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Raman spectra of triplet superconductivity in Sr_2RuO_4

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We study the Raman spectra of spin-triplet superconductors in Sr_2RuO_4. The *p*-wave and *f*-wave symmetries are considered. We show that there is the clapping mode with frequency of $\sqrt{2}\Delta(T)$ and $\Delta(T)$ for *p*-wave and *f*-wave superconductors, respectively. This mode is visible as a huge resonance in the B_{1g} and B_{2g} modes of Raman spectra. We discuss the details of the Raman spectra in these superconducting states.

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I. INTRODUCTION

The superconductivity in Sr_2RuO_4 was discovered in 1994.[1] Shortly after the discovery of superconductivity, a possible triplet *p*-wave superconductivity with the following order parameter was postulated by Rice and Sigrist:[2,3]

$$\hat{\Delta}(\mathbf{k})=\Delta\hat{d}(k_x\pm ik_y), \tag{1}$$

where Δ is the magnitude of the superconducting order parameter. Here the $\hat{d}$ vector is called the spin vector perpendicular to the direction of the spin associated with the condensed pair.[4] Notice that this state is analogous to the *A* phase of ^{3}He and there is a full gap on the Fermi surface. Subsequently, the triplet superconducting nature has been confirmed by the constancy of ^{17}O Knight shift (spin susceptibility) through T_c for the magnetic field parallel to the *a*-*b* plane.[5] The broken time reversal symmetry state have been also confirmed by the spontaneous magnetic moment found in μSR experiment.[6] In general, the triplet superconductors have a variety of collective modes. The spin waves and the clapping mode with the order parameter of Eq. (1) were studied in Refs. 7–9. Further, the effect of the clapping mode on the sound wave was studied.[9] However, this coupling is very small to detect the existence of the clapping mode.

In the meantime, the sample quality of Sr_2RuO_4 has been improved. The cleanest sample shows the transition temperature close to the optimal $T_c=1.5$ K deduced from the T_c dependence of the residual resistivity.[10] All these high quality samples exhibit the character of nodal superconductors; the T^2 behavior of the specific heat,[11] the *T*-linear dependence of superfluid density,[12] the T^3 behavior of $1/T_1$ in NMR,[13] the T^2 behavior of the ultrasonic attenuation,[14] and the $\sqrt{H}$ dependence of the specific heat in a magnetic field[11] at low temperature. These low temperature behaviors of the specific heat and superfluid density are consistent with a *f*-wave superconductor with nodes in the order parameter.[15–19]

On the other hand, the quasi-two-dimensional system with strong paramagnon (the ferromagnetic spin fluctuation) favors the *p*-wave superconductor as given in Eq. (1).[20,21] In order to understand this situation, the nodal structure of the order parameter becomes of crucial importance. In a series of papers, Won and Maki have shown that the magnetothermal conductivity for $T\ll\tilde{v}\sqrt{eH}\ll\Delta(0)$ will provide the direct access to this question,[22,23] where $\tilde{v}=\sqrt{v_a v_c}$ and $v_{a,c}$ are the anisotropic Fermi velocities. Such experiments have been carried out by Tanatar *et al.*[24] and Izawa *et al.*[25] The result is consistent with the *f*-wave superconductor with horizontal nodes of Eq. (2), even though the thermal conductivity data cannot exclude a small admixture, a few % of *p*-wave order parameter (1):

$$\hat{\Delta}(\mathbf{k})=\Delta\hat{d}[(k_x\pm ik_y)\cos(ck_z)], \tag{2}$$

where *c* is the lattice constant along the *c* axis.

Parallel to this development, Zhitomirsky and Rice[26] have proposed an alternative model, multigap model for Sr_2RuO_4. As it is known that the Fermi surfaces in Sr_2RuO_4 consist of three different bands labeled by α, β, and γ bands,[3] it is also believed that the superconductivity arises mainly in the γ band. It was proposed that a full gap with *p* wave, Eq. (1) exists in the active band γ, while line nodes with an *f*-wave order parameter, Eq. (3) develops in the α and β due to proximity effect:[26]

$$\hat{\Delta}_{zr}(\mathbf{k})=\Delta\hat{d}[(k_x\pm ik_y)\cos(ck_z/2)]. \tag{3}$$

