Tomography of pairing symmetry from magnetotunneling spectroscopy — a case study for quasi-1D organic superconductors

Y. Tanuma, K. Kuroki, Y. Tanaka, R. Arita, S. Kashiwaya, and H. Aoki

1Graduate School of Natural Science and Technology, Okayama University, Okayama 700-8530, Japan
2Department of Applied Physics and Chemistry, The University of Electro-Communications, Chofu, Tokyo 182-8585, Japan
3Department of Applied Physics, Nagoya University, Nagoya, 464-8603, Japan
4Department of Physics, University of Tokyo, Hongo, Tokyo 113-0033, Japan
5National Institute of Advanced Industrial Science and Technology, Tsukuba, 305-8568, Japan

(Dated: March 22, 2022)

We propose that anisotropic p-, d-, or f-wave pairing symmetries can be distinguished from a tunneling spectroscopy in the presence of magnetic fields, which is exemplified here for a model organic superconductor (TMTSF)$_2$X. The shape of the Fermi surface (quasi-one-dimensional in this example) affects sensitively the pairing symmetry, which in turn affects the shape (U or V) of the gap along with the presence/absence of the zero-bias peak in the tunneling in a subtle manner. Yet, an application of a magnetic field enables us to identify the symmetry, which is interpreted as an effect of the Doppler shift in Andreev bound states.

One of the most fascinating features of unconventional superconductors with anisotropic pairing symmetries is that Andreev reflection at surfaces can occur in a variety of ways: (i) $p$-wave, (ii) $d$-wave, and (iii) $f$-wave. Namely, at surfaces and interfaces, interference takes place between incident and reflected quasiparticles, which, when the pairing is anisotropic, experience opposite signs of the pair potential depending on the situation, since an anisotropic pairing dictates that the BCS gap function has to have node(s). The interference then becomes constructive, and we end up with the Andreev bound states (ABS) at the surface, which should be observed as a zero-bias conductance peak (ZBCP) in tunneling spectroscopy. This is recognized as a clear signature of an anisotropic pairing. The ZBCP is observed in various anisotropic superconductors such as the high-$T_c$ cuprates and other oxides, $\mathrm{Sr}_2\mathrm{RuO}_4$ and a heavy fermion system UBe$_1_3$. In this context, it is intriguing to investigate whether the ZBCP can be observed in organic superconductors, another class of candidates for anisotropic pairing, such as Bechgaard salts (TMTSF)$_2$X ($X=\mathrm{PF}_6$, $\mathrm{ClO}_4$, etc.)

For (TMTSF)$_2$X, a spin-triplet pairing has been suggested from an observation of a large $H_{c2}$ and an unchanged Knight shift across $T_c$. Anisotropic pairing with nodes on the Fermi surface has been suggested from an NMR measurement, while a thermal conductivity measurement has reported the absence of nodes on the Fermi surface. Theoretically, an anisotropic $p$-wave pairing in which the nodes of the pair potential can be made to avoid intersecting the (quasi-1D) Fermi surface has been proposed in an early stage. The triplet pairing itself, however, is puzzling in an electron mechanism for superconductivity, since the usual wisdom dictates that a triplet pairing should be an outcome of ferromagnetic spin-fluctuation exchange, whereas the superconductivity lies right next to a $2k_F$ SDW in the pressure-temperature phase diagram for TMTSF.

In real materials, however, the question is what will happen for a general shape of the Fermi surface. The warping of the quasi-1D Fermi surface is actually controlled by the ratio between the transfer within the chains and several types of transfers across the chains. So, if the tunneling spectrum is sensitive to the warping, identification of the pairing symmetry will be marred, so we will have to devise some in situ way of probing the symmetry from the tunneling spectroscopy. In the present study, we first show that the tunneling spectrum is in fact sensitive to the warping. This is due to the degradation of the symmetry in the Fermi surface against $k_a \to -k_a$ in $k$ space, and the appearance/disappearance of the ZEP may even be inverted between $d$ and $f$-waves by a small change in the hopping integral.

