Superconducting Gap Structure of Spin-Triplet Superconductor Sr$_2$RuO$_4$ Studied by Thermal Conductivity

K. Izawa$^1$, H. Takahashi$^1$, H. Yamaguchi$^1$, Yuji Matsuda$^1$, M. Suzuki$^2$, T. Sasaki$^2$, T. Fukase$^2$, Y. Yoshida$^3$, R. Settai$^3$, and Y. Onuki$^3$

$^1$Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan
$^2$Institute for materials research, Tohoku University, Sendai 980-8577, Japan
$^3$Graduate School of Science, Osaka University, Toyonaka, Osaka, 560-0043 Japan

To clarify the superconducting gap structure of the spin-triplet superconductor Sr$_2$RuO$_4$, 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.

74.70.Pq, 74.25.Fy, 74.25.Jb

Ever since its discovery in 1994[1], the superconducting properties of the layered ruthenate Sr$_2$RuO$_4$ has been attracting a 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, $\mu$SR 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)$_2$PF$_6$[5], though it most probably is also realized in the recently discovered UGe$_2$[6]. While in $^3$He the simplest $p$-wave pairing state is realized, UPt$_3$ seems to be in a more complicated $f$-wave state. At an early stage, the gap symmetry of Sr$_2$RuO$_4$ was discussed in analogue 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, $d(k)=\Delta_0\hat{z}(k_x+ik_y)$, where $\Delta_0$ is a constant, has been proposed as being likely to be realized [1][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 [7]. This implies that the origin of the triplet pairing is not a simple ferromagnetic interaction. Furthermore, the specific heat $C_p$ and NMR relaxation rate $T_1^{-1}$ on very high quality compounds exhibit the power law dependence of $C_p \propto T^2$[9,10] 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 which might explain consistently the spin-triplet superconductivity in the ruthenates [12][13]. 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 $\kappa$, in which only the unpaired electrons are responsible for the thermal transport in the superconducting state. Compared to the specific heat and NMR measurements, an important advantage of the thermal conductivity is that it is a directional probe, sensitive to the orientation relative to the thermal flow, the magnetic field, and nodal directions of the order parameter [14][15]. In fact, a clear 4-fold modulation of $\kappa$ 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$_2$Cu$_3$O$_{7-\delta}$, 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$_2$RuO$_4$, the experimental resolution were not good enough to identify the nodal directions [22]. In the work reported in this Letter, we have 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 $\bf{H}$ rotating within the RuO$_2$ plane, which is sufficient to resolve the gap structure.

Several single crystals with different $T_c$‘s were grown by the floating-zone method. The thermal conductivity was measured by a steady state method with one heater and two ruthenium-oxide thermometers. In the present measurements, it is very important to rotate $\bf{H}$ within the RuO$_2$ planes with high accuracy because a slight field-misalignment produces a large effect on $\kappa$ due to the large anisotropy. For this purpose, we constructed a system with two superconducting magnets generating $\bf{H}$ in two mutually orthogonal directions and a $^3$He cryostat equipped on a mechanical rotating stage with a minimum step of 1/500 degree at the top of the dewar. Computer-controlling two magnets and rotating stage, we were able to rotate $\bf{H}$ continuously within the RuO$_2$ planes with a misalignment less than 0.015 degree from the plane, which we confirmed by the simultaneous measurement of...
the resistivity.

The inset of Fig. 1(a) shows the $T$-dependence of $\kappa/T$ in zero field. Since the electrical resistivity is very small which is an order of $0.1\mu\Omega\cdot\text{cm}$, the electron contribution well dominates over the phonon contribution [23]. At the superconducting transition, $\kappa/T$ shows a kink. At low temperatures, $\kappa/T$ decreases almost linearly with decreasing $T$ with finite residual values at $T = 0$. The residual $\kappa$ decreases with increasing $T_c$ and is very small in the crystal with highest $T_c (=1.45 \text{ K})$. These $T^2$-dependence and the residual $\kappa/T$ are consistent with the presence of the line nodes [10,11].