While this model could reproduce the specific heat data by Nishizaki *et al.*,[11] and the magnetic penetration depth data by Bonalde *et al.*,[12,27] they have not attempted to calculate the magnetothermal conductivity which should be more revealing. Note that the *f*-wave superconductor associated with α and β bands in Ref. 26 is similar to Eq. (2), but not the same. The model with $\hat{\Delta}_{zr}$ produced much larger ($\sim$30 times) $\cos(2\phi)$ term in the angular dependence of magnetothermal conductivity. The better test of the nodal position can be done through the angle (azimuthal angle of $\mathbf{k}$ from the *a* axis) dependence of the thermal conductivity.[17,28]

More recently, Deguchi *et al.* observed a double transition in the specific heat with the magnetic field near H_{c2}, and this experimental result was interpreted in terms of multigap model.[29] On the other hand, the behavior of the specific heat and the magnetic penetration depth for low temperature ($T\ll T_c$) and the low field ($H\ll H_{c2}$) appears to be consistent with the *f*-wave order parameter given in Eq. (2). At the moment, it is not clear whether the order parameter of Eq. (2), or the multigap model with the order parameters of Eqs. (1) and (3) is adequate. This is the fundamental issue for Sr_2RuO_4.

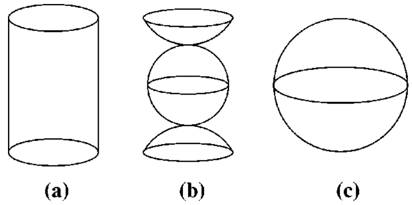

FIG. 1. Order parameters with the cylindrical Fermi surface for (a) p wave, (b) f wave of Eq. (2), (c) f wave of Eq. (3).

In this paper, we study the Raman spectra of spin triplet superconductors in Sr_2RuO_4. Since the Raman spectra S_i in superconductors measure effective density fluctuations, and its strength of the scattering is determined by the Raman vertex, γ_i, the clapping mode can couple to the Raman fluctuations.[4,9,30] We consider three different order parameters given in Eqs. (1), (2), and (3) sketched in Fig. 1: (a) p wave with a full gap, (b) f wave with horizontal nodes, and (c) f wave with node at the zone boundary. We identify the clapping mode[4] in these superconductors and consider its effect on Raman spectra. We show that (1) the Raman spectroscopy can detect the clapping mode and (2) it can discriminate the multigap model from the single gap model. For the p-wave superconductor, the frequency of the clapping mode was found at $\sqrt{2}\Delta(T)$,[9] and this mode exists as a sharp resonance in B_{1g} and B_{2g} modes of Raman spectra. For the f-wave superconductors [Eqs. (2) and (3)], the frequency of the clapping mode $\Delta(T)$ gets smaller than that of the p wave, as we expect due to the existence of node, and this mode is also detectable in B_{1g} and B_{2g} modes of Raman spectra. While the weighted sum of the clapping mode contributions from the p wave and f wave appears in the multigap model, the single gap model of Eq. (2) has the contribution from solely the clapping mode in the f-wave superconductor.

The paper is organized as follows. The formalism to identify the clapping mode and its effect on the Raman spectra is summarized in Sec. II. The results of the Raman spectra of B_{1g} and B_{2g} modes for p-wave and f-wave superconductors are presented in Secs. III and IV, respectively. The conclusion and discussion will be in the Sec. V.

II. FORMALISM OF RAMAN SPECTRA AND CLAPPING MODE

The electronic Raman scattering in superconductors are well described in Ref. 30. Therefore, here we give a brief summary of the Raman spectra. One can select the Raman vertex which allows for different projections on the Fermi surface. The intensity of the each Raman mode provides information on the gap structure along the Fermi surface. The Raman spectra in superconductors is determined from

$$S_i^0(\omega,\mathbf{q}\to 0)=\mathrm{Im}\left[\langle\gamma_i,\gamma_i\rangle-\frac{\langle\gamma_i,1\rangle^2}{\langle 1,1\rangle}\right]. \tag{4}$$