We then show that we can overcome this difficulty by applying a magnetic field as an in situ control. In a magnetic field screening current affects the ABS spectrum,
which is called the Doppler shift, and this can split the ZBCP \[25, 26, 27, 28\]. We find that ZBCP splits into two for d-wave, while it does not for p and f. The way in which the Doppler shift occurs reflects the shape of the Fermi surface, so the magnetotunneling spectrum provides a unique kind of tomography of the gap function symmetry on the Fermi surface.

![Diagram](image1)

**FIG. 1:** (a) The hopping integrals and the surface normal to the a-axis are shown. Cooper pairs between sites separated by \(m_l\) lattice spacings are illustrated for (b) triplet p- or singlet d-wave \((m_p = m_d = 2)\) and (c) triplet f-wave \((m_f = 4)\).

Three of the present authors have derived in Ref. 28 plausible pairing symmetries by considering pairing interactions arising from spin and charge fluctuations. For discussing the tunneling spectroscopy for a given pairing symmetry, it is more convenient to start from the extended Hubbard model that incorporates effective attractions for the given pairing symmetry. So we have

\[
\mathcal{H} = - \sum_{\langle i,j \rangle, \sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} - \sum_{i\sigma} \mu c_{i\sigma}^\dagger c_{i\sigma} - \frac{V}{2} \sum_{\{i-j\} = m_l, \sigma, \sigma'} c_{i\sigma}^\dagger c_{j\sigma'} c_{j\sigma'} c_{i\sigma},
\]

where \(c_{i\sigma}^\dagger\) creates a hole with spin \(\sigma = \uparrow, \downarrow\) at site \(i = (i_a, i_b)\). \(V\) is the effective attraction that is assumed to act on a pair of electrons when they are \(m_l\) lattice spacings apart (along a axis when the system is quasi-1D), where \(m_l = 2(4)\) for \(l = p, d(f)\)-wave pairing [see Fig. 1(b)(c)].

As for the lattice we take here an anisotropic \((t_b \ll t_a)\) 2D system. To warp the shape of the Fermi surface we introduce a diagonal transfer \(t'\), so the system is an anisotropic triangular lattice. The sample edge is assumed to be \(\perp a\) axis. In what follows we vary \(t' \leq 0\) in the range \(0 \leq |t'/t_a| \leq 0.2\) with a fixed \(t_b/t_a = 0.1\), which covers three typical shapes of the Fermi surface (Fig. 3). In actual \((\text{TMTSF})_2X\) salts, \(|t'|\) may be similar to or even greater than \(t_b\) for some anion \(X\) at low temperatures \[25, 26\]. The chemical potential \(\mu\) is set \((\mu = -1.41t_a)\) to make the band quarter-filled as in TMTSF.

In the latter half of the paper we apply a magnetic field parallel to the c-axis \((\perp a, b)\). Since the penetration depth \(\lambda \geq 12000\, \text{Å}\) \[36\] in \((\text{TMTSF})_2X\) is much greater than the coherence length \(\xi \sim 500\, \text{Å} \sim 150a\) \[37, 38\], the vector potential can be taken to be \(A = (0, H, 0)\) \[39\]. Thus the quasiparticle momentum \(k_b\) in the b direction changes as \(k_b' = k_b + H/\pi t_c\), where \(H_{\sigma} = \phi_0/(\pi^2 \lambda)\) \((= 3.5 \times 10^{-3}\, \text{T}\) for TMTSF) with \(\phi_0 = h/(2e)\).

For the sample with an edge, a mean-field study is performed by introducing a site dependent pair potential. The symmetry of the pair potential is best represented with the d vector as \[25\] \(\Delta_{\sigma\sigma'} = \sum q_{\sigma\sigma'}^q i\delta_{ij}^q (\sigma_q \cdot \delta_y)_{\sigma\sigma'}\), with \(\Delta_{ij}^q = \frac{V}{2} \sum_{\sigma,\sigma'} (\sigma_q \cdot \delta_y)_{\sigma\sigma'} (c_{i\sigma}^\dagger c_{j\sigma'})\), where \(\sigma_q, \delta_y, \delta_x\) are Pauli matrices and \(\delta_0 = 1\). We consider only \(\Delta_{ij}(\sigma = \pm E_p)\) with all the other \(\Delta_{ij}^0 = \Delta_{ij}^q = 0\) for triplet p-wave state, \(\Delta^0\) only for singlet d, and \(\Delta^0\) only for triplet f \[28\].

