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Microscopic theory of spin-triplet *f*-wave pairing in quasi-one-dimensional organic superconductors

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We present a microscopic theory of fluctuation-mediated pairing mechanism in organic superconductors $(TMTSF)_2X$, where the experimentally observed coexistence of $2k_F$ charge fluctuation and $2k_F$ spin fluctuation is naturally taken into account. We have studied, within the random phase approximation, the extended Hubbard model at quarter filling on a quasi-one-dimensional lattice, where we consider the off-site repulsive interaction up to third next-nearest neighbors along with the on-site repulsion. The results show that spin-triplet *f*-wave-like pairing can be realized in this system, dominating over singlet *d*-wave-like pairing, if $2k_F$ spin and $2k_F$ charge fluctuations coexist.

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It has been a long standing issue to clarify the superconducting state of quasi-one-dimensional (q1D) organic superconductors $(TMTSF)_2X$ ($X=PF_6$, ClO_4, etc.), so-called the Bechgaard salts.[1,2] One may expect unconventional superconducting states due to the quasi-one-dimensional nature of these materials as well as the electron correlation effects generally seen in organic materials. In fact, an unchanged Knight shift across T_c,[3] along with a large H_{c2},[4,5] supports a realization of spin-triplet pairing. As for the orbital part of the order parameter, there have been NMR experiments[3,6] which may be regarded as evidence for the presence of nodes (although this is still controversial), while a thermal conductivity measurement suggests absence of nodes.[7]

Theoretically, the triplet *p*-wave pairing state in which the nodes of the pair potential do not intersect the Fermi surface has been proposed.[8–10] However, the occurrence of triplet pairing in $(TMTSF)_2X$ is puzzling[11] from a microscopic point of view since superconductivity lies right next to a $2k_F$ spin density wave (SDW) in the pressure-temperature phase diagram.[12] Naively, SDW spin fluctuations should favor spin-singlet *d*-wave-like pairing as suggested by several other authors.[13–15] One should note, however, that the insulating phase is not pure SDW at least for some anions, namely, $2k_F$ charge density wave (CDW) actually coexists with $2k_F$ SDW.[16,17] In fact, one of the present authors has proposed[18] that triplet *f*-wave-like [see Fig. 1(c) for a typical pair potential] pairing may dominate over *p*-wave pairing and become competitive against *d*-wave-like pairing [see Fig. 1(b)] due to a combination of quasi-1D (disconnected) Fermi surface and the coexistence of $2k_F$ SDW and $2k_F$ CDW fluctuations. A similar scenario has been proposed by Fuseya *et al.*[19] Concerning the *f*-wave versus *d*-wave competition, it has also been proposed that magnetotunneling spectroscopy[20] via Andreev resonant states[21] is a promising method to detect the *f*-wave pairing.

However, there has been no *microscopic* theory for this hypothetical *f*-wave pairing in $(TMTSF)_2X$ starting from a Hamiltonian that assumes only purely electronic repulsive interactions.[22] To resolve this issue, we study an extended Hubbard model at quarter filling on a quasi-one-dimensional lattice within the random phase approximation (RPA). We consider off-site repulsions up to third nearest neighbors along with the on-site repulsion in order to naturally take into account the coexistence of $2k_F$ charge and $2k_F$ spin fluctuations. The merit of adopting RPA[23] is that we can easily take into account the off-site repulsion as compared to fluctuation exchange (FLEX) approximation, where it is by no means easy to take into account distant interactions.[24,25]