Here we use the following notational convenience:

$$\langle A,B\rangle=T\sum_n\sum_{\mathbf{p}}\mathrm{Tr}[A\rho_3G(\mathbf{p},\omega_n)B\rho_3G(\mathbf{p}-\mathbf{q},i\omega_n-i\omega_\nu)], \tag{5}$$

where the single particle Green's function $G(i\omega_n,\mathbf{k})$ in Nambu space is given by

$$G^{-1}(i\omega_n,\mathbf{k})=i\omega_n-\xi_{\mathbf{k}}\rho_3-\Delta(\hat{k}\cdot\hat{\rho})\sigma_1. \tag{6}$$

Here ρ_i and σ_i are Pauli matrices acting on the particle-hole and spin space, respectively. $\omega_n=(2n+1)\pi T$ is the fermionic Matsubara frequency and $\xi_{\mathbf{k}}=(k_x^2+k_y^2)/2m-\mu$, where μ is the chemical potential. The Raman vertics, B_{1g} and B_{2g} are written as

$$\gamma_{B1g}=\sqrt{2}\cos(2\phi),\quad \gamma_{B2g}=\sqrt{2}\sin(2\phi), \tag{7}$$

where ϕ is the angle of the wave vector $\mathbf{k}$ on the Fermi surface. The second term of Eq. (4) is a back-flow term due to the charge conservation, which can be seen in the limit of $\gamma_i=$const. In this case, Raman intensity should vanishes because there is no density fluctuation in the homogeneous limit of $\mathbf{q}\to 0$ in a superconductor.

The clapping mode is the fluctuation of the order parameter which can be written as $\delta\Delta\rho_3\sim\exp(\pm 2i\phi)(\sigma_1\pm i\sigma_2)\rho_3$ in the Nambu space, As it is indicated in the fluctuation of the order parameter $\delta\Delta\propto\exp(\pm 2i\phi)$, this mode can directly couple to B_{1g} and B_{2g} channels of the Raman spectra. The clapping mode makes the additional contribution to the Raman intensity given by

$$S_i=S_i^0+\mathrm{Im}\left(\frac{\langle\gamma_i,\delta\Delta\rangle\langle\delta\Delta,\gamma_i\rangle}{g^{-1}-\langle\delta\Delta,\delta\Delta\rangle}\right), \tag{8}$$

where g is the coupling constant which mediates superconducting state and the coupling between the fluctuation of the order parameter and the light scattering with Raman vertex γ_i. On the other hand, the A_{1g} mode does not couple to the clapping mode. Therefore $S_{A_{1g}}$ is given by the first term in Eq. (8). Note that S_i^0 is the same for all three modes (B_{1g}, B_{2g}, and A_{1g}) for each order parameter. This indicates the existence of the axial symmetry and horizontal nodes in the order parameter, if there is any.

III. RAMAN SPECTRA OF B_{1g} AND B_{2g} IN P-WAVE SUPERCONDUCTOR

The order parameter of Eq. (1) has a full gap on the Fermi surface, therefore the bare Raman intensity in the p-wave superconductor is same as that of the s-wave superconductor:

$$S_{B_{1g}}^0=\mathrm{Im}\langle\cos(2\phi),\cos(2\phi)\rangle=\frac{2\pi N(0)\Delta^2}{\omega\sqrt{\omega^2-4\Delta^2}}\theta(\omega^2-4\Delta^2). \tag{9}$$

Note that the Raman intensity is zero for the frequency, $\omega<2\Delta$ due to the presence of the full gap.

The coupling to the collective mode leads to the additional contribution to the Raman spectra. While the A_{1g} mode has the same frequency dependences as B_{1g} and B_{2g} as