By solving the mean-field equation for a region with \(N_{a,\nu}(=10^3)\) sites in the \(a\) direction and one site in the \(b\) direction, we have obtained the eigenenergy \(E_{\nu}\). In terms of the eigenenergy \(E_{\nu}\) and the wave functions \(u_{i\nu}^\dagger, v_{i\nu}^\dagger\), the Bogoliubov-de Gennes equation for the surface with the magnetic field applied field parallel to the c-axis is given by

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

with \(H_{ij}(k_b) = -\sum_{\pm} |t_a \delta_{ij, \pm \delta} + 2t_b \cos(k_b a) \delta_{ij} + t' \epsilon \pm i\delta_a^l \delta_{ij, \pm -\delta}| - \mu \delta_{ij}\).

The pair potential is determined self-consistently as

\[
F_{ij}^l = -\frac{V}{2} \sum_{k_{\nu'}} u_{i\nu}^\dagger v_{j\nu'}^* \tanh \left( \frac{E_{\nu}(k_b)}{2k_B T} \right).
\]

The ABS is probed by the surface density of states calculated with the pair potential determined self-consistently. In order to compare our theory with scanning tunneling microscopy (STM) experiments, we assume that the STM tip is metallic with a constant density of states, and that the tunneling occurs only for the site nearest to the tip. This has been shown to be valid through the study of tunneling conductance of unconventional superconductors \[3\]. The tunneling conductance spectrum is then given at low temperatures by the normalized surface density of states \[\rho_{S}(\omega)\],

\[
\rho(E) = \frac{\int_{-\infty}^{\infty} \mathrm{d}\omega \rho_{S}(\omega) \text{sech}^2 \left( \frac{\omega + E}{2k_B T} \right)}{\int_{-\infty}^{\infty} \mathrm{d}\omega \rho_{S}(\omega) \text{sech}^2 \left( \frac{\omega - 2\Delta_{ij}^q}{2k_B T} \right)},
\]

\[
\rho_S(\omega) = \sum_{k_{\nu}} |u_{i\nu}^\dagger|^2 \delta(\omega - E_{\nu}) + |v_{i\nu}^\dagger|^2 \delta(\omega + E_{\nu})\]
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.

Let us first examine the case of zero magnetic field. The shape of the gap in the surface density of states displayed in Fig. 3 is U-shaped for $p$-wave, while V-shaped for $d$ and $f$. This is because the nodal lines (displayed in Fig. 3 by the dashed lines) in the pair potential intersect the Fermi surface for $d$ and $f$, while they do not for $p$-wave, as noted in Ref. 8 for the case of $t'/t = 0$. If we turn to the ZEP, $p$ and $f$-waves have peak [23 8], which is due to the fact that incident and reflected quasiparticles normal to the surface feel opposite signs of the pair potential, which results in a formation of the ABS (whereas oblique incidence is required for $d$-wave). The situation does not change when $t'$ is turned on because the nodes of the $p$-wave lies away from the Fermi surface, so the warping of the Fermi surface has little effect.

On the other hand, the situation is not so simple for $d$ and $f$-waves, where the nodes of the pair potential intersect the Fermi surface. For $t' = 0$, the ZEP appears for $f$, and does not for $d$ as mentioned in Refs. 23 and 34. This is because when the Fermi surface is symmetric against $k_a \leftrightarrow -k_a$, injected and reflected quasiparticles always feel opposite (same) signs of the pair potential for $f(d)$-wave [see Fig. 3(a)]. This time the situation does change when we turn on $t'$, which makes the Fermi surface asymmetric against $k_a \leftrightarrow -k_a$, so that some of the Andreev reflection [the blue area in Fig. 3(b), left panel] lead to ZEP also for $d$-wave as seen in Fig. 3. For $|t'/t_a| = 0.15$ in particular, the warping of the Fermi surface is such that injected and reflected quasiparticles mostly feel opposite (same) signs for $d(f)$ [see Fig. 3(c)], so the situation for the ZEP is inverted as seen in Fig. 3. Consequently, the surface density of states for $d$- and $f$-wave pairings look similar (having V-shaped gap + ZEP) at around $|t'/t_a| \sim 0.1$, so that it should be difficult to distinguish the two by tunneling spectroscopy.

