Anisotropic Superconducting Gap in the Spin-Triplet Superconductor Sr_2RuO_4: Evidence from a Ru-NQR Study

K. Ishida,[1] H. Mukuda,[1,3] Y. Kitaoka,[1,3] Z. Q. Mao,[2,3] Y. Mori,[2] and Y. Maeno[2,3]

[1]*Department of Physical Science, Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan*
[2]*Department of Physics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan*
[3]*CREST, Japan Science and Technology Corporation, Kawaguchi, Saitama 332-0012, Japan*

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We have investigated a gap structure in the spin-triplet superconductor Sr_2RuO_4 through the measurement of the ^{101}Ru nuclear spin-lattice relaxation rate $^{101}(1/T_1)$ down to 0.09 K at zero magnetic field. In the superconducting state, $1/T_1$ in a high-quality sample with $T_c \sim 1.5$ K exhibits a sharp decrease without the coherence peak, followed by a T^3 behavior down to 0.15 K. This result is in marked contrast to the behavior observed below ~0.4 K in samples with lower T_c, where T_1T is a constant. This behavior is demonstrated to be not intrinsic. We conclude that the gap structure in Sr_2RuO_4 is significantly anisotropic, consistent with line-node-like models.

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There are a lot of experimental evidences that Sr_2RuO_4 [1], with the same crystal structure as a mother compound of high-T_c cuprates, La_2CuO_4 is an unconventional superconductor. The muon-spin-rotation (μSR) experiment revealed that the superconducting (SC) state breaks the time-reversal symmetry [2] and the spin susceptibility of the Cooper pairs extracted from the ^{17}O Knight-shift measurements revealed no change when passing through the SC transition [3]. In particular, the latter result provided decisive evidence that Sr_2RuO_4 is a spin-triplet p-wave superconductor with a finite angular moment of the pairs, as pointed out theoretically at an early stage after the discovery of this compound [4].

In the previous paper, temperature (T) dependence of nuclear spin-lattice relaxation rate, $1/T_1$ of ^{101}Ru was reported on a sample with $T_c \sim 0.7$ K at zero magnetic field by nuclear quadrupole resonance (NQR) [5]. We found that $1/T_1$ exhibits a sharp decrease with no coherence peak just below T_c, followed by a T-linear behavior below 0.4 K. The former result points to an evidence for a non-s-wave SC state and the latter to the existence of the large residual density of states (RDOS) inside the gap. These unconventional features in T_1 data are reminiscent of the relaxation behavior in heavy-fermion superconductors in which the SC gap vanishes along lines and/or on points due to the anisotropic pairing state. The presence of large RDOS was also suggested from the specific-heat measurements, and the RDOS with as large as 60% of the normal-state DOS (N_0), $N_{\rm res}/N_0 \sim 0.6$ was reported in the sample with $T_c \sim 1.14$ K [6]. Since the quality of samples inferred from their residual resistivity was believed to be clean enough, a likely source for the RDOS was expected to originate not from some impurity effect but from the SC nature inherent to Sr_2RuO_4. Several scenarios to interpret the RDOS as an intrinsic SC nature were put forth. A nonunitary p-wave model, realized as in the A_1 phase of superfluid ^{3}He, predicts a large RDOS with $N_{\rm res}/N_0 \sim 0.5$ in the SC state, since the order parameter gives rise to the ungapped branch for the spin up pairs and then a half of the electron system remains ungapped [7,8]. The orbital-dependent superconductivity (ODS) model proposed by Agterberg *et al.* [9], on the other hand, assumes that an interband pairing interaction of quasiparticles is weak between the γ band characterized by the Ru d_{xy} orbital and the α and β bands by the Ru d_{xz} and d_{yz} orbitals so that the SC gap Δ_γ may become an 1 order of magnitude larger than Δ_α and Δ_β in the latter bands. In this case, a partial DOS remains finite in the high-T range where $\Delta_{\alpha,\beta} < k_BT$, and a large RDOS was pointed out to be accountable by this scenario. In order to settle whether the presence of the RDOS is intrinsic or not in the pure Sr_2RuO_4, further experiments on higher quality samples are highly desired.