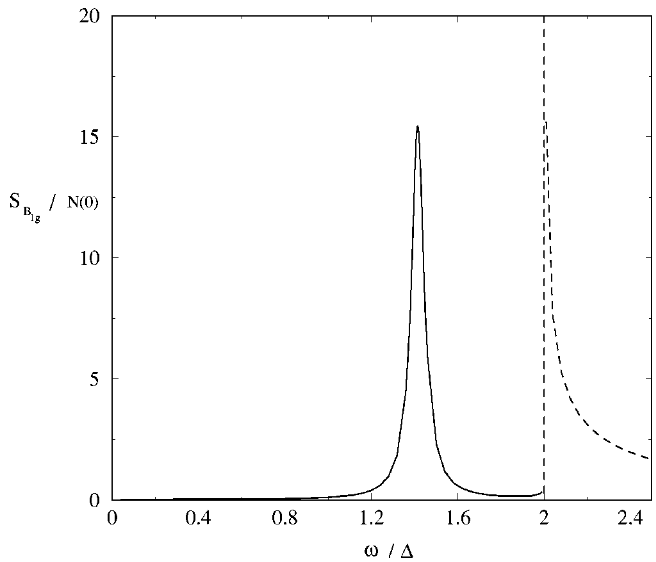

FIG. 2. Raman intensity for p wave with a finite impurity scattering $\Gamma = 0.1\Delta$. The solid line is the contribution from the clapping mode, the dotted line is the bare Raman intensity.

far as the bare Raman intensity, S^0 is concerned, this mode does not couple to the clapping mode. The Raman intensity due to the clapping mode can be obtained by computing the correlation functions, Eq. (8). These are obtained as,

$$\mathrm{Re}\langle \delta\Delta, \gamma_{B_{1g}} \rangle = N(0)[\omega \Delta f],$$

$$\mathrm{Re}\langle \delta\Delta, \delta\Delta \rangle = g^{-1} - N(0)\left[\left(\frac{\omega^2}{2} - \Delta^2\right) f\right], \tag{10}$$

where f is given by

$$f(\omega,T) = \int_\Delta^\infty dE \frac{\tanh(E/2T)}{\sqrt{E^2-\Delta^2}} \frac{1}{4E^2-\omega^2} \tag{11}$$

and

$$g^{-1} = \frac{N(0)}{2} \int_\Delta^\infty \frac{dE}{\sqrt{E^2-\Delta^2}} \tanh\frac{E}{2T}. \tag{12}$$

Since the imaginary part of $\langle \delta\Delta, \cos(2\phi) \rangle$ and $\langle \delta\Delta, \delta\Delta \rangle$ are zero for the frequency, $\omega < 2\Delta$, the clapping mode appears as a resonance in the Raman spectra for B_{1g}, which is given by

$$S_{B_{1g}}^{\mathrm{clapp}}(\omega) = \mathrm{Im}\left(\frac{\langle \gamma_{B_{1g}}, \delta\Delta \rangle \langle \delta\Delta, \gamma_{B_{1g}} \rangle}{g^{-1} - \langle \delta\Delta, \delta\Delta \rangle} \right)$$
$$= 2\pi N(0) \omega^2 \Delta^2 f(\omega,T) \delta(\omega^2 - 2\Delta^2). \tag{13}$$

The Raman intensity $B_{1g}(\omega)$ is plotted in Fig. 2 with a finite impurity scattering $\Gamma = 0.1\Delta$. It is important to note that there is a resonance at the frequency of $\omega = \sqrt{2}\Delta$ with a Lorentzian shape. The same is true for B_{2g} channel.

IV. RAMAN SPECTRA OF B_{1g} AND B_{2g} IN f-WAVE SUPERCONDUCTOR

We consider the f-wave order parameter of Eq. (2), and the cylindrical Fermi surface independent of k_z. Since there is a node along the k_z direction, we expect the finite bare Raman intensity, $\langle \gamma_i, \gamma_i \rangle$ at low frequency ($\omega < 2\Delta$),

$$S_{B_{1g}}^0 = \mathrm{Im}\langle \cos(2\phi), \cos(2\phi) \rangle$$
$$= \frac{4N(0)\Delta}{\omega}\left[K\left(\frac{\omega}{2\Delta}\right) - E\left(\frac{\omega}{2\Delta}\right)\right] \tanh\frac{\omega}{4T}, \tag{14}$$

where K and E are complete elliptic integral of first and second kinds, respectively.

