The effect of interchain interaction on the pairing symmetry competition in organic superconductors (TMTSF)$_2$X

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We investigate the effect of interchain repulsive interaction on the pairing symmetry competition in quasi-one-dimensional organic superconductors (TMTSF)$_2$X by applying random phase approximation and quantum Monte Carlo calculation to an extended Hubbard model. We find that interchain repulsive interaction enhances the $2k_F$ charge fluctuations, thereby making the possibility of spin-triplet $f$-wave pairing dominating over spin-singlet $d$-wave pairing realistic.

KEYWORDS: (TMTSF)$_2$X, superconductivity, spin-triplet pairing, interchain interaction, charge and spin fluctuations

Possible occurrence of unconventional superconductivity in organic conductors has been of great interest recently. Microscopically understanding the mechanism of pairing in those materials is an intriguing theoretical challenge. Among the various candidates of unconventional superconductors, the pairing mechanism of quasi-one-dimensional (q1D) organic superconductors (TMTSF)$_2$X ($X = PF_6, ClO_4$, etc.), so called the Bechgaard salts,$^1,2$ has been quite puzzling. Namely, since superconductivity lies right next to the $2k_F$ spin density wave (SDW) phase in the pressure-temperature phase diagram, a spin-singlet $d$-wave-like pairing (shown schematically in Fig.1(a)) is expected to take place as suggested by several authors.$^3−5$ However, an unchanged Knight shift across $T_c$ and a large $H_{c2}$ exceeding the Pauli limit$^6$ suggest a realization of spin-triplet pairing. As for the orbital part of the order parameter, there have been NMR experiments suggesting the existence of nodes and thus unconventional pairing,$^7$ although a thermal conductivity measurement suggests absence of nodes for (TMTSF)$_2$ClO$_4$.$^9$

As a possible solution for this puzzle of spin-triplet pairing, one of the present authors has phenomenologically proposed that triplet $f$-wave-like pairing (whose gap is shown schematically in Fig.1(b)) may take place due to a combination of quasi-1D (disconnected) Fermi surface and the coexistence of $2k_F$ spin and $2k_F$ charge fluctuations.$^{10}$ Namely, due to the disconnectivity of the Fermi surface, the number of gap nodes that intersect the Fermi surface is the same between $d$ and $f$. Moreover, if the $2k_F$ spin and charge fluctuations have about the same magnitude, spin-singlet and spin-triplet pairing interactions have close absolute values (with opposite signs) as will be explained later. In such a case, spin-triplet $f$-wave pairing should be closely competitive against singlet $d$-wave pairing. As for other possibilities of triplet pairing, the $p$-wave state in which the nodes of the gap (Fig.1(c)) do not intersect the Fermi surface has been considered from the early days,$^{11−13}$ but from a microscopic point of view, spin-triplet pairing interaction has a negative sign for the momentum transfer of $2k_F$ unless spin fluctuations are highly anisotropic, so that a gap that changes sign between the left and right portions of the Fermi surface is unlikely to take place.$^{14}$ A similar phenomenological proposal of $f$-wave pairing in (TMTSF)$_2$X has also been given by Fuseya et al.$^{15}$ Experimentally, the $f$-wave scenario due to the coexistence of $2k_F$ spin and charge fluctuations is indirectly supported by the observation that $2k_F$ charge density wave (CDW) actually coexists with $2k_F$ SDW in the insulating phase lying next to the superconducting phase.$^{16,17}$

As for microscopic theories for the pairing competition, we have previously shown using a ground state quantum Monte Carlo method that $f$-wave strongly dominates over $p$-wave in the Hubbard model that considers only the on-site repulsive interaction.$^{18}$ More recently, we have shown, by applying random phase approximation (RPA) to an extended Hubbard model, that $f$-wave pairing can indeed dominate over $d$-wave pairing when we have large enough second nearest neighbor repulsion $V'$, which has been known for some years to have the effect of stabilizing $2k_F$ CDW configuration.$^{20,21}$ To be more precise, the condition for $f$-wave dominating over $d$-wave is to have $V' \simeq U/2$ (where $U$ is the on-site repulsion) or larger $V'$ because $2k_F$ spin and $2k_F$ charge fluctuations have the same magnitude for $V' = U/2$ within RPA. A similar condition for $f$-wave being competitive against $d$-wave has also been obtained in a recent renor-
fluctuations are given as the singlet and triplet channels due to spin and charge pulsions considered in our previous study. We take as the unit of energy, and we adopt $V_{\text{next nearest}}$ (in addition to the intrachain on-site ($U$) nearest neighbor repulsive interactions, respectively, where we take practically a 3/4 filling system in the electron picture) with.

