Competition between triplet, singlet, and FFLO states in organic superconductors (TMTSF)$_2$X under magnetic field

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Abstract. We study the competition between triplet $f$-wave, singlet $d$-wave, and the FFLO state in a model for (TMTSF)$_2$X using random phase approximation. The result suggests the possibility of singlet $\rightarrow$ FFLO $\rightarrow$ triplet transition upon increasing the magnetic field. The singlet $\rightarrow$ FFLO $\rightarrow$ triplet transition can appear in a large parameter regime on the space of the magnetic field and the off-site repulsions which induce the coexistence with $2k_F$ charge and $2k_F$ spin fluctuations, although decreasing the off-site repulsions suppresses the spin triplet $f$-wave pairing. We also show that the triplet component mixing increases in the FFLO state with increasing the magnetic field, and the off-site repulsions increase the mixing rate. The results seem to suggest that the large triplet mixing results in an enhanced tendency toward the FFLO state over the usual pairing state.

1. Introduction

FFLO (Fulde-Ferrell-Larkin-Ovchinnikov) state [1, 2] is one of the most fascinating superconducting states. In the FFLO state, superconductivity occurs as a non-uniform state since the Cooper pairs have a finite center of mass momentum as $(k+Q_c \uparrow, -k+Q_c \downarrow)$. Generally, when the symmetry breaking in the degree of freedom for space, spin or time takes place, spin singlet pairing and spin triplet with $S_z = 0$ pairing can be mixed. In fact, Matsuo et. al. have shown that mixing of the singlet and the triplet pairing because of the symmetry breaking in the spin space stabilizes the FFLO state. [3] Recent microscopic studies on the Hubbard model have shown that the triplet pairing component mixes in the FFLO state of the spin fluctuation mediated $d$-wave superconductivity. [4, 5]

As for the pairing competition in the quasi one dimensional (Q1D) organic superconductors (TMTSF)$_2$X (X=PF$_6$, ClO$_4$, etc.), some experiments suggest the possibility of spin triplet pairing and/or the FFLO state in the large magnetic field regime. [6, 7, 8, 9] We have previously shown that spin triplet $f$-wave pairing can compete with spin singlet $d$-wave pairing, where $d$-wave and $f$-wave in this study are shown in Figure 1 (b). [10, 11, 12] This is because (i) the $f$-wave and the $d$-wave gap has the same number of nodes intersecting the Fermi surface, and (ii) competing $2k_F$ spin and $2k_F$ charge fluctuations results in a nearly similar magnitude of the singlet and triplet pairing interactions. Recently, we have further found that the triplet pairing mediated by $2k_F$ spin+$2k_F$ charge fluctuations is strongly enhanced by the magnetic field, which
supports the possibility of singlet-triplet transition in (TMTSF)$_2$X under magnetic field. [13] The possibility of the triplet pairing has also been suggested by other studies. [14, 15, 16, 17, 18, 19] On the other hand, several studies have shown the possibility of the FFLO state in the Q1D model for (TMTSF)$_2$X. [20, 21]

In the present study, we investigate the possibility of the FFLO state from a microscopic point of view by applying random phase approximation (RPA) to a model for (TMTSF)$_2$X. We find a close competition between triplet, singlet, and the FFLO states, and propose the possibility of singlet→FFLO→triplet transition upon increasing the magnetic field. We also show that the mixing rate of the triplet with $S_z = 0$ pairing component in the FFLO state strongly increases with the increase of the magnetic field. It seems that the increase of the triplet component results in an enhanced tendency toward the FFLO state over the usual pairing state.