The model Hamiltonian is given as

$$H = -\sum_{\langle i,j\rangle,\sigma} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma} + U\sum_{i} n_{i\uparrow} n_{i\downarrow} + \sum_{|i-j|=m_l} V_{i,j} n_i n_j,$$

where $c_{i\sigma}^{\dagger}$ creates a hole [note that $(TMTSF)_2X$ is actually a 3/4 filling system in the electron picture] with spin $\sigma=\uparrow,\downarrow$ at site $\boldsymbol{i}=(i_a,i_b)$. Here, $\langle \boldsymbol{i},\boldsymbol{j}\rangle$ stands for the summation over nearest neighbor pairs of sites. As for the hopping parameters, we take $t_{ij}=t_a$ for nearest neighbor in the (most conductive) *a* direction, and $t_{ij}=t_b$ for nearest neighbor in the *b* direction. We choose $t_b=0.2t_a$ to take into account the quasi-one-dimensionality. t_a is taken as the unit of energy throughout the study. U and V_{ij} are the on-site and the off-site repulsive interactions, respectively. We take distant off-site repulsions because it has been shown previously that nearest neighbor and second nearest neighbor off-site repulsion is necessary to have coexistence of $2k_F$ spin and $2k_F$ charge density waves.[26] Here we take off-site repulsions up to third nearest neighbors, namely, $V_{\boldsymbol{i},\boldsymbol{j}}=V_0$, V_1, and V_2 with $m=|i_a-j_a|=1,2$, and 3, respectively. The effect of the third nearest neighbor repulsion, V_2, will be discussed at the end of the paper. The effective pairing interactions for the singlet and triplet channels due to spin and charge fluctuations are given as

$$V^s(\boldsymbol{q},\omega_l) = U + V(\boldsymbol{q}) + \tfrac{3}{2}U^2\chi_s(\boldsymbol{q},\omega_l) - \tfrac{1}{2}[U+2V(\boldsymbol{q})]^2\chi_c(\boldsymbol{q},\omega_l), \quad (1)$$

$$V^t(\boldsymbol{q},\omega_l) = V(\boldsymbol{q}) - \tfrac{1}{2}U^2\chi_s(\boldsymbol{q},\omega_l) - \tfrac{1}{2}[U+2V(\boldsymbol{q})]^2\chi_c(\boldsymbol{q},\omega_l), \tag{2}$$

within RPA, where

$$V(\boldsymbol{q}) = 2V_0\cos q_x + 2V_1\cos(2q_x) + 2V_2\cos(3q_x), \tag{3}$$

and ω_l is the Matsubara frequency. Here, χ_s and χ_c are the spin and charge susceptibilities, respectively, which are given as

$$\chi_s(\boldsymbol{q},\omega_l) = \frac{\chi_0(\boldsymbol{q},\omega_l)}{1-U\chi_0(\boldsymbol{q},\omega_l)}$$

$$\chi_c(\boldsymbol{q},\omega_l) = \frac{\chi_0(\boldsymbol{q},\omega_l)}{1+[U+2V(\boldsymbol{q})]\chi_0(\boldsymbol{q},\omega_l)}. \tag{4}$$

Here, χ_0 is the bare susceptibility given by

$$\chi_0(\boldsymbol{q},\omega_l) = \frac{1}{N}\sum_p \frac{f(\epsilon_{p+q})-f(\epsilon_p)}{\omega_l - (\epsilon_{p+q}-\epsilon_p)}$$

with $\epsilon_k = -2t_a\cos k_a - 2t_b\cos k_b - \mu$ and $f(\epsilon_p) = 1/[\exp(\epsilon_p/T)+1]$. χ_0 peaks at the nesting vector $\boldsymbol{Q}$ [$=(\pi/2,\pi)$ here] of the Fermi surface. The terms proportional to χ_s and χ_c in Eqs. (1) and (2) represent effective pairing interactions due to the spin and charge fluctuations, respectively. The chemical potential μ is determined so that the band is quarter-filled, which is $\mu=-1.38$. We take $U=1.6$ throughout the study, which is large enough to have strong $2k_F$ spin fluctuations [large $\chi_s(\boldsymbol{Q})=\chi_s(\boldsymbol{Q},0)$] but not so large as to drive SDW instability at high temperatures. The off-site interactions are chosen so that $2k_F$ charge fluctuations are induced as will be discussed. In the actual numerical calculation, we take $N=400\times 40$ k-point meshes except for low temperatures, where we take $N=800\times 80$ meshes.

