Zero-bias conductance peak splitting due to multiband effect in tunneling spectroscopy

Y. Tanuma, K. Kuroki, Y. Tanaka, and S. Kashiwaya

1 Institute of Physics, Kanagawa University, Rokkakubashi, Yokohama, 221-8686, Japan
2 Department of Applied Physics and Chemistry, The University of Electro-Communications, Chofu, Tokyo 182-8585, Japan
3 Department of Applied Physics, Nagoya University, Nagoya, 464-8603, Japan
4 National Institute of Advanced Industrial Science and Technology, Tsukuba, 305-8568, Japan
(Dated: November 2, 2018)

We study how the multiplicity of the Fermi surface affects the zero-bias peak in conductance spectra of tunneling spectroscopy. As case studies, we consider models for organic superconductors \( \kappa-(BEDT-TTF)_2\text{Cu(NCS)}_2 \) and \( \text{(TMTSF)}_2\text{ClO}_4 \). We find that multiplicity of the Fermi surfaces can lead to a splitting of the zero-bias conductance peak (ZBCP). We propose that the presence/absence of the ZBCP splitting is used as a probe to distinguish the pairing symmetry in \( \kappa-(BEDT-TTF)_2\text{Cu(NCS)}_2 \).

I. INTRODUCTION

An unambiguous determination of pairing symmetry in unconventional superconductors is crucial to understand the pairing mechanism of superconductivity. Strong evidences suggesting \( d_{x^2-y^2} \)-wave pairing symmetry in the high-\( T_c \) cuprates have been provided using several phase-sensitive probes\(^{12,13,14,15,16,17,18,19,20,21,22,23,24,25,26,27,28,29,30,31,32,33} \) including tunneling spectroscopy via Andreev surface bound states (ABS’s).\(^{10,11,12} \) The tunneling spectroscopy via ABS’s enables us to detect the sign change in the pair potential as well as its nodal structure.\(^{8,8,8} \) This state, which originates from the interference effect in the effective pair potential of the \( d_{x^2-y^2} \)-wave symmetry through reflection at a surface or an interface, have significant influences on several charge transport properties.\(^{10,11,12} \) The existence of ABS’s, which manifests itself as a distinct conductance peak at zero-bias in the tunneling spectrum (zero-bias conductance peak, referred to as ZBCP), has been actually observed not only in the high-\( T_c \) cuprates\(^{24,25,26,27,28,29,30,31,32,33} \), but also in ruthenates\(^{14,15,16,17} \), heavy fermion systems\(^{18} \), and more recently MgCNi\(_3\).\(^{19,20} \) In this context, it is of great interest to investigate whether the ZBCP due to the ABS’s can be observed in organic superconductors such as \( \kappa-(BEDT-TTF)_2\text{X} \) and \( \text{(TMTSF)}_2\text{X} \).\(^{20,21,22,23} \)

The tunneling spectroscopy via ABS’s can be used to determine the pairing symmetry if one can prepare well-treated surfaces with arbitrary orientations in the superconducting plane. For high-\( T_c \) cuprates, which has a \( d_{x^2-y^2} \)-wave pair potential, it is theoretically shown that the ZBCP should be observed most prominently for the (110) surfaces or interfaces. Moreover, it has been clarified that the ZBCP may be observed due to atomic-scale roughness even in the (100) surfaces.\(^{21,22,23} \) In fact, Iguchi et al.\(^{24} \) have measured the ZBCP for Ag/YBCO ramp-edge junctions with various orientations, where the injection direction varies continuously from (100) to (110) interfaces. The height of the ZBCP has shown to vary according as the misorientation angle from \( a \)-axis within the plane.

