PHYSICAL REVIEW B, VOLUME 63, 060505(R)

Ultrasonic studies of the spin-triplet order parameter and the collective mode in Sr_2RuO_4

H. Matsui,[1] Y. Yoshida,[2] A. Mukai,[2] R. Settai,[2] Y. Ōnuki,[2] H. Takei,[2] N. Kimura,[3] H. Aoki,[3] and N. Toyota[1]
[1]*Department of Physics, Graduate School of Science and Center for Interdisciplinary Research, Tohoku University, Sendai 980-8578, Japan*
[2]*Graduate School of Science, Osaka University, Toyonaka 560-0043, Japan*
[3]*Center for Low Temperature Science, Tohoku University, Sendai 980-8578, Japan*

(Received 27 August 2000; published 22 January 2001)

The elastic constant C_T and ultrasonic attenuation α_s for $(C_{11}-C_{12})/2$ have been measured on Sr_2RuO_4 with $T_c^*=1.42$ K. In contrast to the A_{1g} and E_g strains, the B_{1g} strain ($\epsilon_{xx}-\epsilon_{yy}$) strongly couples to the superconducting state, which dominantly originate from hybridized Ru-4d_{xy} electrons on the γ Fermi surface. Taken into account impurity induced in-gap states, the T^3 dependence of α_s is found to be consistent with an existence of line nodes. The large reduction in C_T below T_c^* evidences the two-component order parameter and chiral p-wave state. Anomalous C_T and α_s around $T_a=1.34$ K suggest a collective mode.

DOI: 10.1103/PhysRevB.63.060505 PACS number(s): 74.25.Ld, 74.70.−b

Layered perovskite superconductor Sr_2RuO_4 has attracted much attention,[1] because the pairing symmetry could be of p wave. Two-dimensional electronic states are formed on the alternating RuO_2 planes. There exist three cylindrical Fermi surfaces named α, β, and γ running along [001] in a tetragonal structure.[2–4] The nuclear relaxation rate T_1 measured by ^{101}Ru nuclear quadrupole resonance (NQR) never exhibits a Hebel-Slichter coherence peak, and the Knight shift of ^{17}O nuclear magnetic resonance (NMR)[5] is temperature independent below and above T_c. The muon spin relaxation indicates a broken time-reversal symmetry in the superconducting state.[6] These experimental results were interpreted with $\boldsymbol{d}$ vector $\boldsymbol{z}\Delta_0(k_x+ik_y)$ for an isotropic gap.[7] Here $\boldsymbol{z}$ and k_x+ik_y represent spin and orbital parts, and Δ_0 is a constant.

Recent specific heat[8] and ^{101}Ru NQR measurements,[9] however, show power-law behaviors at low temperatures, suggesting an existence of a line-node gap. An anisotropic gap model[10] shows the p-wave superconductivity induced by the short-range ferromagnetic spin fluctuation. Various f-wave pairing states are obtained by the product of two irreducible representations in a D_{4h} point group.[11]

In p-wave superfluid ^{3}He, many collective modes were extensively studied by ultrasonic measurements.[12] However, collective modes caused by multicomponent order parameters in an unconventional superconductor[13] have been little known experimentally at least.

We have already reported a small superconducting anomaly below $T_c=1.2$ K in the longitudinal mode C_{33} inducing a volume strain with A_{1g} representation of D_{4h}.[4] On the other hand, a large temperature dependence is found in the longitudinal mode C_{11}, which is reducible to A_{1g} and B_{1g}. With this indication that the B_{1g} strain $\epsilon_{xx}-\epsilon_{yy}$ might strongly couple to the superconductivity, we have measured the ultrasound velocity and attenuation of $(C_{11}-C_{12})/2$ mode to clarify the superconducting order parameter and the collective mode.

We have used a single crystal of Sr_2RuO_4 grown by a floating zone technique with the size of 3.02([110]) $\times$2.10([1$\bar{1}$0])$\times$2.51([001]) mm^3. Ultrasonic experiments based on a phase comparison method with high sensitivity of $\Delta v/v=10^{-5}\sim10^{-6}$ have been carried out down to 0.17 K with a ^{3}He-^{4}He dilution refrigerator. The attenuation coefficient has been measured by the amplitude variation of the 2nd-echo signal with the input power of $\sim$30 μW. The 5th-higher overtone $\sim$43.4 MHz has been employed to increase the resolution. The transverse ultrasound propagating and polarizing along [110] and [1$\bar{1}$0] is excited and detected by the $LiNbO_3$ transducers stuck to both the parallel sample surfaces by RTV silicone.