In this Letter, we report $1/T_1$ measurements on high-quality single crystals (denoted as high-T_c samples hereafter) with a nearly maximum value of $T_c \sim 1.5$ K. Three single crystals with the same quality as in the previous ^{17}O Knight-shift measurements [3] were used in this measurement. The onset temperature of the SC transition in these crystals is T_c ~1.48 K with a transition width $\Delta T_c \sim 50$ mK. T_1 was measured at 6.555 and 3.275 MHz with a respective peak position of ^{101}Ru-NQR spectra corresponding to the ($\pm 3/2 \leftrightarrow \pm 5/2$) and ($\pm 1/2 \leftrightarrow \pm 3/2$) NQR transitions [5]. T_1 was determined by a single component in $T = 4.2$–0.09 K, confirmed to be independent of the NQR frequency. Thus the reliable T_1 data were obtained down to temperature as low as 0.09 K on the single-crystal samples.

The T dependence of $1/T_1$ in the high-T_c sample is indicated in Fig. 1 together with that reported on a sample with $T_c \sim 0.7$ K (low-T_c sample) [5]. As seen clearly in the figure, a $T_1T =$ const behavior in the low-T_c sample is not observable at all in the high-T_c sample in which $1/T_1$ follows a nearly T^3 behavior down to 0.15 K. A possible fraction of RDOS in the high-T_c sample may be

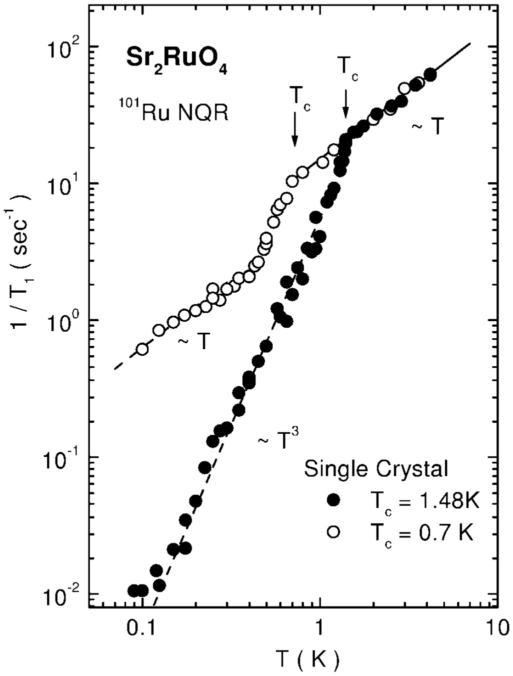

FIG. 1. T dependence of $1/T_1$ in low-T_c and high-T_c samples of Sr_2RuO_4.

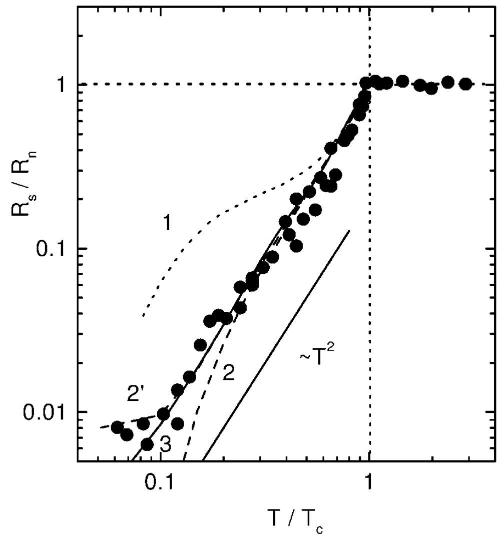

FIG. 2. Plot of R_s/R_n against T/T_c in the high-T_c sample. Solid and dotted lines are the calculated R_s/R_n using the DOS in the superconducting state shown in Fig. 3 (see text).

tentatively estimated to be less than 8.8% from a saturating behavior of $1/T_1$ at low T. This result suggests that the presence of RDOS in the low-T_c samples is extrinsic and caused by their lower quality. By noting that T_c is dramatically decreased with an increase of residual resistivity as reported by Mackenzie *et al.* [10], some nonmagnetic impurity effect [11] should be responsible for inducing the RDOS as observed in unconventional heavy-fermion [12] and high-T_c cuprate superconductors [13,14]. According to a theoretical calculation on T_c/T_{c0} vs $N_{\rm res}/N_0$, where T_{c0} is T_c in pure samples and the nonmagnetic impurity effect is treated in terms of the unitarity limit in two-dimensional (2D) anisotropic superconductors with a line-node gap [14], we estimate $T_{c0} \sim 1.5$ K using an experimental value of $N_{\rm res}/N_0 = 0.6$ and $T_c = 0.7$ K in the low-T_c sample, which is in good agreement with a maximum $T_c \sim 1.5$ K reported so far. The T^3 behavior in the high-T_c sample in $T = 1$–0.15 K suggests a line-node-gap state. These results are incompatible with either the nonunitary p-wave model [7,8] or the ODS model with large RDOS [9]. Furthermore, these observed T_1 behaviors are not in accordance with an isotropic p-wave model of $\boldsymbol{d}(\boldsymbol{k}) = \hat{z}\Delta_0(k_x + ik_y)$ [8], since an exponential decrease of $1/T_1$ should be expected in such a p-wave state. The same conclusion was also presented from the recent specific-heat measurements in the same-quality samples [15].

