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

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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 κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ and (TMTSF)$_2$ClO$_4$. We find that the 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 κ-(BEDT-TTF)$_2$Cu(NCS)$_2$.

I. INTRODUCTION

An unambiguous determination of pairing symmetry in unconventional superconductors is crucial to understand the pairing mechanism of superconductivity. Strong evidence suggesting $d_{x^2-y^2}$-wave pairing symmetry in the high-T_c cuprates has been provided using several phase-sensitive probes[1–3] including tunneling spectroscopy via Andreev surface bound states (ABS's).[4–7] The tunneling spectroscopy via ABS's enables us to detect the sign change in the pair potential as well as its nodal structure.[6,8,9] 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.[10–21] The existence of ABS's, which manifests itself as a distinct conductance peak at zero bias in the tunneling spectrum [zero-bias conductance peak (ZBCP)], has been actually observed not only in the high-T_c cuprates[22–28] but also in ruthenates,[29,30] heavy fermion systems,[31] and more recently MgCNi$_3$.[32] In this context, it is of great interest to investigate whether the ZBCP due to the ABS's can be observed in organic superconductors[33] such as κ-(BEDT-TTF)$_2$X and (TMTSF)$_2$X.[35]

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 $d_{x^2-y^2}$ pair potential, it is theoretically shown that the ZBCP should be observed most prominently for (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.[7,36–38] In fact, Iguchi *et al.*[39] 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 the a axis within the plane.

As regards organic superconductors such as κ-(BEDT-TTF)$_2$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 κ-(BEDT-TTF)$_2$X has a d-wave pair potential similar to high-T_c cuprates. There is now a body of accumulating experimental evidences suggesting that κ-(BEDT-TTF)$_2$X have anisotropy in the pair potential.[40–45] Earlier theories support $d_{x^2-y^2}$-wave pairing,[46–48] while a recent thermal conductivity measurement suggests d_{xy}-wave pairing.[49] 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.[50] According to previous studies,[5–7] if the pairing symmetry of κ-(BEDT-TTF)$_2$X is d wave, ABS is expected to be created at surfaces for arbitrary injection orientations. However, a scanning tunneling microscopy (STM) experiment for κ-(BEDT-TTF)$_2$X by Arai *et al.*[51] 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.[36,37] 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.*[52] have shown, both from analytical and numerical calculation beyond quasiclassical approximations,[53–57] 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.[58] 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, κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ and (TMTSF)$_2$ClO$_4$. The Fermi surface of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, which has been determined by the Shubnikov–de Haas experiment,[59] 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[58] 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

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

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

$$+ \sum_{i,\sigma} (\varepsilon_i - \mu) c_{i,\sigma}^\dagger c_{i,\sigma}, \tag{1}$$

where $c_{i,\sigma}^\dagger$ creates a hole with spin $\sigma = \uparrow, \downarrow$ at site $i = (i_x, i_y)$. As a model for κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, each site corresponds to BEDT-TTF molecule dimmers. We consider five kinds of hopping integrals $t_x (=t)$, $t_{x'}$, t_y, $t_{y'}$, 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 κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ and (TMTSF)$_2$ClO$_4$, we adopt the values of (i) $t' = 0.8t$, $t_{y'} = t_x$, $t_{x'} = t_y$ (Ref. 59) and (ii) $t_y = 0.1t$, $t_{x'} = t_x$, $t_{y'} = t_y$,[60–62] respectively. Two subchains in the x direction alternatively have the site energy $\varepsilon_i = +E_g, -E_g$,
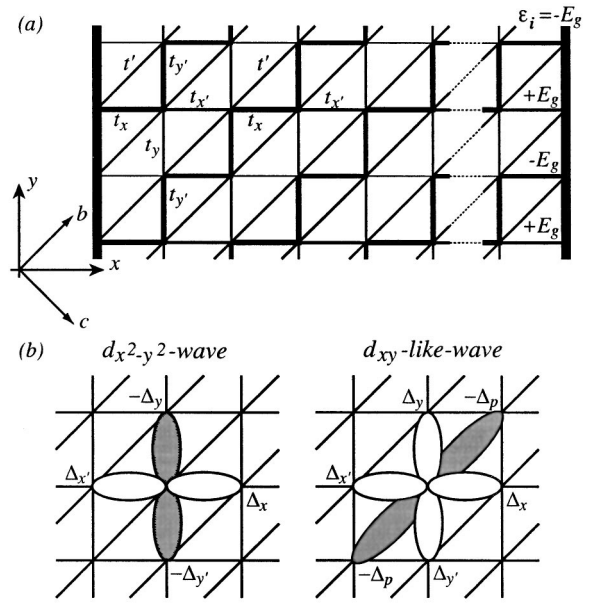