The superconducting transition for the present crystal is determined by the specific heat using a quasiadiabatic heat pulse method. As shown in Fig. 1(a), the mean-field superconducting transition temperature is obtained as $T_c^*=1.42$ K by the mid point of the specific heat jump. The jump between $T_c^{\text{offset}}=1.37$ K and $T_c^{\text{onset}}=1.50$ K is estimated to be $\Delta C_p=4.5\times10^3$ erg/cm^3 K. The transition width is attributed to the sample inhomogeneity due to the spatial distribution of T_c. In a spin-triplet superconductor, the nonmagnetic impurities act as a pair breaker. According to the analysis with the Abrikosov-Gor'kov theory,[14] T_c^0 for an ideal case has been estimated to be 1.5 K,[15] and the quasiparticle life time due to the impurity scattering is obtained as $\tau_s=7.5\times10^{-11}$ sec for the present crystal.

The temperature dependence of the elastic constant $C_T=(C_{11}-C_{12})/2$ is depicted in Fig. 1(b). $\Delta C_T=C_T(T,H=0)-C_T(T,H=0.1\ \text{T})$ is the difference in the elastic constant between superconducting and normal states, where $H=0.1$ T applied along [001] is higher than the upper critical field at $T=0$. The experimental data deviate from $\Delta C_T=0$ below T_c^{onset}. The difference ΔC_T is theoretically calculated by the second derivative of a Helmholtz free energy based on a two-fluid model $\Delta F=-\phi(\epsilon_\Gamma)[1-T^2/T_c^{*2}(\epsilon_\Gamma)]^2$ with respect to the strain ϵ_Γ.[16] Here the condensation energy ϕ at 0 K and T_c^* depends on the strain. In general, the longitudinal elastic constant exhibits a jump at T_c^* due to the first-order derivative term $dT_c^*/d\epsilon_\Gamma$.

For the transverse strain ϵ_Γ,

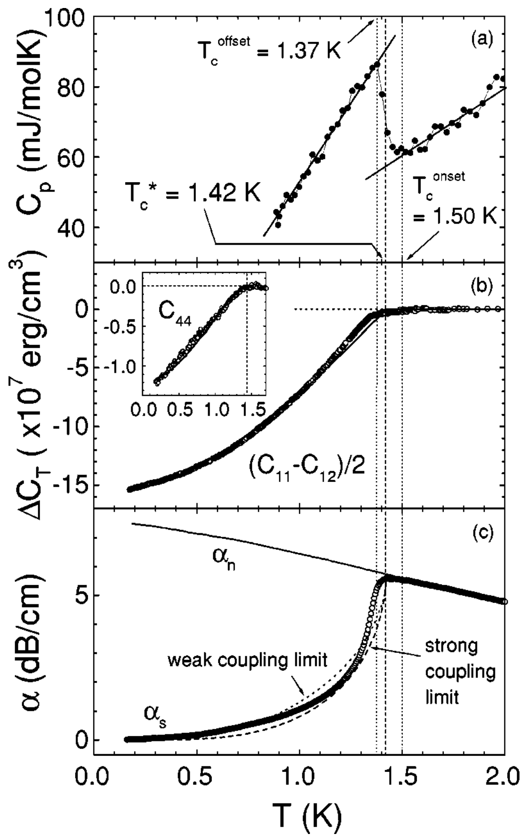

FIG. 1. Temperature dependence of the specific heat (a), the elastic constant (b), and the attenuation coefficient (c) for $(C_{11}-C_{12})/2$. The inset in (b) shows ΔC_T in C_{44}. See the text for details.

$$\Delta C_{\mathrm{T}}=\frac{d^2T_c^*}{d\epsilon_\Gamma^2}\left[-\frac{T^2}{2T_c^{*2}}\left(1-\frac{T^2}{T_c^{*2}}\right)\right]\Delta C_{\mathrm{p}}-\frac{d^2\phi}{d\epsilon_\Gamma^2}\left(1-\frac{T^2}{T_c^{*2}}\right)^2. \tag{1}$$