Fig. 2. The model of the present study is shown.

The model considered in the present study is shown in Fig. 2. In standard notations, the Hamiltonian is given as

$$H = - \sum_{<i,j>\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow} + \sum_{<i,j>} V_{ij} n_{i\uparrow} n_{j\downarrow},$$

where $c_{i\sigma}^\dagger$ creates a hole (note that (TMTSF)$_2$X is actually a 3/4 filling system in the electron picture) with spin $\sigma = \uparrow, \downarrow$ at site $i$. As for the kinetic energy terms, we consider nearest neighbor hoppings $t_{ij} = t$ in the (most conductive) $a$-direction and $t_{ij} = t_\perp$ in the $b$-direction. $t$ is taken as the unit of energy, and we adopt $t_\perp = 0.2t$ throughout the study. $U$ and $V_{ij}$ are the on-site and the off-site repulsive interactions, respectively, where we take into account the nearest neighbor interchain repulsion $V_\perp$ in addition to the intrachain on-site ($U$), nearest ($V$), next nearest ($V'_\perp$), and third nearest ($V''\perp$) neighbor repulsions considered in our previous study.

Within RPA, the effective pairing interactions for the singlet and triplet channels due to spin and charge fluctuations are given as

$$V^s(q) = U + V(q) + \frac{3}{2} U^2 \chi_s(q)$$

$$- \frac{1}{2} (U + 2V(q))^2 \chi_s(q)$$

where

$$V(q) = 2V \cos q_x + 2V' \cos 2q_x + 2V'' \cos 3q_x + 2V_\perp \cos q_y.$$ (3)

$$V^t(q) = V(q) - \frac{1}{2} U^2 \chi_s(q)$$

$$- \frac{1}{2} (U + 2V(q))^2 \chi_c(q),$$ (2)

Here, $\chi_s$ and $\chi_c$ are the spin and charge susceptibilities, respectively, which are given as

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

$$\chi_c(q) = \frac{\chi_0(q)}{1 + (U + 2V(q)) \chi_0(q)}.$$ (4)

Here $\chi_0$ is the bare susceptibility given by

$$\chi_0(q) = \frac{1}{N} \sum_p \frac{f(\epsilon_p + q) - f(\epsilon_p)}{\epsilon_p - \epsilon_p + q}$$

with $\epsilon_k = -2t \cos k_x - 2t_\perp \cos k_y - \mu$ and $f(\epsilon_p) = 1/(\exp(\epsilon_p/T) + 1)$. $\chi_0$ peaks at the nesting vector $Q_{2k_F}$ (= $\pi/2, \pi$ here) of the Fermi surface.

To obtain $T_c$, we solve the linearized gap equation within the weak-coupling theory,

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

The eigenfunction $\Delta^{s,t}$ of this eigenvalue equation is the gap function. The transition temperature $T_c$ is determined as the temperature where the eigenvalue $\lambda$ reaches unity. Note that the main contribution to the summation in the right hand side comes from $k - k' \approx Q_{2k_F}$ because $V^{s,t}(q)$ peaks around $q = Q_{2k_F}$. Although RPA is quantitatively insufficient for discussing the absolute values of $T_c$, we expect this approach to be valid for studying the competition between different pairing symmetries.

Now, from eqs.(3) and (4), it can be seen that $\chi_s(Q_{2k_F}) = \chi_s(Q_{2k_F})$ holds when $V' + V_\perp = U/2$, which in the absence $V_\perp$ of course reduces to the condition $V' = U/2$ obtained in our previous study. This in turn results in $V^s(Q_{2k_F}) = -V^t(Q_{2k_F})$ for the pairing interactions apart from the first order terms as can be seen from eqs.(1) and (2). Thus, considering the fact that the number of nodes intersecting the Fermi surface is the same between $d$ and $f$, the condition for $f$-wave being competitive against $d$-wave should be $V' + V_\perp \approx U/2$. The possibility of this condition being satisfied in actual materials is realistic since $V_\perp$ can be comparable with the intrachain off-site repulsions due to the fact that the lattice constant in the $a$- and $b$- directions are of the same order. An intuitive picture here is that $V_\perp$ tends to "lock" more firmly the $2k_F$ charge configuration induced by $V'$, so that $2k_F$ charge fluctuations are enhanced, thereby stabilizing the spin-triplet $f$-wave state.