The contribution from the coupling to the clapping mode is given by

$$S_{B_{1g}}^{\mathrm{clapp}} = N(0)\omega^2\Delta^2 \frac{4\omega_c^R \omega_c^I D(\omega,T)}{(\omega^2 - \omega_C^R)^2 + 4(\omega_c^R \omega_c^I)^2}, \tag{15}$$

where

$$D(\omega,T) = \frac{2}{\pi}\int_0^\infty \frac{dE}{4E^2-\omega^2} \frac{1}{\Delta}\left[K\left(\frac{E}{\Delta}\right) - E\left(\frac{E}{\Delta}\right)\right] \tanh\frac{E}{2T}. \tag{16}$$

The frequency of the clapping mode and its damping are obtained as

$$\omega_c^R = \Delta, \quad \omega_c^I = 0.57\Delta, \tag{17}$$

where $\omega_c^I = \mathrm{Im}\langle \delta\Delta, \delta\Delta \rangle / N(0)\omega_c^R D(\omega,T)$ and $\mathrm{Im}\langle \delta\Delta, \delta\Delta \rangle$ is given by

$$\mathrm{Im}\langle \delta\Delta, \delta\Delta \rangle = \frac{N(0)}{\omega}\left(\frac{\omega^2}{\Delta}[K(k) - E(k)] - \frac{2\Delta}{3}[(k^2+2)K(k) - 2(k^2+1)E(k)] \right) \tanh\frac{\omega}{4T}, \tag{18}$$

where $k = \omega/2\Delta$. The Raman intensity due the clapping mode is shown in Fig. 3. It is important to note that the shape of the Raman intensity is not Lorentzian, but it has asymmetry. The Raman spectra for the another proposed f-wave, Eq. (3) are identical to the result presented here for the order parameter of Eq. (2).

V. CONCLUSION

Within the framework of the weak-coupling BCS theory, we have studied the Raman spectra of two-dimensional p wave [Eq. (1)] and f wave [Eqs. (2) and (3)] superconductors; the candidate for Sr_2RuO_4. The clapping mode with angular momentum ± 2, parallel to the c axis is common to these ground states, which break the chiral symmetry. We have shown that the clapping mode has the frequency of $\sqrt{2}\Delta(T)$, and $\Delta(T)$ for p- and f-wave superconductors, respectively. We have also shown that the clapping mode couples both sound wave,[9] and B_{1g} and B_{2g} modes of the Raman spectra. While the coupling to the sound wave is very

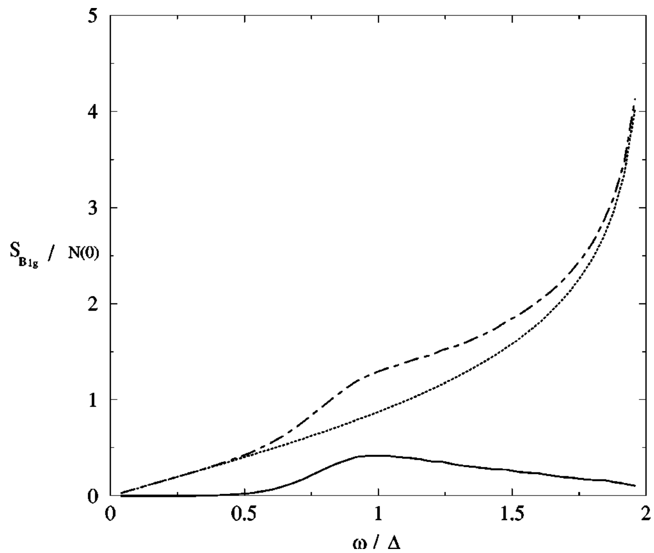

FIG. 3. Raman intensity for *f* wave. The solid line is the contribution from the clapping mode, the dotted line is the bare Raman intensity, and the dashed line is the total Raman intensity.

small, the clapping mode appears as a huge resonance in Raman spectra. Therefore, if the present experimental difficulty is overcome, the Raman spectroscopy provides a unique window to probe the clapping mode.