Equation (1) only includes the second-order derivative term $d^2T_c^*/d\epsilon_\Gamma^2$, so that the discontinuity in the slope $d\Delta C_T/dT$ occurs at T_c^*. As shown by a solid line in Fig. 1(b), Eq. (1) with parameters $d^2T_c^*/d\epsilon_{B1g}^2=7\times10^4$ K and $\phi^{-1}(d^2\phi/d\epsilon_{B1g}^2)=2\times10^5$, where $\epsilon_{B1g}=\epsilon_{xx}-\epsilon_{yy}$, can be fit well to the data except just below T_c^*.

For the transverse mode C_{44} with E_g inducing $\epsilon_{Eg}=\epsilon_{zx}$ in the inset of Fig. 1(b), we obtain $d^2T_c^*/d\epsilon_{Eg}^2=3\times10^3$ K and $\phi^{-1}(d^2\phi/d\epsilon_{Eg}^2)=1\times10^4$. Equation (1) reproduces the data quite well just below T_c^*. In case of C_{33},[4] $d^2T_c^*/d\epsilon_{A1g}^2=2\times10^4$ K, while the first-order derivative term is extremely small because the longitudinal elastic constant does not exhibit a remarkable jump at T_c. Therefore the superconducting electrons couple strongly to the transverse B_{1g} strain, which induces the in-plane perturbation for the two-dimensional electronic states.

Here we consider the electron-strain coupling which is measurable via an area coefficient Λ_Γ derived by the acoustic de Haas–van Alphen (dHvA) effect. The γ oscillation has not been detected by this effect owing to the large effective mass. The coefficient for C_{33} with the α Fermi surface,[4] Λ_{A1g}^{α}, is quite large, about 20, while those of $(C_{11}-C_{12})/2$ with α and β Fermi surfaces are quite small,[17] less than 1. Therefore, the α and β Fermi surfaces constructed by the hybridized Ru-4$d_{yz(zx)}$ orbitals as predicted from the band-structure calculations[2] couple strongly with the A_{1g} strain and quite weakly with the B_{1g} strain. These results are totally consistent with the orbital-strain symmetry. Since the superconducting state exclusively couples to the B_{1g} strain as mentioned above, the hybridized Ru-4d_{xy} orbital electron on the γ Fermi surface is concluded to be dominantly responsible for the superconductivity.

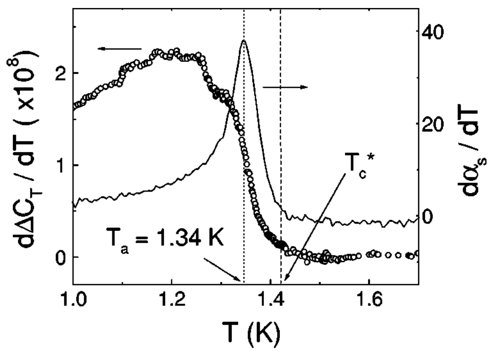

FIG. 2. Temperature derivative of ΔC_T (open circles) and α_s (solid line).

The temperature dependence of the attenuation coefficient is shown in Fig. 1(c) for both the superconducting state (α_s) at $H=0$ and the normal one (α_n) under $H=0.1$ T. With decreasing temperature, α_n monotonically increases as expected for a pure metal. At T_c^{onset}, α_s starts to deviate from α_n as well as the elastic constant mentioned above, and then drops sharply well below T_c^*.

The attenuation coefficient for a conventional superconductor is written as:[18]

$$\frac{\alpha_s}{\alpha_n}=2f_0(\Delta(T))=\frac{2}{1+\exp(\Delta/kT)}. \tag{2}$$

Here Δ is a temperature-dependent energy gap isotropically opened on a Fermi surface. The dotted and broken curves in Fig. 1(c) show the calculated results with Eq. (2) for a weak and strong coupling limit $[\Delta(0)/k_BT_c^*=3.52$ and $3.88]$. Both curves become exponentially zero, while the data are still temperature dependent down to 0.17 K.