To obtain the onset of the superconducting state, we solve the gap equation within the weak-coupling theory:

$$\lambda\Delta(\boldsymbol{k}) = -\sum_{\boldsymbol{k}'} V^{s,t}(\boldsymbol{k}-\boldsymbol{k}',0)\frac{\tanh(\beta\epsilon_{\boldsymbol{k}'}/2)}{2\epsilon_{\boldsymbol{k}'}}\Delta(\boldsymbol{k}'). \tag{5}$$

The transition temperature T_C is determined by the condition, $\lambda=1$. In the weak coupling theory, ω dependence of the pair potential $\Delta(\boldsymbol{k})$ is neglected. Although this approximation is quantitatively insufficient, it is expected to be valid for studying the pairing symmetry of $\Delta(\boldsymbol{k})$ mediated by both spin and charge fluctuations. In the following calculations, we study triplet and singlet cases with $\Delta(\boldsymbol{k})=-\Delta(-\boldsymbol{k})$ and $\Delta(\boldsymbol{k})=\Delta(-\boldsymbol{k})$, respectively. We define $\phi_s(\boldsymbol{k})=\Delta(\boldsymbol{k})/\Delta_M$ and $\phi_t(\boldsymbol{k})=\Delta(\boldsymbol{k})/\Delta_M$ for singlet and triplet pairing, respectively, where Δ_M is the maximum value of the pair potential.

Equations (1)–(4), show that when $U\sim-[U+2V(\boldsymbol{Q})]$ is satisfied, $|V^s(\boldsymbol{Q})|\sim|V^t(\boldsymbol{Q})|$ holds, apart from the first order terms, which are negligible in the limit of strong spin and /or charge fluctuations (but turn out to be important in the actual cases considered later). This, along with the disconnectivity of the Fermi surface (note that the number of nodes intersecting the Fermi surface is the same between f and d waves), is expected to make spin triplet f-wave pairing competitive against singlet d-wave pairing.[18]

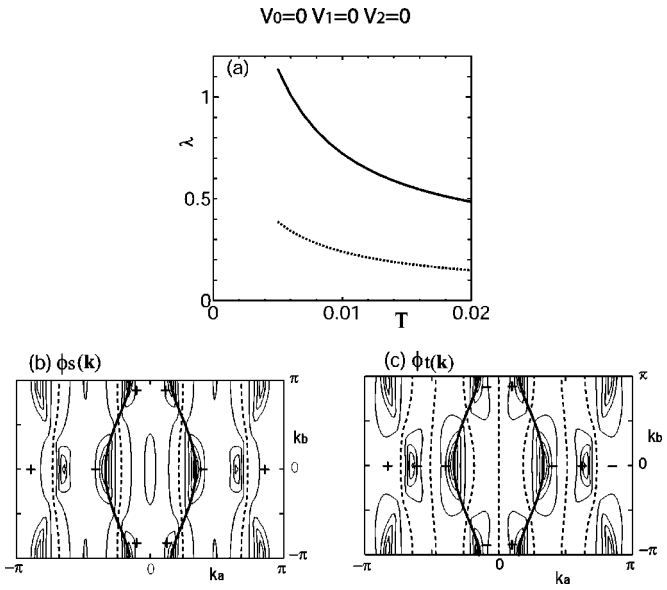

FIG. 1. Calculation results for $U=1.6$, $V_0=V_1=V_2=0$: (a) Temperature dependence of λ for singlet (solid line) and triplet (dotted line) pairings. Contour plots of (b) $\phi_s(\boldsymbol{k})$ and (c) $\phi_t(\boldsymbol{k})$ at $T=0.01$. In (b) and (c), the solid lines represent the Fermi surface and the dotted lines denote the nodal lines of the pair potentials.