(a)

(b) $d_{x^2-y^2}$-wave d_{xy}-like-wave

FIG. 1. (a) Schematic of the (100) surface in the 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.

$+E_g, \ldots,$ in the y direction.[63,64] The chemical potential μ is determined such that the band in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ [(TMTSF)$_2$ClO$_4$] 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 $N_L(=500)$ sites in the x direction and two sites in the y direction, we obtain the eigenenergy E_ν. In terms of the eigenenergy E_ν and the wave functions u_i^ν, v_i^ν, the Bogoliubov–de Gennes equation for the (100) surface in the xy plane is given by

$$\sum_j \begin{pmatrix} H_{ij} & F_{ij} \\ F_{ij}^* & -H_{ij}^* \end{pmatrix} \begin{pmatrix} u_j^\nu \\ v_j^\nu \end{pmatrix} = E_\nu \begin{pmatrix} u_i^\nu \\ v_i^\nu \end{pmatrix}, \tag{2}$$

with

$$H_{ij}(k_y) = -t_x \eta_+ \delta_{j_x, i_x+1} - t_{x'} \eta_- \delta_{j_x, i_x+1} - t_y e^{-2ik_y \delta_{i_y,2}} \eta_+ \delta_{j_y, i_y+1} - t_{y'} e^{-2ik_y \delta_{i_y,2}} \eta_- \delta_{j_y, i_y+1} - t' e^{-2ik_y \delta_{i_y,2}} \delta_{j_x, i_x+1} \delta_{j_y, i_y+1}$$

$$- t_x \eta_- \delta_{j_x, i_x-1} - t_{x'} \eta_+ \delta_{j_x, i_x-1} - t_y e^{2ik_y \delta_{i_y,1}} \eta_- \delta_{j_y, i_y-1} - t_{y'} e^{2ik_y \delta_{i_y,1}} \eta_+ \delta_{j_y, i_y-1} - t' e^{2ik_y \delta_{i_y,1}} \delta_{j_x, i_x-1} \delta_{j_y, i_y-1}$$

$$+ \{(-1)^{i_y+1} E_g - \mu\} \delta_{i_x, j_x} \delta_{i_y, j_y}, \tag{3}$$

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 κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, we consider $d_{x^2-y^2}$-wave pairing given by

$$F_{ij}(k_y) = \Delta_x \eta_+ \delta_{j_x, i_x+1} + \Delta_{x'} \eta_- \delta_{j_x, i_x+1} - \Delta_y e^{-2ik_y \delta_{i_y,2}} \eta_+ \delta_{j_y, i_y+1} - \Delta_{y'} e^{-2ik_y \delta_{i_y,2}} \eta_- \delta_{j_y, i_y+1} + \Delta_x \eta_- \delta_{j_x, i_x-1}$$

$$+ \Delta_{x'} \eta_+ \delta_{j_x, i_x-1} - \Delta_y e^{2ik_y \delta_{i_y,1}} \eta_- \delta_{j_y, i_y-1} - \Delta_{y'} e^{2ik_y \delta_{i_y,1}} \eta_+ \delta_{j_y, i_y-1} \tag{4}$$