Figure 2 shows the temperature-derivative of ΔC_T and α_s. There appears a sharp peak in $d\alpha_s/dT$ at $T_a=1.34$ K, around which the discontinuous change in $d\Delta C_T/dT$ takes place. These anomalies corresponding to the anomalous rounding in ΔC_T and α_s do occur not at T_c^* but at T_a well below T_c^*.

In Fig. 3, the ratio α_s/α_n near T_c^* is shown by the solid circles as a function of $(T-T_c^*)/T_c^*$. The broken curve is a BCS result with Eq. (2) for a strong coupling limit. To take account of the inhomogeneity effect, a Gaussian distribution function for T_c is applied to Eq. (2): $\Psi(T_c)=(2\pi\sigma)^{-1}\exp(-(T_c-T_c^*)^2/2\sigma^2)$, the sum of which for all the T_c's are normalized to unity. The most probable standard

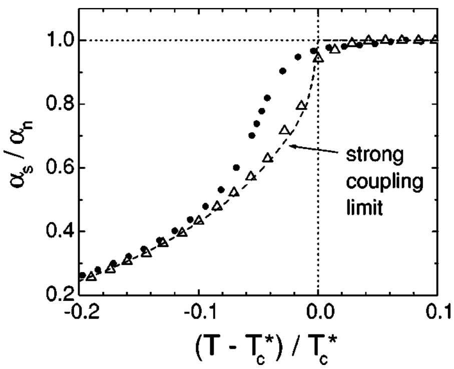

FIG. 3. The ratio α_s/α_n close to T_c^* as a function of $(T - T_c^*)/T_c^*$.

deviation is obtained as $\sigma = 0.025$. For the sake of simplicity, we divide the temperature region with 0.02 K step. The open triangles in Fig. 3 are obtained by summing up Eq. (2) weighted by $\Psi(T_c)$. The data above T_c^* are well reproduced by the inhomogeneity effect. However this effect below T_c^*, which is so small due to the overwhelming temperature dependence [Eq. (2)], cannot explain the data at all. Therefore some additional attenuation exists at temperatures ~ 1.2 K $\leq T \leq T_c^*$.

Next we discuss the nonexponential dependence of the attenuation as shown in Fig. 1(c). At high temperatures, say above 0.8 K, the attenuation no longer follows a power-law behavior because of the temperature-dependent gap. The low-temperature data are well fitted to T^2 for 0.17 K$\lesssim T \lesssim$0.4 K and T^3 for 0.3 K$\lesssim T \lesssim$0.8 K, as shown in Fig. 4. The crossover from T^3 to T^2 dependence occurs around 0.4 K with decreasing temperatures. Since the absolute attenuation coefficient due to the electrons is hardly obtained in general, the extrapolation of a T^2 line to $T=0$ is set to zero.

When we determine the power-law in an anisotropic superconductor, the impurity effect inducing a low-lying in-gap states should be taken into account. The rate of an impurity scattering is represented by the pair-breaking parameter:[10,19] $\eta_c = \hbar[2\tau_s\Delta(0)]^{-1}$, where $\Delta(0)$ is a maximum gap at $T=0$. When we assume $\Delta(0) = (1.75–2.5)k_B T_c^*$, η_c is calculated to be 0.014–0.021 for the present crystal. The residual density of states (DOS) is estimated to be about $0.3N_F$, where N_F is a normal-state DOS. Thus the impurity effect plays a significant role below $T_{imp} = \eta_c^{1/2} T_c^* \sim 0.2$ K, which locates in the T^2 regime. Therefore, the crossover could be attributed to the impurity effect, which is observed in T_1^{-1} of the recent ^{101}Ru NQR[9] deviating from T^3 dependence below 0.15 K.