As regards organic superconductors such as \( \kappa-(BEDT-TTF)_2\text{X} \), the pairing symmetry of the pair potentials still remains to be a controversial problem. It has indeed become an issue of great interest whether \( \kappa-(BEDT-TTF)_2\text{X} \) has a \( d \)-wave pair potential similar to high-\( T_c \) cuprates. There is now a body of accumulating experimental evidences suggesting that \( \kappa-(BEDT-TTF)_2\text{X} \) have anisotropy in the pair potential.\(^{25,26,27,28,29,30,31,32,33} \) Earlier theories support \( d_{x^2-y^2} \)-wave pairing,\(^{34} \) Concerning this issue, two of the present authors have theoretically shown that a \( d_{xy} \)-like pairing may slightly dominate over \( d_{x^2-y^2} \) pairing when the dimerization of the BEDT-TTF molecules is not so strong.\(^{35} \) According to previous studies\(^{36,37,38} \), if the pairing symmetry of \( \kappa-(BEDT-TTF)_2\text{X} \) is \( d \)-wave, ABS is expected to be created at surfaces for arbitrary injection orientations. However, a scanning tunneling microscopy (STM) experiment for \( \kappa-(BEDT-TTF)_2\text{X} \) by Arai et al.\(^{39} \) showed the absence of ZBCP for arbitrary injection angle from the \( c \)-axis in the \( bc \)-plane, which is in contrast with the case of the high-\( T_c \) cuprates. The presence/absence of the ZBCP of \( d \)-wave superconductors is sensitive to several factors: (i) roughness effect of surfaces or interfaces, (ii) random impurity scattering effect near the interfaces, (iii) the shape of the Fermi surface, and (iv) the degradation of surfaces. The disappearance of the ZBCP in \( d \)-wave superconductors due to reason (i) has been studied previously.\(^{40,41,42,43} \) Depending on the shape of the Fermi surface and the geometry of the surface, the atomic-size wave nature of the zero energy ABS’s (ZES), \( i.e., \) the oscillatory behavior of the wave function of ZES induces an interference effect which locally destroys the ZBCP. In fact, it is by no means easy to make well-oriented cleavage surfaces in organic materials, so this point may be important. As regards point (ii), Asano et al.\(^{44} \) have shown, both from analytical and numerical calculation beyond quasiclassical approximation,\(^{45} \) that impurity scattering near the interface in the high-\( T_c \) cuprates can induce a splitting or a disappearance of the ZBCP.

As for point (iii), we have recently studied the disappearance of ZBCP due to the warping of quasi-1D Fermi
surface as in (TMTSF)$_2$X. The results indicate that the ABS's are sensitive to the shape of the Fermi surface. However, most of the theoretical studies on tunneling spectroscopy via ABS's up to date have been performed for single band systems. It has not been clarified how the multiplicity of the Fermi surface influences the ZBCP.

Motivated by this point, here we investigate the surface density of states in systems having multiple Fermi surfaces, where we focus on two organic superconductors as case studies, namely, $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ and (TMTSF)$_2$ClO$_4$. The Fermi surface of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, which has been determined by Shubnikov-de Haas experiment, consists of two portions separated by small gaps. The Fermi surface of (TMTSF)$_2$ClO$_4$ is also separated by a small gap, which is due to anion ordering. In this paper, we extend our previous studies on anisotropic triangular lattice by taking into account these multiplicity of the Fermi surface.

The organization of the paper is as follows. The formulation of calculating the tunneling spectrum on anisotropic triangular lattice is presented in Sec. II. In Sec. III, results of the numerical calculations are discussed in detail. Finally, we summarize the paper in Sec. IV.

II. FORMULATION

FIG. 1: (a) Schematic of (100) surface in $xy$-plane with next-nearest neighbor hopping $t'$. (b) Cooper pairs in real space for $d_{x^2-y^2}$ and $d_{xy}$-like wave pairings.

In the present study, we start from an extended Hubbard model given by

$$H = -\sum_{i,j,\sigma} t_{ij} c_i^\dagger c_j + \frac{V}{2} \sum_{i,j,\sigma,\sigma'} c_i^\dagger c_j^\dagger c_j c_i,$$

where $c_i^\dagger$ creates a hole with spin $\sigma$ at site $i$ at site $i = (x, y)$. As a model for $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, each site corresponds to BEDT-TTF molecule dimers. We consider five kinds of hopping integrals, $t_x, t_y, t_{x'}$, and $t'$ in the $xy$ plane on the anisotropic triangular lattice as shown in Fig. 1(a). In order to reproduce the shape of Fermi surface for $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ and (TMTSF)$_2$ClO$_4$, we adopt the values of (i) $t' = 0.8t$, $t_y = t_x = t_{x'} = 0.4t$, and (ii) $t_y = 0.1t$. $t_{x'} = t_x$, $t' = t_y$. Two subchains in the $x$ direction alternatively have the site energy $\varepsilon_i = \pm E_g = -E_g, +E_g, \ldots$ in the $y$ direction. The chemical potential $\mu$ is determined such that the band in $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ is half-filled [quarter-filled]. The effective attraction $V$ is assumed to act on a pair of electrons.

