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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*

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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.

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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 $T_c^{\rm offset}=1.37$ K and $T_c^{\rm onset}=1.50$ 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). $\Delta C_T=C_T(T,H=0)-C_T(T,H=0.1\ \mathrm{T})$ 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 $T_c^{\rm onset}$. 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,

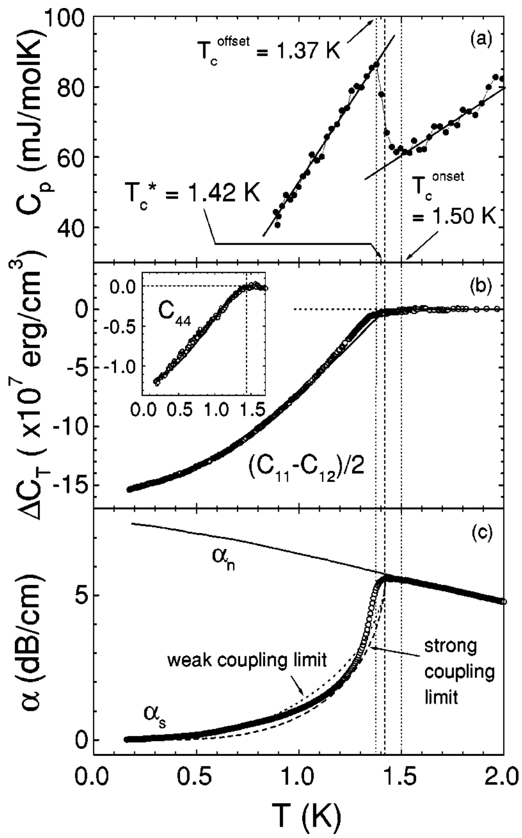

FIG. 1. Temperature dependence of the specific heat (a), the elastic constant (b), and the attenuation coefficient (c) for $(C_{11}-C_{12})/2$. The inset in (b) shows ΔC_T in C_{44}. See the text for details.

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

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.

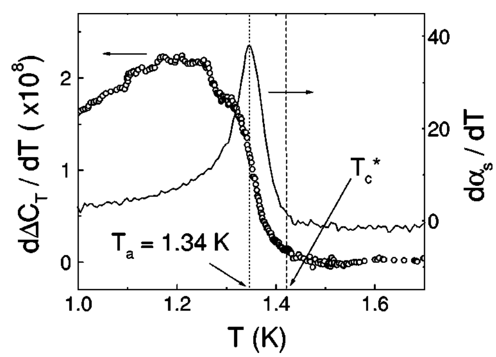

FIG. 2. Temperature derivative of ΔC_T (open circles) and α_s (solid line).

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

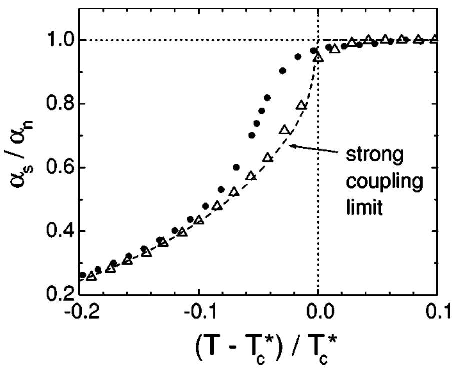

FIG. 3. The ratio α_s/α_n close to T_c^* as a function of $(T-T_c^*)/T_c^*$.

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 $\sim$1.2 K $\leqslant T \leqslant T_c^*$.

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 $\Delta(0)=(1.75\text{–}2.5)k_BT_c^*$, 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), $\Delta_0\boldsymbol{z}(k_xk_y)(k_x+ik_y)$,[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

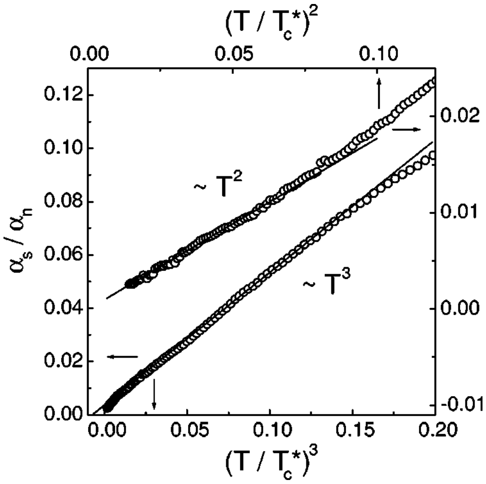

FIG. 4. The ratio α_s/α_n as a function of $(T/T_c^*)^n$ ($n=2$ and 3).

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^{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.

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