Figures 1(a) and (b) show the $H$-dependence of $\kappa$ for the sample with $T_c=1.45 \text{ K}$ in perpendicular ($H \perp ab$-plane) and parallel fields ($H \parallel ab$-plane), respectively. In both orientations, $\kappa$ increases with $H$ after the initial decrease at low fields. The consequent minimum is much less pronounced at lower temperatures. At low $T$, $\kappa$ increases linearly with $H$. We note that the $H$-linear dependence of $\kappa$ is observed only in the very clean crystals with $T_c > 1.3 \text{ K}$ and $\kappa$ increases with an upward curvature in samples with lower $T_c$. In parallel field $\kappa$ rises very rapidly as $H$ approaches $H_{c2}$ and attains its normal value with a large slope ($d\kappa/dH$), while $\kappa$ in perpendicular field remains linear in $H$ up to $H_{c2}$. The understanding of the heat transport for superconductors with nodes have largely progressed during past few years [17,24,25]. 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 $\langle \varepsilon(p) \rangle \rightarrow \varepsilon(p) - v_s \cdot \vec{p}$ in the circulating supercurrent flow $\vec{v}_s$ [24]. This effect becomes important at such positions where the local energy gap becomes smaller than the Doppler shift term ($\Delta < v_s \cdot \vec{p}$), 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 $\langle N(\varepsilon) \rangle \propto \varepsilon$, $N(H)$ increases in proportion to $\sqrt{H}$. While the Doppler shift enhances the DOS [26], it also leads to a suppression of both the impurity scattering time and Andreev scattering time off the vortices [21,23]. 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, $\kappa$ increases in proportion to $H$ in the "superclean regime" where the condition, $\frac{\Gamma}{\Delta} \ll \frac{H_{c2}}{T_c}$, is satisfied. Here $\Gamma$ is the pair breaking parameter estimated from the Abrikosov-Gorkov equation $\Psi(1/2 + \Gamma/2\pi T_c) - \Psi(1/2) = \ln(T_c/T_0)$, where $\Psi$ is a digamma function and $T_0$ is the transition temperature in the absence of the pair breaking. Assuming $T_0=1.50 \text{ K}$ and $\Delta = 1.76T_c$, $\Gamma/\Delta$ is estimated to be 0.025 (0.067) for $T_c=1.45 \text{ K}$ ($T_c=1.37 \text{ K}$), 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 $\kappa$ of superconductors with line nodes. The steep increase of $\kappa$ in the vicinity of $H_{c2}$ in parallel field is also observed in pure Nb [27]. When the vortices are close enough near $H_{c2}$, tunneling of the quasiparticle excitations from core to core becomes possible, which leads to large enhancement of quasiparticle mean free path and $\kappa$. The absence of a steep increase in 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 $\kappa$ is present in $H \parallel c$ while is absent in $H \perp b$ [23].

We now move on to the angular variation of the thermal conductivity in parallel field. Figures 2(a) and (b) depict $\kappa(H,\theta)$ as a function of $\theta = (\vec{q},\vec{H})$. No hystere-
sis of \( \kappa \) related to the pinning of the vortices was observed in rotating \( \theta \). 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 \( \kappa_0 \) is \( \theta \)-independent, \( \kappa_{2\theta}(H) = C_{2\theta}(H) \cos 2\theta \) is a term with 2-fold symmetry, and \( \kappa_{4\theta}(H) = C_{4\theta}(H) \cos 4\theta \) with 4-fold symmetry with respect to the in-plane rotation. Figures 3 (a)-(d) show \( \kappa_{4\theta}/\kappa_n \) after the subtraction of \( \kappa_0 \)- and \( \kappa_{2\theta} \)-term from \( \kappa \).

The sign and magnitude of \( C_{2\theta} \) and \( C_{4\theta} \) provide important information on the gap structure. The term \( \kappa_{4\theta} \) appears as a result of difference of the effective DOS for quasiparticles travelling parallel to the vortex and for quasiparticles moving in the perpendicular direction. In the presence of vertical nodes, the term \( \kappa_{4\theta} \) appears as a result of two effects. The first one is the DOS oscillation associated with the rotating \( \mathbf{H} \) within the plane. This effect arises from the fact that DOS depends sensitively on the angle between \( \mathbf{H} \) 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, \( \kappa \) attains the maximum value when \( \mathbf{H} \) is directed to the antinodal directions and becomes minimum when \( \mathbf{H} \) is directed along the nodal directions \([10] \). The second one is the quasiparticle lifetime from the Andreev scattering off the vortex lattice, which has the same symmetry as the gap function \([2,21,24] \). This effect is important at very low fields where \( \kappa \) decreases with \( H \). In addition to the 4-fold symmetry associated with vertical nodes, there is another contribution to \( \kappa_{4\theta} \)-term, which originates from the tetragonal band structure inherent to the Sr₂RuO₄ crystal. We will discuss this effect later.