The T dependence of R_s/R_n [$\equiv (1/T_{1s}T)/(1/T_{1n}T)$] in the high-$T_c$ sample is plotted against T/T_c in Fig. 2. Here $1/T_{1s}(1/T_{1n})$ corresponds to the data in the SC (normal) state. In general, R_s/R_n is related to the DOS in the SC state, $N_s(E)$, and is expressed as

$$\frac{R_s}{R_n} = \frac{2}{k_{\rm B}T}\int_0^\infty [N_s(E)^2 + M(E)^2]f(E)[1 - f(E)]\,dE\,, \tag{1}$$

where $M(E)$ and $f(E)$ are the so-called "anomalous" DOS arising from the coherence effect of scattering inherent to a spin-singlet SC state and the Fermi-distribution function, respectively [16]. Note $M(E) = 0$ for any spin-triplet states. Needless to say, our previous result of the ^{17}O Knight-shift measurement in the SC state excluded the possibility of the spin-singlet state [5]. The sharp decrease of R_s/R_n in Sr_2RuO_4 can be explained reasonably by the anisotropic pairing state such as p- or f-wave states due to the weak divergence of N_s at $E = \Delta$. It should be noted that the T^2 behavior in R_s/R_n at low temperatures evidences $N_s \propto E$ as $E \to 0$ and is hence consistent with an anisotropic SC model with a line-node gap.

The previous Knight-shift results revealed that the $\boldsymbol{d}$ vector of the spin-triplet state is parallel to the z axis expressed by $\boldsymbol{d}(k) = \hat{z}\Delta_0 f(k)$ where the orbital part $f(k)$ determines a gap anisotropy. In order to account for an anisotropic structure of the gap, renewed models have been proposed recently. First is the model proposed by Miyake and Narikiyo (denoted as "MN model" hereafter) [17] in which isotropic $f(k) = k_x + ik_y$ is modified to $f(k) = \sin k_x + i\sin k_y$ due to the $\boldsymbol{k}$ dependence of the pairing interaction mediated by short-range ferromagnetic spin fluctuations. They showed that the amplitude of the gap is described approximately by a fourfold-symmetry gap function of $|\boldsymbol{d}(\boldsymbol{k})| = \Delta_0[1 - r\cos(4\theta_k)]$ with $r = 0.692$. R_s/R_n was calculated by this gap function. The SC DOS on this model is shown in Fig. 3 where

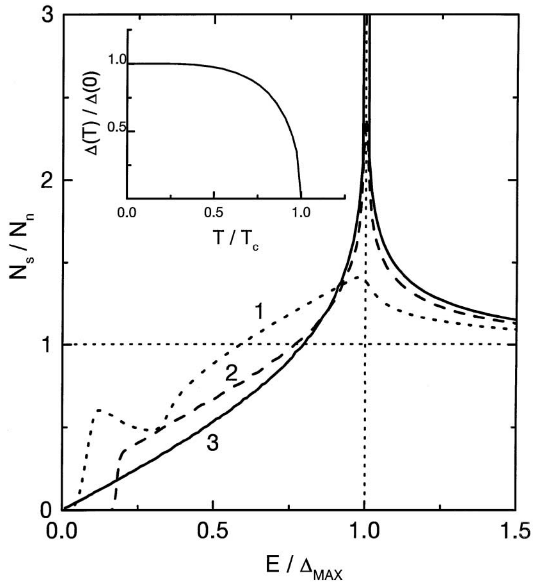

FIG. 3. Density of states in the superconducting state by the several models; ODS model (labeled 1), MK model (labeled 2), and line-node-gap model (labeled 3). The inset is the reduced temperature T/T_c dependence of superconducting gap $\Delta(T)$ divided by $\Delta(0)$, calculated by Eq. (2) with $\Delta(0)/k_B T_c = 1.75$ and $(\Delta C/C_n) = 0.7$.

a finite gap exists below $0.2\Delta_{\rm Max}$ when some impurity effect is neglected.