Bearing the above analysis in mind, we now move on to the RPA calculation results for the pairing symmetry competition between $f$- and $d$-waves. We first focus on the case where the parameter values satisfy the condition for $\chi_s(Q_{2k_F}) = \chi_s(Q_{2k_F})$, that is when $V' + V_\perp = U/2$ holds. Here we take $U = 1.7$, $V = 0.8$, $V' = 0.45$, $V'' = 0.2$, and $V_\perp = 0.4$ in units of $t$. Note that $V'$ is much smaller than $U/2$. As expected, the singlet pairing having the largest eigenvalue $\lambda$ has a $d$-wave gap, while the triplet pairing with the largest $\lambda$ has a $f$-wave gap, as seen in Fig.3. In Fig.4, we plot $\lambda$ as functions of temperature for $d$-wave and $f$-wave pairings. The two pairings closely compete with each other, but
The largest eigenvalue in the singlet and the triplet channels are plotted as functions of temperature for $U = 1.7$, $V = 0.8$, $V' = 0.45$, $V'' = 0.2$, and $V_\perp = 0.4$.

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Fig. 4. The largest eigenvalue in the singlet and the triplet channels are plotted as functions of temperature for $U = 1.7$, $V = 0.8$, $V' = 0.45$, $V'' = 0.2$, and $V_\perp = 0.4$.

To summarize, we have studied the pairing symmetry competition in a model for (TMTSF)$_2$X which considers not only the intrachain repulsions but also the interchain repulsion. We find that the possibility of satisfying the condition for realizing $f$-wave pairing becomes more realistic in the presence of the interchain repulsion. It would be an interesting future study to investigate whether this condition is actually satisfied in (TMTSF)$_2$X using first principles or quantum chemical calculations. Experimentally, it would be interesting to further confirm spin-triplet pairing by using probes complementary to those in the previous studies, for example, a phase sensitive tunneling spectroscopy study with or without applying a magnetic field, or those based on a newly developed theory for triplet superconductors, which has been proposed by one of the present authors.

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interaction part. The summation over the Stratonovich variables are taken by Monte-Carlo importance sampling. Using this method, correlation functions and susceptibilities can be calculated for finite size systems (16 sites in the $a$-direction and 4 sites in the $b$-direction=64 sites in the present study), and the results are exact within the statistical errors. A defect of this approach is that we cannot go down to very low temperatures in the presence of off-site repulsions such as $V$, $V'$ and $V_\perp$ due to the negative sign problem, so that it is difficult to look into the pairing symmetry competition itself. Nevertheless, we can check the validity of RPA at moderate temperatures of the order of $0.1t$. Here we compare the values of $\chi_f(Q_{2k_F})$ and $\chi_d(Q_{2k_F})$ calculated by AFQMC at $T = 0.25$, fixing $V = 0.9$, $V'' = 0$ and $V_\perp = 0.3$. In Fig.6, we show the “phase diagram” in $U - V'$ plane, where we find that the AFQMC boundary for $\chi_f(Q_{2k_F}) = \chi_d(Q_{2k_F})$ is very close to the RPA boundary $V' + V_\perp = U/2$. This result suggests that the RPA condition for $\chi_f(Q_{2k_F}) = \chi_d(Q_{2k_F})$ is reliable at least at moderate temperatures.
Fig. 6. AFQMC result for the competition between $\chi_s(Q_{2k_F})$ and $\chi_c(Q_{2k_F})$ shown in $U-V'$ space. $V = 0.9, V'' = 0, V_\perp = 0.3$, and $T = 0.25$. The dashed line represents the RPA condition for $\chi_s(Q_{2k_F}) = \chi_c(Q_{2k_F})$.

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