By solving the mean-field equation for a unit cell with $N_u (=500)$ sites in the $x$ direction and two sites in the $y$ direction, we obtain the eigenenergy $E_\nu$. In terms of the eigenenergy $E_\nu$ and the wave functions $u_\nu^i, v_\nu^i$, the Bogoliubov-de Gennes equation for the (100) surface in the $xy$ plane is given by

$$\sum_{j} \left( \begin{array}{cc} H_{ij} & F_{ij} \\ F_{ij}^\dagger & -H_{ij} \end{array} \right) \left( \begin{array}{c} u_\nu^j \\ v_\nu^j \end{array} \right) = E_\nu \left( \begin{array}{c} u_\nu^i \\ v_\nu^i \end{array} \right),$$

with

$$H_{ij}(k_y) = -t_x \eta_+ \delta_{j_x,i_x+1} - t_x \eta_- \delta_{j_x,i_x-1} - t_y \eta_+ \delta_{j_y,i_y+1} - t_y \eta_- \delta_{j_y,i_y-1} - t_y' \eta_+ \delta_{j_y,i_y+1} - t_y' \eta_- \delta_{j_y,i_y-1}.$$

where we define $\eta_+ = \frac{1}{2} \{ 1 + (-1)^{i_x+i_y} \}$ and $\eta_- = \frac{1}{2} \{ 1 - (-1)^{i_x+i_y} \}$.

As for plausible pairing symmetries in $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, we consider $d_{x^2-y^2}$-wave pairing given
with the Fermi surfaces. For \( \tau_d \) and \( \kappa \) face is continuously connected. In the actual (TMTSF) \(_2\)ClO\(_4\), the orientational order of the anions ClO\(_4\) doubles the unit cell, leading again to a splitting of the Fermi surface. Although the pairing symmetry for (TMTSF)\(_2\)X remains to be undetermined, here we assume singlet \( d \)-wave pairing as an example in which the multiband effect is prominent. In this case \( d \)-wave is a pairing separated by two lattice spacings given by

\[
F_{ij}(k_y) = \Delta_x \eta_+ \delta_{j_x,i_x} + \Delta_x' \eta_- \delta_{j_x,i_x+1} - \Delta_y e^{-2ik_y \delta_{j_y,i_y}} \eta_+ \delta_{j_y,i_y+1} - \Delta_y' e^{-2ik_y \delta_{j_y,i_y}} \eta_- \delta_{j_y,i_y+1} + \Delta_y e^{2ik_y \delta_{j_y,i_y}} \eta_- \delta_{j_y,i_y-1} - \Delta_y' e^{2ik_y \delta_{j_y,i_y}} \eta_+ \delta_{j_y,i_y-1}.
\]

and \( d \)-wave-like pairings given by

\[
F_{ij}(k_y) = \Delta_x \eta_+ \delta_{j_x,i_x} + \Delta_x' \eta_- \delta_{j_x,i_x+1} + \Delta_y e^{-2ik_y \delta_{j_y,i_y}} \eta_+ \delta_{j_y,i_y+1} + \Delta_y' e^{-2ik_y \delta_{j_y,i_y}} \eta_- \delta_{j_y,i_y+1} + \Delta_y e^{2ik_y \delta_{j_y,i_y}} \eta_- \delta_{j_y,i_y-1} + \Delta_y' e^{2ik_y \delta_{j_y,i_y}} \eta_+ \delta_{j_y,i_y-1} - \alpha \Delta_p e^{2ik_y \delta_{j_y,i_y}} \eta_+ \delta_{j_y,i_y-1} - \alpha \Delta_p e^{-2ik_y \delta_{j_y,i_y}} \eta_- \delta_{j_y,i_y+1},
\]

with \( \alpha = 0.8t \) in accord with Ref. 33. The pairing in real space is shown in Fig. 1(b). Here, we select \( \Delta_x' = \Delta_y \) and \( \Delta_x = \Delta_y' = \Delta_y = \Delta_0 \), where \( \Delta_0 \) is a bulk value.

The upper and lower panels of Fig. 2 show the \( d_{x^2-y^2} \) and \( d_{xy} \)-like pair potentials in momentum space along with the Fermi surfaces. For \( t_{x'} = t_x \), the Fermi surface is continuously connected. In the actual \( \kappa \)-(BEDT-TTF)\(_2\)Cu(NCS)\(_2\), however, BEDT-TTF dimmers are further dimerized so that \( t_{x'} \neq t_x \), which leads to

\[
\rho(E) = \int_{-\infty}^{\infty} d\omega \rho_s(\omega) \text{sech}^2\left(\frac{\omega + E}{2k_B T}\right).
\]

The anion potential \( E_y \) is estimated from experimental measurement of angle dependent magnetoresistance 45.