Second is the E_u model with a line-node gap represented by $f(k) = k_x$ or k_y. This model is incompatible with the experimental results, since this SC state does not break the time-reversal symmetry [2]. Quite recently, Hasegawa *et al.* have proposed a phenomenological model with $\boldsymbol{d}(\boldsymbol{k}) = \hat{z}\Delta_0(k_x + ik_y)\cos(ck_z)$ with the node running parallel to the basal plane to account for all the experimental results [18]. For the model of the line-node gap, the gap function of $\Delta(\phi) = \Delta_0 \cos(\phi)$ was assumed in the 2D cylindrical Fermi surface, where ϕ is the angle in the Fermi surface. This DOS is also indicated in Fig. 3. For comparison, we also present DOS of the ODS model in Fig. 3. Here $(N_\alpha + N_\beta)/N_\gamma = 0.435/0.55$ and $\Delta_{\alpha,\beta}/\Delta_\gamma = 0.1$ are assumed [9].

As for the calculation of $1/T_1$, we adopt the T dependence of the gap which is nearly the same as the BCS theory, given by

$$\Delta(T) = \Delta(0)\tanh\left[\frac{\pi k_B T_c}{\Delta(0)}\sqrt{\left(\frac{\Delta C}{C_n}\right)\left(\frac{T_c}{T} - 1\right)}\right], \quad (2)$$

where $(\Delta C/C_n)$ is a jump in specific heat and $(\Delta C/C_n) = 0.7$ in the high-T_c sample [15], which is shown in the inset of Fig. 3. The calculated results of R_s/R_n based on the three models are indicated in Fig. 2 by dashed, dotted, and solid lines labeled as 1 (ODS model), 2 (MN model), and 3 (line-node-gap model).

R_s/R_n based on the ODS model with $\Delta_0/k_B T_c = 2.5$ (curve 1) is not in accordance with the experimental data below $0.6T_c$. In the ODS model, a tiny gap with $\Delta_{\alpha,\beta}/\Delta_\gamma \sim 0.1$ opens for $k_B T < \Delta_{\alpha,\beta}$. One would expect some anomaly well below T_c as seen actually in curve 1 in Fig. 2. This is, however, not the case. Furthermore, from experimental R_s/R_n we may tentatively estimate a possible fraction of RDOS, $N_{\rm res}/N_0 < 0.1$ at $0.1T_c$. The present T_1 results seem to be unaccountable based on the ODS model at least with the parameters in Ref. [9].

Curve 2 in the MN model with $\Delta_0/k_B T_c = 1.75$ gradually deviates from the experimental R_s/R_n below $0.3T_c$ due to the existence of a finite gap as expected from the DOS in Fig. 3. However, if some impurity effect is taken into account, the calculated R_s/R_n is altered into curve 2' especially in a low-T regime [17]. In curve 2', a fraction of RDOS with $N_{\rm res}/N_0 \sim 0.09$ is in good agreement with the experimental result as seen in Fig. 2. From the relation of T_c/T_{c0} vs $N_{\rm res}/N_0 \sim 0.09$ and using $T_c = 1.48$ K [17], $T_{c0} \sim 1.5$ K can also be estimated. Curve 3 in the line-node-gap model with $\Delta/k_B T_c = 2$ appears to be consistent with the experimental result as well [19]. In this model, it would be possible that some deviation of R_s/R_n from a T^2-like behavior is due to the nonmagnetic impurity effect which induces the RDOS. Although it is difficult to distinguish between the MN model and the line-node-gap model from the present experiment, it is clear that the SC gap in Sr_2RuO_4 possesses a highly anisotropic nature characterized by the line-node-like models and that the large RDOS seen in the low-T_c sample was induced by impurities and/or crystal imperfection. As a result, we conclude that the ODS model with the parameters in Ref. [9] and the nonunitary p-wave model [7,8] will be ruled out.