This is where the in situ control of the tunneling by magnetic field comes in. Namely, we now apply magnetic fields along the $c$-axis. In Fig. 3, we show the magnetic field dependence of the surface density of states for $d$ and $f$-waves for $t' = -0.08t_a$. The ZEP for the $d$-wave is seen to split with the magnetic field, while that for $f$ (or $p$; not shown) does not. The magnitude of applied magnetic field here is $H < 30H_c$, where $30H_c$ roughly corresponds to $H_{c2}^{(F)}(\sim 0.1 \text{ T})$, i.e., the upper critical field parallel to $c$-axis, of $(\text{TMTSF})_2\text{PF}_6$ [17].

The result can be interpreted as follows. For $d$-wave, the ZEP is mainly formed by the quasiparticles having $k_b \sim \pi/2$, where $v_{Fb}(k_a)$ and $v_{Fb}(-k_a)$ have the same sign [see Fig. 3(b), left panel]. This situation is similar to those studied previously [4 8 33 34], where the ZEP is strongly affected by the Doppler shift. Namely, the magnetic field gives in this case the injected and reflected quasiparticles additional phase shifts with the same sign, which degrades the constructive interference for the formation of ZEP. By contrast, for $p$ and $f$-waves quasiparticles with $k_b = 0$ mainly contribute to the formation of the zero-energy states [4 41]. For $k_b = 0$, $v_{Fb}(k_a)$ and $v_{Fb}(-k_a)$ have opposite signs regardless of the value of $t'$ [see Fig. 3(b), right panel]. In such a
case, the magnetic field gives the injected and reflected quasiparticles additional phase shifts with opposite signs, which almost cancel out when added, so that the formation of ZEP is barely affected \[2\]. This is physically why we can distinguish \(d\) and \(f\)-waves through the appearance/disappearance of ZEP splitting in the presence of magnetic field.

![Fig. 4](image-url)

**FIG. 4:** The surface density of states normal to \(a\) axis for \(|t'/t_s| = 0.08\) in the presence of the magnetic field for the (a) \(d\)-wave and (b) \(f\)-wave.

Although we have here exemplified the tunneling spectra for the quasi-1D model, the physics involved (i.e., the signs of the pair potential and the signs of \(v_F\) for the incident and reflected particles) is quite general, so that the principle for distinguishing various singlet and triplet pairing symmetries should apply for other, non-cubic (or non-tetragonal) systems.

Y.T. would like to thank K. Maki and K. Yamaji for pointing out the possible importance of the realistic shape of the Fermi surface. He also acknowledges a financial support of Japan Society for the Promotion of Science for Young Scientists. This work was in part supported by the Core Research for Evolutional Science and Technology (CREST) of the Japan Science and Technology Corporation. The computations were performed at the Supercomputer Center of Institute for Solid State Physics, and the Computer Center in the University of Tokyo.