2. Formulation
The extended Hubbard model taking into account the Zeeman effect is given as

$$H = \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + \sum_i U n_{i\uparrow} n_{i\downarrow} + \sum_{i,j,\sigma,\sigma'} V_{ij} n_{i\sigma} n_{j\sigma'} + h_z \sum_{i,\sigma} \text{sgn}(\sigma) c_{i\sigma}^\dagger c_{i\sigma},$$

(1)

where $c_{i\sigma}^\dagger$ creates an electron with spin $\sigma$ at the $i$-th site. Here, as a model for (TMTSF)$_2$X, we consider the extended Hubbard model at 1/4-filling with the parameters shown in Figure 1 (a). The hopping parameter $t_{ij}$ is taken as $t_x$ and $t_y$. $U$ is the on-site repulsion and the off-site repulsions $V_{ij}$ are taken as $V_x$, $V_{x2}$ and $V_{x3}$, which are 1st, 2nd and 3rd nearest neighbor intra-chain interactions, and $V_y$ is the interchain interaction. $h_z$ is the Zeeman energy, where we consider the magnetic field parallel to the conductive plane, thus we ignore the orbital effect and assume a sufficiently large Maki parameter.

Within RPA, the effective pairing interactions for the opposite spin and equal spin pairing mediated by spin and charge fluctuations are given as

$$V_{bab}^\sigma (k) = U + V (k) + \frac{U^2}{2} \chi_{sp}^{zz} (k) - \frac{[U + 2V (k)]^2}{2} \chi_{ch} (k),$$

(2)

$$V_{lad}^\sigma (k) = U^2 \chi_{sp}^{+-} (k),$$

(3)

$$V_{bab}^\sigma (k) = V (k) - 2[U + V (k)] V (k) \chi^{\sigma\sigma} (k) - V (k)^2 \chi^{\sigma\sigma} (k) - [U + V (k)]^2 \chi^{\sigma\sigma} (k),$$

(4)

$$V_{lad}^\sigma (k) = 0,$$

(5)

where $V (k)$ is the Fourier transform of the off-site repulsions. The longitudinal spin and the charge susceptibility are obtained by $\chi_{sp}^{zz} (k) = \left[\chi^{\uparrow\uparrow} (k) + \chi^{\downarrow\downarrow} (k) - \chi^{\uparrow\downarrow} (k) - \chi^{\downarrow\uparrow} (k)\right]/2$ and $\chi_{ch} (k) = \left[\chi^{\uparrow\uparrow} (k) + \chi^{\downarrow\downarrow} (k) + \chi^{\uparrow\downarrow} (k) + \chi^{\downarrow\uparrow} (k)\right]/2$. Here, $\chi^{\sigma\sigma} (k)$ and $\chi^{\sigma\bar{\sigma}} (k)$ are obtained as

$$\chi^{\sigma\sigma} (k) = \frac{[1 + \chi_0^{\sigma\sigma} (k) V (k)] \chi_0^{\sigma\sigma} (k)}{[1 + \chi_0^{\sigma\sigma} (k) V (k)] [1 + \chi_0^{\bar{\sigma}\bar{\sigma}} (k) V (k)] - [U + V (k)]^2 \chi_0^{\sigma\sigma} (k) \chi_0^{\bar{\sigma}\bar{\sigma}} (k)},$$

(6)

$$\chi^{\sigma\bar{\sigma}} (k) = \frac{-\chi_0^{\sigma\sigma} (k) [U + V (k)] \chi_0^{\bar{\sigma}\bar{\sigma}} (k)}{[1 + \chi_0^{\sigma\sigma} (k) V (k)] [1 + \chi_0^{\bar{\sigma}\bar{\sigma}} (k) V (k)] - [U + V (k)]^2 \chi_0^{\sigma\sigma} (k) \chi_0^{\bar{\sigma}\bar{\sigma}} (k)}. $$

(7)

The transverse spin susceptibility is given as $\chi_{sp}^{+-} (k) = \chi_0^{+-} (k)/[1 - U \chi_0^{+-} (k)]$, where we ignore the effect of the off-site repulsions on the transverse spin susceptibility for simplicity. The longitudinal and transverse bare susceptibilities are given as

$$\chi_0^{\sigma\sigma} (k) = -\frac{1}{N} \sum_q \frac{f (\xi_\sigma (q + k) - f (\xi_\sigma (q))}{\xi_\sigma (q + k) - \xi_\sigma (q)},$$

(8)

$$\chi_0^{+-} (k) = -\frac{1}{N} \sum_q \frac{f (\xi_\sigma (q + k)) - f (\xi_\sigma (q))}{\xi_\sigma (q + k) - \xi_\sigma (q)},$$

(9)

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where $\xi_\sigma(k)$ is the single electron dispersion that considers the Zeeman effect measured from the chemical potential $\mu$, and $f(\xi_\sigma(k))$ is the Fermi distribution function.

