Raman Spectra of Triplet Superconductor in Sr$_2$RuO$_4$

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We study the Raman spectra of spin-triplet superconductors in Sr$_2$RuO$_4$. 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 $1.02\Delta(T)$ for $p$-wave and $f$-wave superconductors, respectively. This mode is visible as a huge resonance in the B1g and B2g 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$_2$RuO$_4$ 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.$^2,3$

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

(1)

where $\Delta$ is the magnitude of the superconducting order parameter. Here $\hat{d}$-vector is called spin vector perpendicular to the direction of the spin associated with the condensed pair.$^1$ 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 $\mu$SR experiment.$^2$ 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 Ref. 3 even though the thermal conductivity data cannot exclude a small admixture, a few $\%$ of $p$-wave order parameter, Eq. (1).

$$\Delta(k) = \Delta\hat{d}(k_x \pm ik_y)\cos(k_z),$$

(2)

where $c$ is the lattice constant along the $c$-axis.

Parallel to this development, Zhitomirsky and Rice$^2$ have proposed an alternative model, multi-gap model for Sr$_2$RuO$_4$. As it is known that the Fermi surfaces in Sr$_2$RuO$_4$ consist of three different bands labeled by $\alpha$-, $\beta$-, and $\gamma$-bands,$^2$ it is also believed that the superconductivity arises mainly in the $\gamma$-band. It was proposed that a full gap with $p$-wave, Eq. (1) exists in the active band $\gamma$, while line nodes with $f$-wave order parameter, Eq. (2) develops in the $\alpha$ and $\beta$ due to proximity effect.$^2$

$$\Delta_{\gamma\gamma}(k) = \Delta\hat{d}(k_x \pm ik_y)\cos(k_z/2).$$

(3)

While this model could reproduce the specific heat data by Nishizaki et al$^1$, and the magnetic penetration depth data by Bonalde et al$^2,27$, they have not attempted to calculate the magneto-thermal conductivity which should be more revealing. Note that the $f$-wave superconductor associated with $\alpha$ and $\beta$-bands in Ref. 24 is similar to the Eq. 2 but not the same. The model with $\Delta_{\gamma\gamma}$ produced much larger ($\sim 30$ times) $\cos(2\phi)$ term in the angular dependence of magneto-thermal conductivity. The better test of the nodal position can be done through the angle (azimuthal angle of $k$ from the $a$-axis) dependence of the thermal conductivity.$^1,28$

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.$^28$ 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. \(3\). At the moment, it is not clear whether the order parameter of Eq. \(2\) or the multigap model with the order parameters of Eq. \(1\) and Eq. \(3\) is adequate. This is the fundamental issue for \(\text{Sr}_2\text{RuO}_4\).

In this paper, we study the Raman spectra of spin triplet superconductors in \(\text{Sr}_2\text{RuO}_4\). We consider three different order parameters given in Eq. \(1\), \(2\), 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 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 \(\sqrt{2}\Delta(T)\); 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 gets smaller than that of the p-wave, 1.02\(\Delta\) as expected 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 the 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 the section II. The results of the Raman spectra of \(B_{1g}\) and \(B_{2g}\) modes for p-wave and f-wave superconductors are presented in the section III and IV, respectively. The conclusion and discussion will be followed in the section V. The details of computing the correlation functions are presented in Appendix.

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. The Raman spectra \(S_i\) in superconductors measure effective density fluctuations, and its strength of the scattering is determined by the Raman vertex, \(\gamma_i\). 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

\[
S^{\gamma_i}_0(\omega, q \to 0) = \text{Im} \left( \langle \gamma_i, \gamma_i \rangle - \frac{\langle \gamma_i, 1 \rangle \langle 1, \gamma_i \rangle}{1} \right). \tag{4}
\]

Here we use the following notational convenience,

\[
\langle A, B \rangle = T \sum_n \sum_p \text{Tr}[A \rho_3 G(p, \omega_n) B \rho_3 G(p-q, \omega_n-i\omega_p)], \tag{5}
\]

where the single particle Green’s function, \(G(i\omega_n, k)\), in the Nambu space is given by

\[
G^{-1}(i\omega_n, k) = i\omega_n - \xi_k \rho_3 - \Delta(k \cdot \hat{p}) \sigma_1. \tag{6}
\]

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

\[
\gamma_{B_{1g}} = \sqrt{2} \cos(2\phi) \quad \gamma_{B_{2g}} = \sqrt{2} \sin(2\phi), \tag{7}
\]

where \(\phi\) is the angle of the wave-vector \(k\) on the Fermi surface. The second term of Eq. \(2\) is a back-flow term due to the charge conservation, which can be seen in the limit of \(\gamma_i = \text{const}\). In this case, Raman intensity should vanishes because there is no density fluctuation in the homogeneous limit of \(q \to 0\) 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

\[
S_i = S^{\gamma_i}_0 + \text{Im} \left( \frac{\langle \gamma_i, \delta \Delta \rangle \langle \delta \Delta, \gamma_i \rangle}{g^{-1} - \langle \delta \Delta, \delta \Delta \rangle} \right), \tag{8}
\]

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, \(\gamma_i\). 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 \(S^{\gamma_i}_0\) is the same for all three modes \((B_{1g}, B_{2g}, A_{1g})\) 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 p-wave superconductor is same as that of s-wave superconductor.