[1] L.J. Buchholtz and G. Zwicknagl, Phys. Rev. B 23, 5788 (1981).
[2] C.R. Hu, Phys. Rev. Lett. 72, 1526 (1994).
[3] Y. Tanaka and S. Kashiwaya, Phys. Rev. Lett. 74, 3451 (1995); S. Kashiwaya and Y. Tanaka, Rep. Prog. Phys. 63, 1641 (2000); T. Löfwander, V. S. Shumeiko and G. Wendin, Supercond. Sci. Technol. 14, R53 (2001).
[4] M. Fogelström, D. Rainer, and J.A. Sauls, Phys. Rev. Lett. 79, 281 (1997).
[5] C. Honerkamp and M. Sigrist, J. Low Temp. Phys. 111, 895 (1998).
[6] M. Yamashiro, et al., Phys. Rev. B 56, 7847 (1997).
[7] J. Geerk, X.X. Xi, and G. Linker, Z. Phys. B. 73, 329 (1988).
[8] M. Covington, et al., Phys. Rev. Lett. 79, 277 (1997).
[9] J.Y.T. Wei, et al., Phys. Rev. Lett. 81, 2542 (1998).
[10] L. Alff, et al., Phys. Rev. B 55, (1997) R14757.
[11] F. Laube, et al., Phys. Rev. Lett. 84, 1595 (2000).
[12] Z.Q. Mao, et al., Phys. Rev. Lett. 87, 037003 (2001).
[13] Ch. Wälti, et al., Phys. Rev. Lett. 84, 5616 (2000).
[14] Tunneling experiment on (TMTSF)$_2$ClO$_4$ has recently been performed by T. Arai, et al., to be published in Synthetic Metals.
[15] D. Jérôme, et al., J. Phys. Lett. (France) 41, L92 (1980).
[16] K. Bechgaard, et al., Phys. Rev. Lett. 46, 852 (1981).
[17] I.J. Lee, et al., Phys. Rev. Lett. 78, 3555 (1997); Phys. Rev. B 62, R14669 (2000).
[18] I.J. Lee, et al., Phys. Rev. Lett. 88, 017004 (2002).
[19] M. Takigawa, H. Yasuoka, and G. Saito, J. Phys. Soc. Jpn. 56, 873 (1987).
[20] S. Belin and K. Behnia, Phys. Rev. Lett. 79, 2125 (1997).
[21] A.A. Abrikosov, J. Low Temp. Phys. 53, 359 (1983).
[22] Y. Hasegawa and H. Fukuyama, J. Phys. Soc. Jpn. 56, 877 (1987).
[23] A.G. Lebed, Phys. Rev. B 59, R721 (1999); A.G. Lebed, K. Machida, and M. Ozaki, ibid. 62, R795 (2000).
[24] R.L. Greene and E.M. Engler, Phys. Rev. Lett. 45, 1587 (1980).
[25] H. Shimahara, J. Phys. Soc. Jpn. 58, 1735 (1989).
[26] K. Kuroki and H. Aoki, Phys. Rev. B 60, 3060 (1999).
[27] H. Kino and H. Kontani, J. Low Temp. Phys. 117, 317 (1999).
[28] K. Kuroki, R. Arita, and H. Aoki, Phys. Rev. B 63, 094509 (2001).
[29] K. Sengupta, et al., Phys. Rev. B 63, 144531 (2001).
[30] Y. Tanuma, et al., Phys. Rev. B 64, 214510 (2001).
[31] We use the term ZEP for the peak in the surface density of states to distinguish it from the conductance peak (ZBCP).
[32] Quite recently, the *bulk* quasiparticle density of states of (TMTSF)$_2$X has been examined for various pairing symmetries in R.D. Ducan, R.W. Cerng and A.R. Sa de Melo, unpublished [cond-mat/0203570].
[33] M. Aprili, E. Badica and L.H. Greene, Phys. Rev. Lett. 83, 4630 (1999).
[34] R. Krupke and G. Deutscher, Phys. Rev. Lett. 83, 4634 (1999).
[35] L. Ducasse, et al., J. Phys. C 19, 3805 (1986).
[36] K. Yamaji, J. Phys. Soc. Jpn. 55, 860 (1986).
[37] L.P. Le, et al., Phys. Rev. B 48, 7284 (1993).
[38] H. Yoshino, et al., J. Phys. Soc. Jpn. 68, 3142 (1999).
[39] M. Sigrist and K. Ueda, Rev. Mod. Phys. 63, 239 (1991).
[40] Y. Tanaka, et al., J. Phys. Soc. Jpn. 71, 271 (2002).
[41] Y. Tanaka, et al., Phys. Rev. B 60, 6308 (1999).
[42] Y. Tanaka, et al., unpublished.
[43] For \(d\)-wave with further increase of \(|t'|\), the imaginary part of the pair potential turns out to be significant near the surface due to a mixing of \(p\)-wave, which results in a broken time-reversal symmetry state, and makes the splitting of the ZEP much wider. However, we believe that the study of the symmetry mixing in the actual (TMTSF)$_2$X would require a model with *spin dependent* electron-electron interaction, so we have restricted ourselves to \(|t'| \leq 0.08t_s\) in the presence of magnetic field, where the symmetry mixing is negligible.