In order to compare our theory with STM experiments, we assume that the STM tip is metallic with a constant density of states, and that tunneling occurs only to the site nearest to the tip. This has been shown to be valid through the study of tunneling conductance of unconventional superconductors 5. The tunneling conductance spectrum is then given at low temperatures by the normalized surface density of states.

\[
\rho_s(\omega) = \sum_{k_x, \nu} \left| |\nu'|^2 \delta(\omega + E_{\nu}) + |\nu|^2 \delta(\omega + E_{\nu}) \right|.
\]
FIG. 4: Tunneling spectrum for (a) \( d_{x^2-y^2} \) and (b) \( d_{xy} \)-like waves fixed in \( \Delta_{x'} = \Delta_x \).

Here \( \rho_s(\omega) \) denotes the surface density of states for the superconducting state while \( \rho_N(\omega) \) the bulk density of states in the normal state.

III. RESULTS OF IN-PLANE TUNNELING SPECTRUM

In this section, we present the calculation results. First, let us focus on the model for \( \kappa \)-(BEDT-TTF)\(_2\)X. We examine the case of the tunneling spectrum at (100) surface on the \( xy \) plane as shown in Fig. 4. As seen in Fig. 4 in the case of \( t_{x'} = t_x \), where the Fermi surface is elliptical but continuous, there exists a distinct peak at zero energy, which resembles those obtained in previous theories assuming round shape Fermi surface. The ZEP arises because incident and reflected (including oblique incidence) quasiparticles normal to the surface feel opposite signs of the pair potential, which results in a formation of the ABS. If we turn on the multiband effect by letting \( t_{x'} \neq t_x \), the ZEP is found to split into two.

We have further studied the \( t_{x'}/t_x \) dependence of the ZEP splitting. In Fig. 5, the width of the ZEP splitting \( \delta \) is plotted as functions of \( t_{x'}/t_x \) for \( d_{x^2-y^2} \) and \( d_{xy} \)-like pairings. \( \delta \) for the \( d_{xy} \)-like pair potential is almost proportional to \( t_{x'}/t_x \), and larger than that for \( d_{x^2-y^2} \). In the regime of \( t_{x'}/t_x > 0.9 \), in particular, we see no splitting for the \( d_{x^2-y^2} \) pairing. Since \( t_{x'}/t_x \) is estimated to be \( \sim 0.9 \), we may be able to distinguish between \( d_{x^2-y^2} \) and \( d_{xy} \)-like pairings through the presence/absence of ZEP splitting.

We have also performed similar calculation by letting \( \Delta_{x'}/\Delta_x \) deviate from unity, which should be the case this is reminiscent of the ZEP splitting originating from broken time reversal symmetry states.\( ^7,48,49,50\)

FIG. 5: The \( t_{x'}/t_x \) v.s. the zero-energy peak splitting width \( \delta \).

FIG. 6: Tunneling spectrum for various \( \Delta_{x'}/\Delta_x \) in \( t_{x'}/t_x = 0.9 \).
when $t_x$ deviates from $t_x$. The results are plotted in Fig. 6 for various $\Delta x'/\Delta x$ with $t_{x'/x}$ fixed at 0.9. In this case, we observe an overall shift of the splitted ZEP, while the magnitude of the splitting remains unchanged.

Let us now move on to the model for $(\text{TMTSF})_2\text{ClO}_4$. In this model, as the Fermi surface becomes asymmetric with respect to $k_x \rightarrow -k_x$ transformation, some injected and reflected quasiparticles feel different signs and the ZEP appears in the tunneling spectrum. When $E_g$ is turned on, a minigap opens at $k_y = \pm \pi/4$. This effect again leads to ZEP splitting, of which the magnitude increases as $E_g$ is increased. [Fig. 7]

Although it is by no means easy to pinpoint the origin of the ZEP splitting analytically, it can be qualitatively explained as follows. For single band models, the ZEP appears due to the sign change of the pair potential felt by quasiparticles at the interface. In multiband systems, injected and reflected quasiparticles have different band indices, so that an additional phase factor is expected to appear due to interband scattering. This additional phase factor induces the ZEP splitting as in the case of pair potentials with broken time reversal symmetry.

IV. CONCLUSIONS

To summarize, we have investigated the multiband effect on tunneling spectroscopy. As case studies, we have focused on models for $\kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2$ and $(\text{TMTSF})_2\text{ClO}_4$. We find that the multiplicity of the Fermi surface can lead to a splitting of the ZEP. As regards $\kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2$, since $t_{x'/x}$ is estimated to be $\sim 0.9$ we can distinguish between $d_{x^2-y^2}$ and $d_{xy}$-like pairings through the presence/absence of ZEP splitting.