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

(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 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 presented in the Appendix. Here we summarize the result of the correlation functions.

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

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

where $f$ is given by

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

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

$$S_{B_{1g}}(\omega) = Im\left( \frac{\langle \gamma_{B_{1g}}, \delta \Delta \rangle \langle \delta \Delta, \gamma_{B_{1g}} \rangle}{g^{-1} - \langle \delta \Delta, \delta \Delta \rangle} \right) = 2\pi N(0)\omega^2 \Delta^2 f(\omega, T) \delta(\omega^2 - 2\Delta^2). \quad (12)$$

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.

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 with $K$ and $E$ are complete Elliptical integral of first and second kinds, respectively.

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

$$S_{B_{1g}}^{clapp} = N(0)\omega^2 \Delta^2 \frac{4\omega R^2 \omega c^T D(\omega, T)}{(\omega^2 - \omega R^2)^2 + 4(\omega R \omega c^T)^2}, \quad (14)$$

where

$$D(\omega, T) = \frac{2}{\pi} \int_{0}^{\infty} \frac{dE}{4E^2 - \omega^2} \frac{1}{\Delta} \left( K(\frac{E}{\Delta}) - E(\frac{E}{\Delta}) \right) \tanh \frac{E}{2T} \quad (15)$$

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

$$\omega_c^R = 1.02\Delta, \quad \omega_c^I = 0.57\Delta. \quad (16)$$

where the $\omega_c^I$ is computed from the formula, $\text{Im} \langle \delta \Delta, \delta \Delta \rangle / N(0)\omega^2 D(\omega, T)$, and $\text{Im} \langle \delta \Delta, \delta \Delta \rangle$ is given in the Appendix, Eq. 10. 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.

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,

$$S^0_{B_{1g}} = Im\langle \cos(2\phi), \cos(2\phi) \rangle = \frac{4N(0)\Delta}{\omega} \left[ K(\frac{\omega}{2\Delta}) - E(\frac{\omega}{2\Delta}) \right] \tanh \frac{\omega}{4T}, \quad (13)$$

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$_2$RuO$_4$. The clapping mode with angular momentum $\pm 2$, parallel to the
Therefore, for the multigap model, the contribution from $\Delta_{\text{BCS}}$ is important to note that the maximum gap of the p-wave (Fig. 2) and f-wave (Fig. 3) contributions. It is also shown that the clapping mode couples both sound wave[8] and $B_{1g}$ and $B_{2g}$ modes of the Raman spectra. While the coupling to the sound wave is very 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$_2$RuO$_4$. The single 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 contributions. It is important to note that the maximum gap of $\Delta_{2\sigma}\approx (0.2 \sim 0.5)\Delta_{\text{BCS}}$ on the $\alpha$- and $\beta$-bands, while the maximum of $\Delta$ on $\gamma$-band with p-wave is set to be $\Delta_{\text{BCS}}$ 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 $(0.2 \sim 0.5)\Delta$ (with the contribution from p-wave around $\sqrt{2}\Delta$), while single gap model gives the peak around $1.02\Delta$. The Raman spectroscopy can discriminate the multigap model from the single gap model.

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APPENDIX A

1. the clapping mode and its contribution on Raman, $B_{1g, 2g}$ in p-wave superconductor

The fluctuation of the order parameter corresponds to the clapping mode, and it can be written as $\delta \Delta \rho_3 \sim e^{\pm \Delta} (\sigma_1 \pm i \sigma_2) \rho_3$. Its coupling to the $\gamma_{B1g}$ mode of Raman spectra, and its propagator, $\langle \delta \Delta, \delta \Delta \rangle$ are obtained by computing the following correlation function within weak coupling theory.

$$\langle \delta \Delta, \cos(2\phi) \rangle(i\omega_{\nu}, q) = T \sum_{n} \sum_{p} \text{Tr}[\delta \Delta \rho_3 G(p, \omega_n) \cos(2\phi) \times \rho_3 G(p - q, i\omega_n - i\omega_{\nu})],$$

$$\langle \delta \Delta, \delta \Delta \rangle(i\omega_{\nu}) = T \sum_{n} \sum_{p} \text{Tr}[\delta \Delta \rho_3 G(p, \omega_n) \delta \Delta \times \rho_3 G(p - q, i\omega_n - i\omega_{\nu})]. \quad (A1)$$