and d_{xy}-like pairing given by

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

$$+ \Delta_x \eta_- \delta_{j_x,i_x-1} + \Delta_{x'} \eta_+ \delta_{j_x,i_x-1} + \Delta_y e^{2ik_y\delta_{i_y,1}} \eta_- \delta_{j_y,i_y-1} + \Delta_{y'} e^{2ik_y\delta_{i_y,1}} \eta_+ \delta_{j_y,i_y-1} - \alpha\Delta_p e^{2ik_y\delta_{i_y,1}} \delta_{j_x,i_x-1} \delta_{j_y,i_y-1},$$
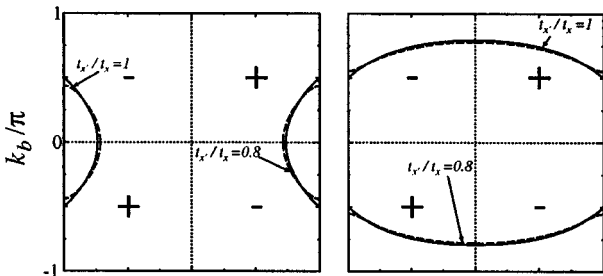
$$(5)$$

with $\alpha = 0.8t$ in accord with Ref. 50. The pairing in real space is shown in Fig. 1(b). Here, we select $\Delta_{x'} = \Delta_y$ and $\Delta_x = \Delta_{y'} = \Delta_p = \Delta_0$, where Δ_0 is a bulk value. For organic superconductors, at the present stage, we can only assume that the pairing symmetry at the surface is the same as that in the bulk.

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 κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, however, BEDT-TTF dimers are further dimerized so that $t_{x'} \neq t_x$, which leads to a splitting of the Fermi surface at around $(k_c, k_b) = (\pm\pi, \pm\pi/2)$.

In (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.[65] Although the pairing symmetry for (TMTSF)$_2X$ remains to be undetermined, here we assume singlet d-wave pairing as an example in which the multiband effect is prominent. In this case, as shown in Fig. 3(a), d-wave is a pairing separated by two lattice spacings given by

$$F_{ij} = \Delta_{2x}\delta_{j_x,i_x+2} + \Delta_{2x}\delta_{j_x,i_x-2}. \qquad (6)$$

The anion potential E_g shown in Fig. 3(b) is estimated from experimental measurement of angle dependent magneto-resistance.[64]
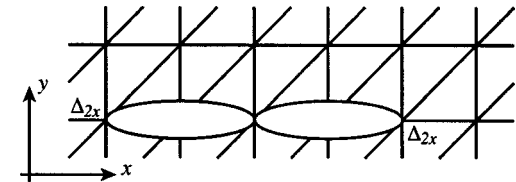
In order to compare our theory with STM experiments, we assume that the STM tip is metallic with a constant density of states, and that tunneling occurs only to the site nearest to the tip. This has been shown to be valid through the study of tunneling conductance of unconventional superconductors.[6] The tunneling conductance spectrum is then given at low temperatures by the normalized surface density of states[6]

$$\rho(E) = \frac{\int_{-\infty}^{\infty} d\omega \rho_S(\omega)\mathrm{sech}^2\left(\frac{\omega+E}{2k_BT}\right)}{\int_{-\infty}^{\infty} d\omega \rho_N(\omega)\mathrm{sech}^2\left(\frac{\omega-2\Delta_0}{2k_BT}\right)}, \qquad (7)$$

$$\rho_S(\omega) = \sum_{k_b,\nu} [|u_1^\nu|^2 \delta(\omega-E_\nu) + |v_1^\nu|^2 \delta(\omega+E_\nu)]. \qquad (8)$$

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.