The T^3 dependence seen at temperatures higher than T_{imp} is compared with the theory. According to the analysis for the transverse attenuation in an anisotropic superconductor,[19] the T^3 dependence is characteristic for an existence of line nodes. The $\boldsymbol{d}$ vector might be expressed by $\Delta_0 \boldsymbol{z}(k_x^2 - k_y^2)$ $(k_x + ik_y)$, $\Delta_0 \boldsymbol{z}(k_x k_y)(k_x + ik_y)$,[11] or $\Delta_0 \boldsymbol{z}(\sin k_x + i \sin k_y)$,[10] all of which have the same symmetry with $\Delta_0 \boldsymbol{z}(k_x + ik_y)$.[7] It is noted that the $\boldsymbol{d}$ vector does not necessarily contain B_{1g}, although the superconducting state strongly couples to $\epsilon_{xx} - \epsilon_{yy}$. Because the order parameter

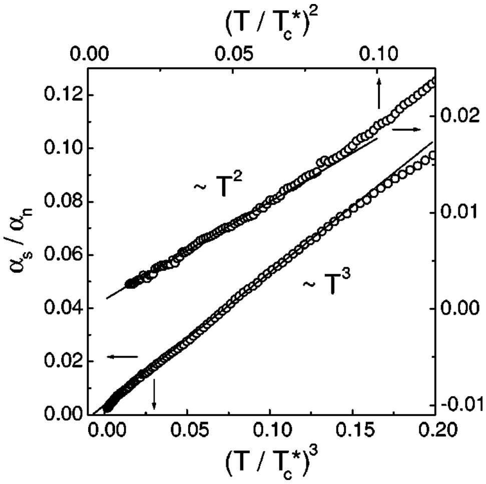

FIG. 4. The ratio α_s/α_n as a function of $(T/T_c^*)^n$ ($n=2$ and 3).

mentioned later [Eq. (3)] always appears in square in the strain-order parameter coupling.[20]

The elastic anomalies are discussed from the viewpoint of the strain-order parameter coupling.[13,20,21] The complex order parameter is defined as $(\eta_1, \eta_2) = |\eta|(\cos\theta, e^{i\phi}\sin\theta)$ belonging to the two-dimensional E_u. The strain couples linearly to the bilinear products of the order parameter.[13] The free energy for the strain-order parameter coupling of the lowest order is expressed as

$$F_{so} = g_{A1g}\epsilon_{zz}(|\eta_1|^2 + |\eta_2|^2) + g_{B1g}(\epsilon_{xx} - \epsilon_{yy}) \times (|\eta_1|^2 - |\eta_2|^2) + g_{B2g}\epsilon_{xy}(\eta_1^* \eta_2 + \eta_1 \eta_2^*), \quad (3)$$

where the parameters g_Γ denote the coupling constants. According to the small change in C_{33} below T_c mentioned before,[4] the amplitude mode $|\eta|$ relating to A_{1g} does not make a dominant contribution. It is noted that $|\eta|$ plays an important role in UPt_3 and URu_2Si_2, where the noticeable jump is observed at T_c in longitudinal modes.[21] From the 2nd term, the coupling of $\epsilon_{xx} - \epsilon_{yy}$ to the order parameter originates from the phase mode θ. The large change in ΔC_T below T_c^*, which is a direct evidence for the two-component order parameter and the chiral p-wave superconducting state,[20] results from the significant contribution of θ to the superconducting state. The phase mode ϕ in the 3rd term could be clarified by the measurement of C_{66}.

Finally we discuss the additional attenuation around T_a (Fig. 3), where the whole volume of the sample completes the transition to the superconducting state below $T_c^{\rm offset}$. Moreover, ΔC_T around T_a is larger than the calculated curve in Fig. 1(b), and thus there should exist some mechanism supplying the excess entropy. Such anomaly, however, is absent in C_{33} and C_{44}. These facts could be interpreted as an excitation of the superconducting collective mode by the transverse wave in $(C_{11} - C_{12})/2$. From the previous discussions on the strain-order parameter coupling, the phase mode θ associated with a chirality should correspond to the collective mode. Several collective order-parameter modes are theoretically discussed for a *p*-wave pairing with $\mathbf{z}\Delta_0(k_x+ik_y)$.[22] In longitudinal modes of UPt_3, a clear peak is emerged just below T_c, which could originate from the density correlation.[23] From the viewpoint of ultrasonic studies, it is evident that the *p*-wave superconducting state in Sr_2RuO_4 is quite different from that in UPt_3. Since the present measurements have been performed under hydrodynamic condition with large damping, further ultrasonic experiments with high frequency are encouraged to clarify the collective mode.