Finally, we compare in Fig. 4 the T dependence of $1/T_1$ in Sr_2RuO_4 [20,21] with that in the first odd-parity anisotropic superconductors UPt_3 [22,23]. The T^3 behavior in the SC state is remarkable in both the compounds, indicative of an anisotropic SC gap. A T-linear behavior of $1/T_1$ characteristics of the Fermi liquid (FL) state is observed below $T_F = 5$ K in UPt_3 and $T_F = 30$ K in Sr_2RuO_4. We note that $T_c/T_F \ll 1$ in these spin-triplet superconductors, suggesting that the triplet superconductivity occurs under the well-established FL state. On the other hand, it is notable that $T_c/T_F \geq 1$ in the d-wave superconductors of $CeCu_2Si_2$ and $La_{1.85}Sr_{0.15}CuO_4$, suggesting that the onset of superconductivity emerges before the stabilization of the FL state and/or even in the non-FL state. Further remarkable difference between the spin-triplet p-wave and the singlet d-wave superconductors is seen in the dependence of T_c on their residual resistivity. We remark that the superconductivity in the former emerges only for the very clean sample. By contrast, the T_c in the latter is not so sensitive to the residual resistivity. As pointed out by Rice [24], for the cuprates it seems that short-range antiferromagnetic coupling, which

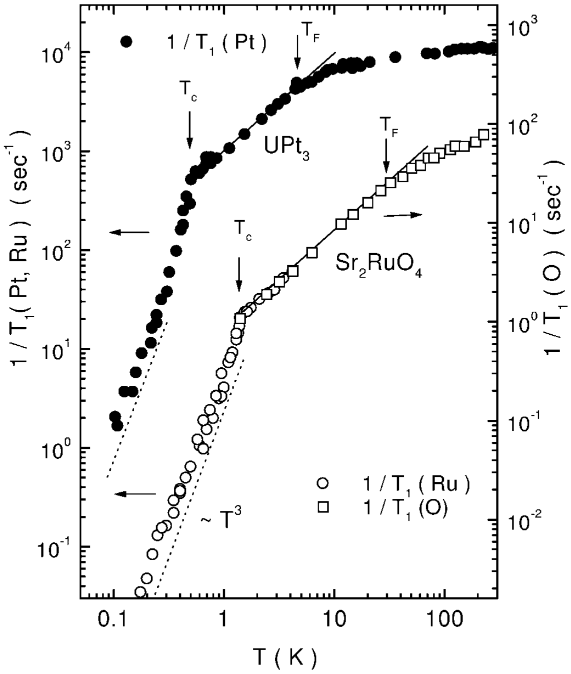

FIG. 4. $1/T_1$ in spin-triplet superconductors, UPt_3, and Sr_2RuO_4 [22] (see text).

presumably mediates the spin-singlet d-wave Cooper pairing, brings about a strong pairing interaction under the formation of the resonant valence bond state for the d-wave superconductivity. By contrast, the triplet p-wave pairing interaction, which is possibly mediated via ferromagnetic spin fluctuations, appears to be weak under the well-established FL state.

In conclusion, the measurement of $1/T_1$ down to 0.09 K on the spin-triplet p-wave superconductor Sr_2RuO_4 with $T_c = 1.48$ K clarified that the superconducting gap possesses the highly anisotropic structures. $1/T_1$ below T_c shows the sharp decrease without the coherence peak just below T_c, followed by a T^3 dependence down to 0.15 K. This is indicative of the line-node state. The origin of the $T_1T =$ const behavior for the sample with $T_c = 0.7$ K was ascribed to the impurity effect. Through the comparison with several models proposed so far, we showed that the nonunitary p-wave model [7,8] and the ODS model with the parameters in Ref. [9] are inadequate, but the p-wave models described by either $\boldsymbol{d}(\boldsymbol{k}) = \hat{z}\Delta_0(\sin k_x + i\sin k_y)$ with a strongly anisotropic gap or $\boldsymbol{d}(\boldsymbol{k}) = \hat{z}\Delta_0(k_x + ik_y)\cos(ck_z)$ with the line node are promising if the nonmagnetic impurity effect is incorporated.

Quite recently, an NMR experimental result was presented which supports the hypothesis of spin-triplet superconductivity in $(TMTSF)_2PF_6$ [25]. In addition, it was reported that ferromagnetic compound UGe_2 shows the superconductivity under pressure, which can be a candidate for spin-triplet superconductivity [26]. The physics of the spin-triplet superconductivity will be discussed more vigorously with new perspective.

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