(a) $d_{x^2-y^2}$-wave

(b) d_{xy}-like-wave
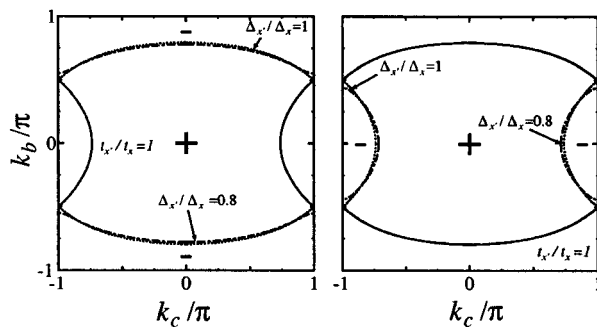
FIG. 2. The Fermi surface and d-wave pairings. (a) $d_{x^2-y^2}$- and (b) d_{xy}-like-wave.
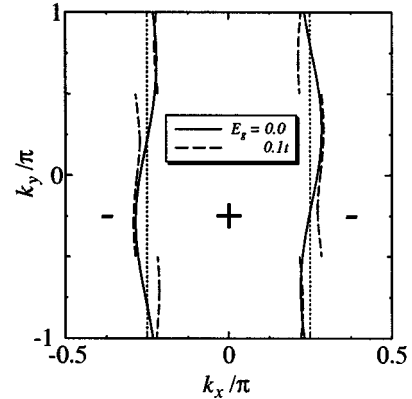
(a) d-wave

(b) $t' = -0.08t$

FIG. 3. (a) Cooper pairs with d-wave symmetry separated by two lattice space and (b) quasi-1D Fermi surface in $t_y = 0.1t_x$ and $t' = -0.08t_x$.
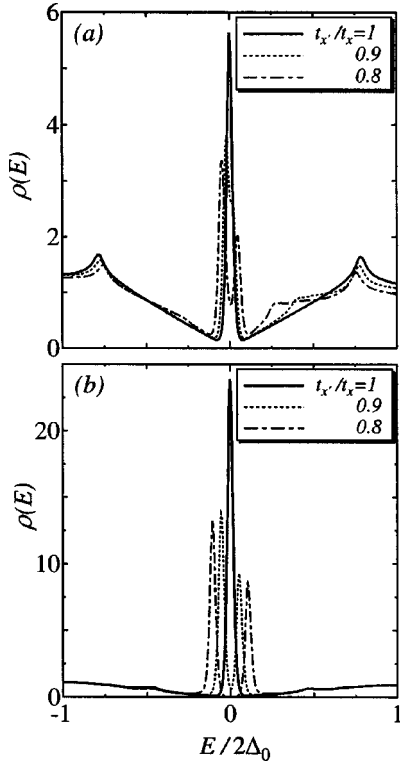
FIG. 4. Tunneling spectrum for (a) $d_{x^2-y^2}$- and (b) d_{xy}-like-waves fixed in $\Delta_{x'} = \Delta_x$.

III. RESULTS OF IN-PLANE TUNNELING SPECTRUM

In this section, we present the calculation results. First, let us focus on the model for κ-(BEDT-TTF)$_2$X. We examine the case of the tunneling spectrum at (100) surface on the xy plane as shown in Fig. 1. 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. This is reminiscent of the ZEP splitting originating from broken time reversal symmetry states.[7,37,67–69]

We have further studied the $t_{x'}/t_x$ dependence of the ZEP splitting. In Fig. 5, the width of the ZEP splitting δ is plotted as functions of $t_{x'}/t_x$ for $d_{x^2-y^2}$ and d_{xy}-like pairings. δ 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 ~ 0.9,[34] 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 when t_x deviates from t_x. The results are plotted in Fig. 6 for various $\Delta_{x'}/\Delta_x$ with $t_{x'}/t_x$ fixed at 0.9. In this case, we observe an overall shift of the splitted ZEP, while the magnitude of the splitting remains unchanged.
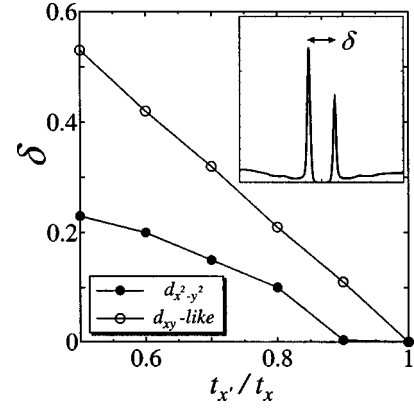

FIG. 5. The $t_{x'}/t_x$ vs the zero-energy peak splitting width δ.

