provides additional important information on the gap structure. According to Ref. [15], a large twofold amplitude, $|C_{2\theta}|/\kappa_n \gtrsim 25\%$ is expected for type I and II symmetries when **q** is injected parallel to the nodes. To check this, we applied **q** along [110] and [100] directions, as shown in Figs. 2(a) and 2(b). In both cases $|C_{2\theta}|/\kappa_n$ is about 1%, which is again much less than expected for the case of vertical line nodes. Thus *both twofold and fourfold symmetries of the thermal conductivity are incompatible with any model with vertical line nodes.*

We now examine the type III symmetry without $\kappa_{4\theta}$ term associated with the nodes. The magnitude of $C_{2\theta}$ provides a clue toward distinguishing between the two gap functions listed under the category of type III. According to Ref. [14], a large magnitude of $|C_{2\theta}|/\kappa_n > 30\%$ is expected for $\mathbf{d}(\mathbf{k}) = \Delta_0\hat{\mathbf{z}}k_z(k_x + ik_y)^2$. In fact, a large twofold oscillation is observed in the B phase of UPt_3 with this symmetry [16]. On the other hand, a much smaller $|C_{2\theta}|/\kappa_n \sim 8\%$ is expected at $T = 0$ for $\mathbf{d}(\mathbf{k}) = \Delta_0\hat{\mathbf{z}}(k_x + ik_y)(\cos ck_z + \alpha)$. Although the value is still several times larger (which may be due to the finite temperature effects which reduce $|C_{2\theta}|$), it is much closer to the experimental result. These results lead us to conclude that the gap symmetry which is most consistent with the in-plane variation of thermal conductivity is $\mathbf{d}(\mathbf{k}) = \Delta_0\hat{\mathbf{z}}(k_x + ik_y)(\cos ck_z + \alpha)$, in which a substantial portion of the Cooper pairs occurs between the neighboring RuO_2 planes. These results impose strong constraints on models that attempt to explain the mechanism of the triplet superconductivity.

In summary, the in-plane thermal conductivity of Sr_2RuO_4 has been measured in **H** rotating within the planes. The angular dependence is incompatible with any model with vertical line nodes and strongly indicated the presence of horizontal line nodes.

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Note added.—After completion of this paper, we became aware of the result of Tanatar *et al.* who reached a similar conclusion by measuring the c-axis phonon thermal conductivity [31].

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