The most important subject is “Is the observed \( \kappa_{4\theta} \) a consequence of the vertical line nodes?” Before analyzing the data, we list up the various proposed gap functions \([13] \).

1. Type-I: Vertical nodes at \((\pm \pi, 0)\) and \((0, \pm \pi)\) \([2,13] \); \( d(k) = \Delta_0 \hat{z}(\sin k_x + i \sin k_y) \) and \( d(k) = \Delta_0 \hat{z}(k_x + ik_y) \).
2. Type-II: Vertical nodes at \((\pm \pi, \pm \pi)\) \([15,29] \); \( d(k) = \Delta_0 \hat{z}(k_x^2 - k_y^2)(k_x + ik_y) \).
3. Type-III: Horizontal nodes \([3,11] \); \( d(k) = \Delta_0 \hat{z}(k_x + ik_y)(\cos c_k + \alpha) \) with \( \alpha \leq 1 \) (\( c \) is the interlayer distance) and \( d(k) = \Delta_0 \hat{z} k_x(k_x + ik_y)^2 \).

As shown in Figs. 3(a)-(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 \( \kappa \) increases steeply, \( |C_{4\theta}|/\kappa_n \) is of the order of a several % (see Fig. 3(a)). However, \( |C_{4\theta}|/\kappa_n \) decreases rapidly and is about 0.2-0.3% at lower field where \( \kappa \) increases linearly with \( H \) (see Figs. 3(b) and (c)). At very low field where \( \kappa \) decreases with \( H \), no discernible 4-fold 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 where \( \kappa \) obeys an \( H \)-linear dependence \([5] \). We will examine our result in accordance with Ref. \([5] \). For both type I and II symmetries, \( |C_{4\theta}|/\kappa_n \) is expected to be about 6% at low field. Apparently, the observed \( |C_{4\theta}|/\kappa_n \) < 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 4-fold symmetry is an indication of vertical line nodes. We then consider the tetragonal band structure as an origin of $\kappa_{4\theta}$. This effect can be roughly estimated by the in-plane anisotropy of $H_{c2}$. 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 $\phi$ is the angle between $H$ and $a$-axis. In Fig. 4, we plot $|C_{4\theta}| = |A|H d\kappa(H)/dH$ calculated from the in-plane anisotropy of $H_{c2}$ with no fitting parameter. The calculation reproduces the data, indicating that the 4-fold symmetry of $\kappa$ is indeed mainly due to tetragonal band structure.

We next discuss $\kappa_{2\theta}$ which provides an additional important information on the gap structure. According to Ref. [1], a large 2-fold 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 (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 2- and 4-fold 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. [2], a large magnitude of $|C_{2\theta}|/\kappa_n > 30\%$ is expected for $d(k) = \Delta_0 \hat{z}(k_x + ik_y)^2$. In fact, a large 2-fold oscillation is observed in the B-phase of UPt$_3$ with this symmetry $\Gamma_4$. On the other hand, much a smaller $|C_{2\theta}|/\kappa_n \sim 8\%$ is expected at $T = 0$ for $d(k) = \Delta_0 \hat{z}(k_x + ik_y)(\cos ck_z + \alpha)$. Although the value is still several times larger (which may be due to finite temperature effect 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 $d(k) = \Delta_0 \hat{z}(k_x + ik_y)(\cos ck_z + \alpha)$, in which the 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. We finally comment on the orbital-dependent superconductivity scenario, in which three different bands have different superconducting gaps $\Gamma_4$. In this case, our main conclusion can be applicable to the band with the largest gap (presumably the $\gamma$-band).

In summary, the in-plane thermal conductivity of Sr$_2$RuO$_4$ have 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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