Let us now move on to the model for (TMTSF)$_2$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.[58] 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 κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ and (TMTSF)$_2$ClO$_4$. We find that the multiplicity of the Fermi surface can lead to a splitting of the ZEP. As regards
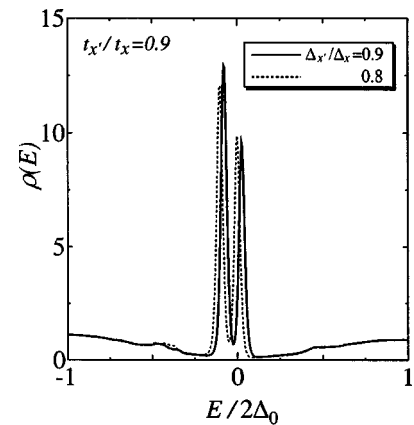

FIG. 6. Tunneling spectrum for various $\Delta_{x'}/\Delta_x$ in $t_{x'}/t_x = 0.9$.
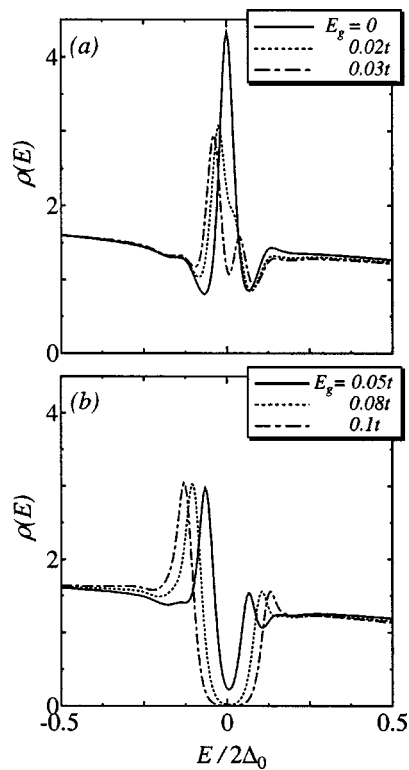
FIG. 7. Tunneling spectrum in $t_y = 0.1t_x$ and $t' = -0.08t_x$ with anion potential E_g.

κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, since t'_x/t_x is estimated to be ~ 0.9,[34] 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[51] actually find no ZBCP in the tunneling spectrum of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. At this stage, it is not easy to make a well oriented surface of organic superconductors due to its fragile crystal structures. For this reason, it is very difficult to observe ZBCP. The situation is different from high-T_c cuprate case, where well oriented surface can be accessible and ZBCP is observed consistent with theoretical prediction.[6,8,39] We hope for the substantial advance of microfabrication technique of organic films in order to observe ZBCP in the near future.

Since many experiments suggest the existence of nodes in the pair potential in κ-(BEDT-TTF)$_2X$, 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,[53–57] namely, point (i) or (ii) mentioned in the Introduction. As far as the roughness of the surface is concerned, we believe it is necessary to study tunneling spectroscopy of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ in the presence of atomic scale roughness as done by Tanuma *et al.* on a lattice model.[36,37] As for the issue of random scattering effect by impurities, Asano *et al.*[52] have shown, both from analytical and numerical calculations 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 κ-(BEDT-TTF)$_2$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 superconductors.[58,66,70] It would also be interesting to apply this probe to κ-(BEDT-TTF)$_2$Cu(NCS)$_2$.

It is well known that ABS's have serious influence on the Josephson current. There are many works on the Josephson effect in unconventional superconductors from both theoretical and experimental view points.[71–78] A future problem is the study of the Josephson effect in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ and (TMTSF)$_2$ClO$_4$.

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