After summing over the frequency, the correlation functions are written as

$$\text{Re}\langle \delta \Delta, \gamma_{B1g} \rangle = N(0) \int_{\Delta} d\omega_{\nu} \frac{\omega \Delta}{\sqrt{E^2 - \Delta^2}} \frac{1}{4E^2 - \omega^2} \tanh \frac{E}{2T} = [N(0)\omega \Delta f], \quad (A2)$$

where $f(\omega, T)$ is given by Eq. 1. There is no imaginary part of the above correlation function. On the other hand, the correlation function of the order parameter fluctuation is given by

$$\text{Re}\langle \delta \Delta, \delta \Delta \rangle = N(0) \int_{\Delta} d\omega \frac{2E^2 - \Delta^2}{\sqrt{E^2 - \Delta^2}} \frac{1}{4E^2 - \omega^2} \tanh \frac{E}{2T} = g^{-1} - \left[ \left( \frac{\omega^2 - \Delta^2}{2} \right) f \right], \quad (A3)$$

where

$$g^{-1} = \frac{N(0)}{2} \int_{\Delta} d\omega \frac{1}{\sqrt{E^2 - \Delta^2}} \tanh \frac{E}{2T} \quad (A4)$$

Now the imaginary part of the above correlation function is

$$\text{Im}\langle \delta \Delta, \delta \Delta \rangle = -N(0)\frac{\pi}{2\omega} \int_{\Delta} d\omega \frac{E^2 - \Delta^2}{\sqrt{E^2 - \Delta^2}} \tanh \frac{E}{2T} \times (\delta(\omega - 2E) + \delta(\omega + 2E)) = N(0)\frac{\pi}{2\omega} \frac{\omega^2 - 2\Delta^2}{\sqrt{\omega^2 - 4\Delta^2}} \tanh \frac{\omega}{4T} \theta(\omega - 2\Delta). \quad (A5)$$

Note that $\text{Im}\langle \delta \Delta, \delta \Delta \rangle$ is 0 for $\omega < 2\Delta$, which is the regime we are interested in. Therefore, the Raman intensity for $B_{1g}$ as well as $B_{2g}$ for p-wave for $\omega < 2\Delta$ should be

$$\text{Im} \left( \frac{[\gamma, \delta \Delta \rho_3]}{g^{-1} - [\delta \Delta \rho_3, \delta \Delta \rho_3]} \right) = 2\pi N(0)\omega^2 \Delta^2 f \left[ \frac{\omega^2 - 2\Delta^2}{\omega^2 - 4\Delta^2} + i\Gamma \right] = 2\pi N(0)\omega^2 \Delta^2 f \delta(\omega^2 - 2\Delta^2), \quad (A6)$$
where a finite $\Gamma$ could come from the impurity scattering. The clapping mode appears as a huge resonance with Lorentzian shape (with a finite $\Gamma$) at the frequency of $\sqrt{2}\Delta$ shown in Fig. 2.

2. the clapping mode and its contribution on Raman, $B_{1g,2g}$ in f-wave superconductor

Using the correlation functions, Eq. [A1], we obtained the following results for the f-wave superconductor.

$$\text{Re}\langle \delta \Delta, \cos (2\phi) \rangle = \frac{2N(0)\omega D}{\pi} \int_0^\infty \frac{dE}{\omega^2 - 4E^2} \times \left( A\left( \frac{E}{\Delta} \right) \right) \tanh \frac{E}{2T}. \quad (A7)$$

Using the definition of $D(\omega, T)$, Eq. [13], the above correlation function can be written as

$$\text{Re}\langle \delta \Delta, \cos (2\phi) \rangle = N(0) [\omega \Delta D(\omega, T)] \quad (A8)$$

On the other hand, the propagator of the clapping mode is given by

$$\text{Re}\langle \delta \Delta, \delta \Delta \rangle = g^{-1} - \frac{2N(0)}{\pi} \int_0^\infty \frac{dE}{4E^2 - \omega^2} \times \left( \frac{\omega^2}{2} A\left( \frac{E}{\Delta} \right) - \Delta^2 B\left( \frac{E}{\Delta} \right) \right) \tanh \frac{E}{2T}$$

$$\times \tanh \frac{\omega}{4T}, \quad (A9)$$

where

$$A(k) = \frac{1}{\Delta} (K(k) - E(k))$$

$$B(k) = \frac{1}{3\Delta^2} \left[ (k^2 + 2)K(k) - 2(k^2 + 1)E(k) \right]$$

$$\omega_c = 2\Delta^2 B(k)/A(k). \quad (A10)$$

Therefore, one can determine the position of the clapping mode, $\omega_c^R$, using the following relations.

$$2k^2 = \frac{1}{3} \left( 2 + \frac{k^2K(k) - 2k^2E(k)}{K(k) - E(k)} \right), \quad (A11)$$

where $k = \omega_c^R/(2\Delta)$. This gives

$$\omega_c^R = 1.02\Delta. \quad (A12)$$

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