To obtain the superconducting state and see the competition between the different pairing states, we solve the linearized gap equation (within the weak-coupling theory) which takes into account the center of mass momentum $Q_c$

$$\lambda_{Q_c}^{\sigma\sigma'} \phi^{\sigma\sigma'}(k) = \frac{1}{N} \sum_{k'} V^{\sigma\sigma'}(k,k') \left[ f(\xi_\sigma(k+Q_c)) - f(-\xi_\sigma(-k'+Q_c)) \right] \phi^{\sigma\sigma'}(k'),$$

where $V^{\sigma\sigma'}(k,k')$ is the pairing interactions as $V^{\sigma\sigma'}(k,k') = \left[ V_{bub}^{\sigma\sigma'}(k-k') + V_{lad}^{\sigma\sigma'}(k+k') \right]$ using Eqs. (2)-(5) and $\phi^{\sigma\sigma'}(k)$ is the gap function. As for the opposite spin pairing, we define the gap functions as

$$\phi^{\downarrow}(k) = \left[ \phi^{\uparrow\uparrow}(k) - \phi^{\downarrow\uparrow}(k) \right] / 2, \quad \phi^{\downarrow}(k) = \left[ \phi^{\uparrow\downarrow}(k) + \phi^{\downarrow\downarrow}(k) \right] / 2.$$

The critical temperature $T_c$ is determined as the temperature where the eigenvalue $\lambda_{Q_c}^{\sigma\sigma'}$ reaches unity. Although RPA may be considered as quantitatively insufficient for discussing the absolute value of $T_c$, we expect this approach to be valid for studying the competition between different pairing symmetries by using $\lambda_{Q_c}^{\sigma\sigma'}$ on the each pairing channels.

3. Results

Before proceeding to the calculation results, let us summarize the findings in our previous studies. We have found that the triplet $f$-wave pairing can dominate over the singlet $d$-wave pairing when the $2k_F$ charge fluctuations are present as same order as the $2k_F$ spin fluctuations, where the $2k_F$ spin+$2k_F$ charge fluctuations arise satisfying the condition $V_{x2} + V_y \geq U/2$. [10, 11, 12] Even if $2k_F$ charge fluctuations are slightly smaller than $2k_F$ spin fluctuations such as unsatisfying the condition $V_{x2} + V_y \geq U/2$, applying the magnetic field strongly enhances the spin triplet $f$-wave pairing with $S_z = 1$ due to $2k_F$ spin+$2k_F$ charge fluctuations, the triplet pairing still has a chance of taking place for large magnetic field, [13] although the magnetic field enhancement of the triplet $f$-wave with $S_z = 1$ disappear when the $2k_F$ spin fluctuations are strongly dominant and the $2k_F$ charge fluctuations are sufficiently small as $V_{x2} + V_y \ll U/2$. Bearing these in mind, we move on to the calculation results, where $t_x = 1.0$, $t_y = 0.2$ and $U = 1.7$ are fixed on the 1024×64 sites. Note that $\lambda_{Q_c}^{x}$ in the FFLO state takes maximum value with the $Q_c = (Q_{cx}, 0)$ and $\lambda_{Q_c}^{y}$ takes maximum value with the $Q_c = (0, 0)$ in this study. We call the opposite spin pairing with $Q_c = (0, 0)$ as the spin singlet(triplet with $S_z = 0$) pairing state when $\phi^{\uparrow}/\phi^{\downarrow} \gg 1(\phi^{\uparrow}/\phi^{\downarrow} \ll 1)$, the opposite spin pairing with $Q_c \neq (0, 0)$ as the FFLO state and the equal spin pairings with $\sigma = \uparrow(\sigma = \downarrow)$ as the spin triplet with $S_z = 1(S_z = -1)$ state.