We now discuss the calculation results. First, we focus on the case where spin fluctuation is dominant, e.g., $V_0=V_1=V_2=0$. As shown in Fig. 1, the magnitude of λ for the singlet case is much larger than that for the triplet case. The resulting singlet pair potential $\phi_s(\boldsymbol{k})$ changes sign as $+-+-$ along the Fermi surface [see Fig. 1(b)]. We call this d-wave pairing, where $\phi_s(\boldsymbol{k})$ is roughly proportional to $\cos(2k_x)$. On the other hand, the triplet pair potential $\phi_t(\boldsymbol{k})$ changes sign as $+-+-+-$ along the Fermi surface [see Fig. 1(c)]. We call this f wave, where $\phi_t(\boldsymbol{k})$ is roughly proportional to $\sin 4k_x$. The results here are expected from the previous FLEX study.[18]

We now discuss the cases where we turn on the off-site repulsions. In order to to have the coexistence of $2k_F$ spin and $2k_F$ charge fluctuations as experimentally observed, namely, to have $\chi_s(\boldsymbol{Q})\sim-\chi_c(\boldsymbol{Q})=-\chi_c(\boldsymbol{Q},0)$, $U\sim-[U+2V(\boldsymbol{Q})]$ must be satisfied as mentioned earlier. To accomplish this, V_1 has to be close to $U/2$, as can be seen from Eq. (3). Namely, since the x component of $\boldsymbol{Q}$ is $Q_x=\pi/2$, the V_1 term in Eq. (3) is dominant for $q\simeq\boldsymbol{Q}$, making $U\sim-[U+2V(\boldsymbol{Q})]$ if $V_1\simeq U/2$. Thus, we choose $V_1=U/2=0.8$. As for the other off-site repulsions, we first choose $V_0=1.2$ and $V_2=0.5$ as a typical value. As shown in Figs. 2(a) and 2(b), χ_s and χ_c peak around $(k_a,k_b)=\pm(\pi/2,\pi),\pm(\pi/2,-\pi)$ and the maximum values are both about 9.1, so that we have the situation where $2k_F$ spin and $2k_F$ charge fluctuations coexist. As shown in Fig. 2(c), the magnitude of λ for triplet pairing is now much larger than that for singlet pairing. The corresponding singlet pair potential $\phi_s(\boldsymbol{k})$ has the d-wave form as shown in Fig. 2(d), while the triplet one $\phi_t(\boldsymbol{k})$ has the f-wave form as shown in Fig. 2(e). Note that the result of λ_{triplet}

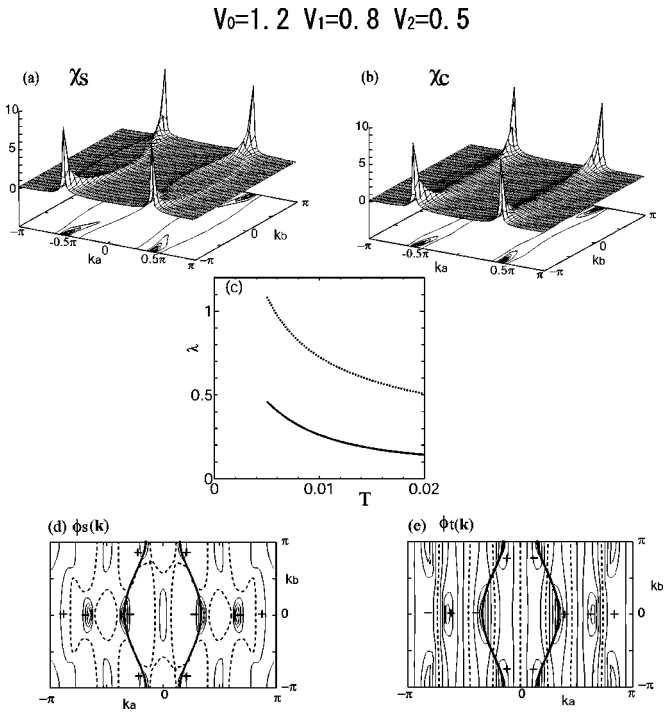

FIG. 2. Calculation results for $U=1.6$, $V_0=1.2$, $V_1=0.8$, $V_2=0.5$: (a) $\chi_s(\boldsymbol{k})$ at $T=0.01$; (b) $\chi_c(\boldsymbol{k})$ at $T=0.01$; (c) temperature dependence of λ for singlet (solid line) and triplet (dotted line) pairings. Contour plots of (d) $\phi_s(\boldsymbol{k})$ and (e) $\phi_t(\boldsymbol{k})$ at $T=0.01$. In (d) and (e), the solid lines represent the Fermi surface and the dotted lines denote the nodes of the pair potentials.