As mentioned in the Introduction, however, a scanning tunneling measurement actually find no ZBCP in the tunneling spectrum of $\kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2$. Since many experiments suggest the existence of nodes in the pair potential in $\kappa-(\text{BEDT-TTF})_2\text{X}$, we believe that the absence of ZBCP is not because the pairing symmetry is a simple $s$-wave, but because of the roughness of the surface or the random scattering effect by impurities near the interface, namely, point (i) or (ii) mentioned in the Introduction. As regards the roughness of the surface, we believe it is necessary to study tunneling spectroscopy of $\kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2$ in the presence of atomic scale roughness as done by Tanuma et al. on a lattice model. As for the issue of random scattering effect by impurities, Asano et al. have shown, both from analytical and numerical calculation beyond quasiclassical approximations, that impurity scattering near the interface in the high-$T_C$ cuprates can induce a splitting or a disappearance of the ZBCP. From this viewpoint, it would also be interesting to study the impurity scattering effect in $\kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2$.

In order to clearly determine the pairing symmetry, other complementary probes should also be used. Recently, we have shown that magnetotunneling spectroscopy is a promising method to identify the detailed paring symmetry of the unconventional superconductor. It would also be interesting to apply this probe to $\kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2$.

It is well known that ABS’s have serious influence on the Josephson current. There are many works on Josephson effect in unconventional superconductors both from theoretical and experimental view point. It is also a future problem to study Josephson effect in $\kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2$ and $(\text{TMTSF})_2\text{ClO}_4$.

Acknowledgments

The computations have been performed at the Supercomputer Center of Yukawa Institute for Theoretical Physics, Kyoto University.
Y. Tanaka, Y. Tanuma, and S. Kashiwaya, Phys. Rev. B 64, 054510 (2001); Y. Tanaka, Yu. V. Nazarov and S. Kashiwaya, Phys. Rev. Lett. 90, 167003 (2003); Y. Tanaka, A. A. Golubov and S. Kashiwaya, Phys. Rev. B 68 (2003); N. Kitaura, H. Itoh, Y. Asano, Y. Tanaka, J. Inoue, Y. Tanuma and S. Kashiwaya, J. Phys. Soc. Jpn. 72, (2003); A. A. Golubov, M. Y. Kupriyanov, Pis’ma Zh. Eksp. Teor. Fiz. 69, 242 (1999); Sov. Phys. JETP Lett. 69, 262 (1999); 67, 478 (1998); Sov. Phys. JETP Lett. 67, 501 (1998); A. Poenicke, Yu. S. Barash, C. Bruder, and V. Istyukov, Phys. Rev. B 59, 7102 (1999); K. Yamada, Y. Nagato, S. Higashitani, and K. Nagai, J. Phys. Soc. Jpn. 65, 1540 (1996); T. Lück, U. Eckern, and A. Shelankov, Phys. Rev. B 63, 064510 (2002).

Y. Tanuma, K. Kuroki, Y. Tanaka, R. Arita, S. Kashiwaya, and H. Aoki, Phys. Rev. B 66, 094507 (2002).

K. Oshima, T. Mori, H. Inokuchi, H. Urayama, H. Yamochi, and G. Saito, Phys. Rev. B 38, R938 (1988).

L. Ducasse, M. Abderrabba, J. Hoarau, M. Pesquer, B. Galliès, and J. Gaultier, J. Phys. C 19, 3805 (1986).

P. M. Grant, J. Phys. (France) 44, C3-847 (1983).

K. Yamaji, J. Phys. Soc. Jpn. 55, 860 (1986).

H. Shimahara, Phys. Rev. B 61, R14936 (2000).

H. Yoshino, A. Oda, T. Sasaki, T. Hanajiri, J. Yamada, S. Nakatsuji, H. Anzai, and K. Murata, J. Phys. Soc. Jpn. 68, 3142 (1999).

J. P. Pouget, G. Shirane, K. Bechgaard, and J. M. Fabre, Phys. Rev. B 27, 5203 (1983).

Y. Tanuma, Y. Tanaka, K. Kuroki, and S. Kashiwaya, Phys. Rev. B 66, 174502 (2002).

M. Matsumoto and H. Shiba, J. Phys. Soc. Jpn. 64, 4867 (1995); J. Phys. Soc. Jpn. 64, 3384 (1995).

Y. Tanuma, Y. Tanaka, and S. Kashiwaya, Phys. Rev. B 64, 214519 (2001).