We would like to thank M. Sigrist for valuable discussions and suggestions. This work was supported by the Grant-in-Aid for Encouragement of Young Scientists from Japan Society for the Promotion of Science under Grant No. 11740182, and REIMEI Research Resources of Japan Atomic Energy Research Institute.

[1] Y. Maeno, H. Hashimoto, K. Yoshida, S. Nishizaki, T. Fujita, J.G. Bednorz, and F. Lichtenberg, Nature (London) **372**, 532 (1994).
[2] Y. Yoshida, A. Mukai, R. Settai, K. Miyake, Y. Inada, Y. Ōnuki, K. Betsuyaku, H. Harima, T. Matsuda, Y. Aoki, and H. Sato, J. Phys. Soc. Jpn. **68**, 3041 (1999).
[3] A.P. Mackenzie, S.R. Julian, A.J. Diver, G.J. McMullan, M.P. Ray, G.G. Lonzarich, Y. Maeno, S. Nishizaki, and T. Fujita, Phys. Rev. Lett. **76**, 3786 (1996).
[4] H. Matsui, M. Yamaguchi, Y. Yoshida, A. Mukai, R. Settai, Y. Ōnuki, H. Takei, and N. Toyota, J. Phys. Soc. Jpn. **67**, 3687 (1998).
[5] K. Ishida, H. Mukuda, Y. Kitaoka, K. Asayama, Z.Q. Mao, Y. Mori, and Y. Maeno, Nature (London) **396**, 658 (1998).
[6] G.M. Luke, Y. Fudamoto, K.M. Kojima, M.I. Larkin, J. Merrin, B. Nachumi, Y.J. Uemura, Y. Maeno, Z.Q. Mao, Y. Mori, H. Nakamura, and M. Sigrist, Nature (London) **394**, 558 (1998).
[7] M. Sigrist, Physica B **280**, 154 (2000).
[8] S. Nishizaki, Y. Maeno, and Z.Q. Mao, J. Phys. Soc. Jpn. **69**, 572 (2000).
[9] K. Ishida, H. Mukuda, Y. Kitaoka, Z.Q. Mao, Y. Mori, and Y. Maeno, Phys. Rev. Lett. **84**, 5387 (2000).
[10] K. Miyake and O. Narikiyo, Phys. Rev. Lett. **83**, 1423 (1999).
[11] Y. Hasegawa, K. Machida, and M. Ozaki, J. Phys. Soc. Jpn. **69**, 336 (2000).
[12] B.K. Sarma, J.B. Ketterson, S. Adenwalla, and Z. Zhao, in *Physical Acoustics*, edited by M. Levy (Academic Press, Boston, 1992), Vol. XX, p. 23
[13] M. Sigrist and K. Ueda, Rev. Mod. Phys. **63**, 239 (1991).
[14] A.A. Abrikosov and L.P. Gor'kov, Zh. Éksp. Teor. Fiz. **39**, 1781 (1960) [Sov. Phys. JETP **12**, 1243 (1961)].
[15] A.P. Mackenzie, R.K.W. Haselwimmer, A.W. Tyler, G.G. Lonzarich, Y. Mori, S. Nishizaki, and Y. Maeno, Phys. Rev. Lett. **80**, 161 (1998).
[16] L.R. Testardi, Phys. Rev. B **12**, 3849 (1975).
[17] H. Matsui, Y. Yoshida, A. Mukai, R. Settai, Y. Ōnuki, H. Takei, N. Kimura, H. Aoki, and N. Toyota, J. Phys. Soc. Jpn. **69**, 3769 (2000).
[18] J. Bardeen, L.N. Cooper, and J.R. Schrieffer, Phys. Rev. **108**, 1175 (1957).
[19] K. Miyake, J. Magn. Magn. Mater. **63&64**, 411 (1987).
[20] M. Sigrist (private communication and unpublished).
[21] P. Thalmeier, B. Wolf, D. Weber, G. Bruls, B. Lüthi, and A.A. Menovsky, Physica C **175**, 61 (1991).
[22] L. Tewordt, Phys. Rev. Lett. **83**, 1007 (1999).
[23] V. Müller, D. Maurer, E.W. Scheidt, Ch. Roth, K. Lüders, E. Bucher, and H.E. Bömmel, Solid State Commun. **57**, 319 (1986).