$\gg \lambda_{\text{singlet}}$ is rather unexpected from the previous phenomelogical argument[18] because when $\chi_c(\boldsymbol{Q})=\chi_s(\boldsymbol{Q})$, the *f* wave is only *degenerate* with the *d* wave in the previous theory. The origin of this discrepancy is the first order terms in Eqs. (1) and (2), which are neglected in the phenomelogical theory. Thus, we have obtained a remarkable result here, namely, the *f* wave can *completely* dominate over the *d* wave when $2k_F$ spin and charge fluctuations coexist.

In order to further look into this point, we next reduce V_1 from 0.8, thereby suppressing the charge fluctuation. The maximum value of χ_c (not shown) is 5.2 and 3.2 for $V_1=0.78$ and $V_1=0.75$, respectively. Although the maximum value of χ_c is smaller than that of χ_s (=9.1) in these cases, λ for the triplet case is still larger than ($V_1=0.78$) or competitive against ($V_1=0.75$) that for the singlet case as seen in Fig. 3. This means that *f*-wave pairing has a chance to be realized *even if spin fluctuation dominates, as far as* $2k_F$ *charge fluctuation exists*.[18]

Finally, in order to look into the effect of the third nearest neighbor interaction V_2, we set $V_2=0$ leaving the other parameters the same as in Fig. 2. As seen in Fig. 4(a), the peak of χ_c is slightly shifted towards $4k_F$ ($q_x=\pi$) when $V_2=0$, compared to the case of $V_2=0.5t$ shown in Fig. 2(a). [χ_s is the same as Fig. 2(a) since V_2 does not affect χ_s within the present formalism.] In this case, the singlet-triplet competition becomes much more subtle as seen in Fig. 4(b). The reason for this can be found in Fig. 4(c), namely, $\phi_s(\boldsymbol{k})$ in this case has the same sign on most of the portion of the Fermi surface. In other words, it is more like the *s* wave than

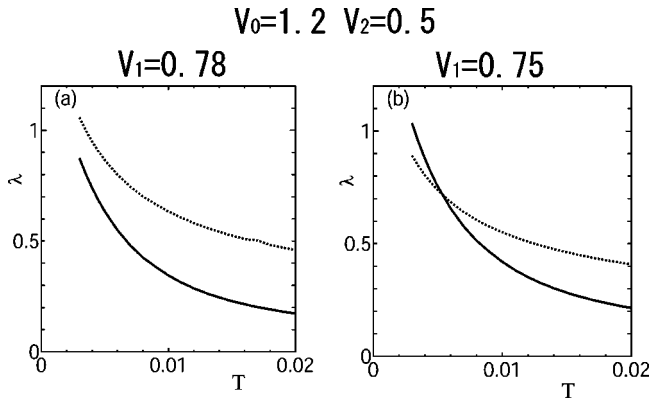

FIG. 3. Calculation results for $U=1.6$, $V_0=1.2$, and $V_2=0.5$: (a) Temperature dependence of λ for singlet (solid line) and triplet (dotted line) pairings for $V_1=0.78$; (b) temperature dependence of λ for singlet (solid line) and triplet (dotted line) pairings for $V_1=0.75$.

d wave. This *s* wave pairing is induced by charge fluctuation, *which does not totally cancel out with spin fluctuation in Eq. (1)* because the wave vector at which χ_c peaks deviates from that for χ_s. Since the *s*-wave pair potential has the same sign on most of the portion of the Fermi surface, almost all the pair scattering processes on the Fermi surface, mediated by the *attractive* interaction [note the minus sign in Eq. (1)] due to charge fluctuation, have positive contributions to superconductivity, making singlet pairing much more enhanced compared to the case with nonzero V_2. Conversely, the present results show that V_2 has the effect of stabilizing $2k_F$ charge fluctuation, which has a tendency to shift towards $4k_F$