Investigating the clapping mode in Raman spectra will also probe the order parameter of Sr_2RuO_4. The single-gap model of the *f* wave shows only contribution from the clapping mode of *f*-wave order parameter (Fig. 3). On the other hand, the multigap model proposed by Zhitomirsky and Rice should be the weighted sum of the *p* wave (Fig. 2) and *f* wave (Fig. 3) contribution. It is important to note that the maximum gap of $\Delta_{zr} \approx (0.2-0.5)\Delta_{BCS}$ on the α and β bands, while the maximum of Δ on γ band with *p* wave is set to be Δ_{BCS} which is the energy scale we used in our figures. Therefore, for the multigap model, the contribution from *f*-wave order parameter, Eq. (3) should be peaked around $(0.2-0.5)\Delta$ (with the contribution from *p* wave around $\sqrt{2}\Delta$), while single gap model gives the peak around Δ. The Raman intensity of the *f*-wave cases are rather small, and it would be practically difficult to distinguish from its background. On the other hand, the intensity of the *p*-wave case appears as a huge resonance well separated from its bare intensity. Therefore, the Raman spectroscopy can discriminate the multigap model from the single gap model.

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[1] Y. Maeno *et al.*, Nature (London) **372**, 532 (1994).
[2] T.M. Rice and M. Sigrist, J. Phys.: Condens. Matter **7**, L643 (1995).
[3] Y. Maeno, T.M. Rice, and M. Sigrist, Phys. Today **54**, 42 (2001).
[4] *The Superfluid Phases of Helium 3*, edited by D. Vollhardt and P. Wölfle (Tayor & Francis, New York, 1990).
[5] K. Ishida *et al.*, Nature (London) **396**, 658 (1998); J.A. Duffy *et al.*, Phys. Rev. Lett. **85**, 5412 (2000).
[6] G.M. Luke *et al.*, Nature (London) **394**, 558 (1998).
[7] L. Tewordt, Phys. Rev. Lett. **83**, 1007 (1999).
[8] H.-Y. Kee, Y.B. Kim, and K. Maki, Phys. Rev. B **61**, 3584 (2000).
[9] H.-Y. Kee, Y.B. Kim, and K. Maki, Phys. Rev. B **62**, 5877 (2000); S. Higashitani and K. Nagai, *ibid.* **62**, 3042 (2000); D. Fay and L. Tewordt, Phys. Rev. B **62**, 4036 (2000) incorrectly referred the paper (Ref. 9) by Kee *et al*., saying that the coupling of the clapping mode to charge density vanishes. However, one should note that the coupling of the clapping mode to the sound is not the coupling to the *uniform* charge density.
[10] A.P. Mackenzie *et al.*, Phys. Rev. Lett. **80**, 161 (1998).
[11] S. Nishizaki, Y. Maeno, and Z.Q. Mao, J. Phys. Soc. Jpn. **69**, 572 (2000).
[12] I. Bonalde *et al.*, Phys. Rev. Lett. **85**, 4775 (2000).
[13] K. Ishida *et al.*, Phys. Rev. Lett. **84**, 5387 (2000).
[14] C. Lupien *et al.*, Phys. Rev. Lett. **86**, 5986 (2001).
[15] Y. Hasegawa *et al.*, J. Phys. Soc. Jpn. **69**, 336 (2000).
[16] H. Won and K. Maki, Europhys. Lett. **52**, 427 (2000).
[17] T. Dahm, K. Maki, and H. Won, cond-mat/0006301 (unpublished).
[18] K. Miyake and O. Narikiyo, Phys. Rev. Lett. **83**, 1423 (1999).
[19] M.J. Graf and A.V. Balatsky, Phys. Rev. B **62**, 9697 (2000).
[20] M. Sigrist *et al.*, Physica C **317**, 134 (1999).
[21] M. Sato and M. Kohmoto, J. Phys. Soc. Jpn. **69**, 3505 (2000).
[22] H. Won and K. Maki, cond-mat/0004105 (unpublished).
[23] H. Won and K. Maki, Curr. Appl. Phys. **1**, 291 (2001).
[24] M.A. Tanatar *et al.*, Phys. Rev. Lett. **86**, 2649 (2001).
[25] K. Izawa *et al.*, Phys. Rev. Lett. **86**, 2653 (2001).
[26] M.E. Zhitomirsky, and T.M. Rice, Phys. Rev. Lett. **87**, 057001 (2001).
[27] H. Kusunose and M. Sigrist, Europhys. Lett. **60**, 28 (2002).
[28] H. Won and K. Maki, Physica B **312-313**, 44 (2002).
[29] K. Deguchi *et al.*, cond-mat/0210537 (unpublished).
[30] T.P. Devereaux and D. Einzel, Phys. Rev. B **51**, 16 336 (1995).