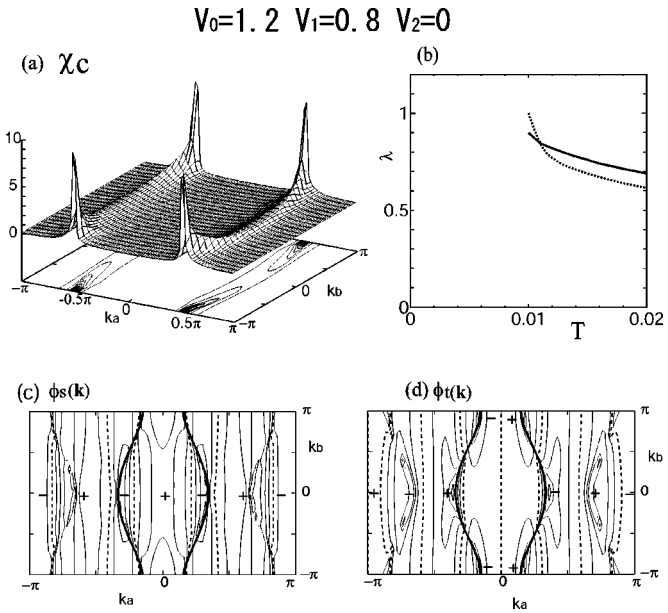

FIG. 4. Calculation results for $U=1.6$, $V_0=1.2$, $V_1=0.8$, $V_2=0$: (a) $\chi_c(\boldsymbol{k})$ at $T=0.01$; (b) temperature dependence of λ for singlet (solid line) and triplet (dotted line) pairings. Contour plots of (c) $\phi_s(\boldsymbol{k})$ and (d) $\phi_t(\boldsymbol{k})$ at $T=0.01$. In (c) and (d), the solid lines represent the Fermi surface and the dotted lines denote the nodes of the pair potentials.

fluctuation when only V_0 and V_1 are present, and this effect in turn suppresses singlet pairing because in that case a strong cancellation occurs between the third and the fourth terms in Eq. (1). Since the screening effect is known to be weak in quasi-one-dimensional systems, it is likely that such a distant off-site repulsion is present in the actual $(TMTSF)_2X$.

To summarize, we have presented a microscopic theory of pairing mechanism in organic superconductors $(TMTSF)_2X$, where we have taken into account the coexistence of the $2k_F$ charge fluctuation and $2k_F$ spin fluctuation by considering off-site repulsions up to third nearest neighbors. We have shown that the *f*-wave triplet pairing symmetry can be realized in this system when $2k_F$ charge density fluctuation and $2k_F$ spin density fluctuation coexists. Surprisingly, the condition for realizing *f*-wave pairing is eased compared to that in the previous phenomelogical theory.[18]

In this paper, we have neglected the realistic shape of the Fermi surface observed in the actual $(TMTSF)_2X$.[27] Although the influence of this effect on the *f*-wave pairing is expected to be small because the *x* component of the nesting vector is close to $\pi/2$ a detailed analysis remains as a future study.

Let us also comment on the one-dimensional fluctuation effects that are not taken into account in the present RPA approach. We have recently performed ground state quantum Monte Carlo study for the Hubbard model with only the on-site repulsion on a quasi-1D lattice, in which SDW fluctuations strongly dominate over CDW fluctuations.[28] In such a case, it is expected that *d*-wave pairing strongly dominates over *f*-wave pairing from the standpoint of RPA, but our Monte Carlo results show that *f*-wave pairing is surprisingly competitive against *d*-wave even though spin fluctuations $>$ charge fluctuations clearly holds. Thus, the one-dimensional effects that are not taken into account in the present approach are expected to work in favor of the *f*-wave pairing. In this sense, we believe that *f*-wave pairing can dominate over *d* wave in a parameter regime with off-site repulsions smaller than those adopted in the present study. This point remains as an interesting future problem. In any case, our overall conclusion that *f*-wave superconductivity is likely to be taking place in the $(TMTSF)_2X$ compounds should not be altered even if we take into account the one-dimensional effects properly.

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