The magnetic field dependence of the center of mass momentum $Q_{cx}$ on the maximum eigenvalue $\lambda_{Q_c}^{x}$ in the opposite spin pairing and the competition between the singlet pairing, triplet pairing with $S_z = 1$ and the FFLO state is shown in Figure 2 in the absence of the off-site repulsions, for which the $2F$ spin fluctuations are strongly dominant over the $2k_F$ charge fluctuations (Figure 2(a)) and for the case with off-site repulsions ($V_x, V_{x2}, V_{x3}, V_y$) = (0.9, 0.4, 0.1, 0.4), for which the $2F$ charge fluctuations are slightly smaller than the $2k_F$ spin fluctuations (Figure 2(b)). Note that the triplet pairing with $S_z = -1$ never dominates over the others so that it is omitted in Figure 2 and the triplet pairing with $S_z = 0$ is mixed in the FFLO state. In the case without the off-site repulsions, the spin singlet $d$-wave(SSd) dominates in the small field regime, but the FFLO state takes place with increasing the magnetic field. The case with the off-site interaction shows an interesting competition: the FFLO state again dominates over the SSd as the field is increased, but it gives way to the spin triplet $f$-wave with $S_z = 1(\text{STT}_{f^{1\uparrow}})$.
in the large field regime. We propose that singlet → FFLO → triplet transition with increasing the magnetic field may occur in the actual (TMTSF)$_2$X compounds since the coexistence of $2k_F$ spin-density-wave (SDW) and $2k_F$ charge-density-wave (CDW) is observed in the SDW phase adjacent to the superconducting phase in (TMTSF)$_2$PF$_6$. [22, 23]

Figure 1. (Color online) The model(a) and the $d$-wave(left) and $f$-wave(right) gaps(b) along with the Fermi surface, where red solid curves are the Fermi surface and the blue dashed lines represent the nodes of the gap. The arrow is the nesting vector.

Figure 2. (Color online) The $h_z$-dependence of $x$ component of $Q_c$(upper) and the $\lambda^{\sigma\sigma'}_{Q_c}$(lower) in the absence (a) and presence (b) of the off-site repulsions. SS$d$ is the spin singlet $d$-wave, ST$f^{\uparrow\downarrow}$ represents the spin triplet $f$-wave with $S_z = 1$.

Although the triplet pairing is suppressed with decreasing the off-site repulsions which induces the $2k_F$ spin+charge fluctuations, the singlet → FFLO → triplet transition appears with increasing the magnetic filed in the “pairing symmetry competition” phase diagram obtained by comparing the $\lambda^{\sigma\sigma'}_{Q_c}$ in the each pairings as seen in Figure 3. When $V_{x2} + V_y \simeq U/2$, the SDW and/or CDW appears in the large field regime since the presence of the off-site repulsions affects the contribution of the bubble-type bare susceptibility, which we have shown. [13]
Finally, we investigate the mixing of singlet and triplet with $S_z = 0$ pairings in the FFLO state. To see the difference between the absence and the presence of the off-site repulsions, the singlet and the triplet with $S_z = 0$ pairing component of the gap function in the FFLO state are shown for the two cases in Figure 4(a). Although the triplet component is already mixed with the singlet component even in the absence of the off-site repulsions, the triplet component mixing is strongly enhanced when the off-site repulsions are present. Figure 4(b) shows the ratio of $\lambda_{FFLO}$ and $\lambda_{SSd}$. It seems from this result that the large mixing of the triplet with $S_z = 0$ component results in an enhanced tendency toward the FFLO state over the usual pairing state.

Figure 3. (Color online) Pairing phase diagram in $(V_{x2} + V_{y}) - h_z$ space. A black solid curve represents the singlet→triplet transition line which we have obtained in Ref. [13].

Figure 4. (Color online) The $h_z$-dependence of (a) the mixing rate of the $S_z = 0$ triplet component $\phi_{STf^{11}} / \phi_{SSd}$ in the FFLO state, where ST$f^{11}$ denotes the spin triplet $f$-wave with $S_z = 0$, and (b) the $\lambda_{FFLO} / \lambda_{SSd}$. The red solid (blue dashed) curves are for the case in the presence (absence) of the off-site repulsions.

From the above results, we suggest the possibility of singlet→FFLO→triplet transition with increasing the magnetic field in (TMTSF)$_2$X compounds. As for the possible pairing state in the
actual compounds, we guess that the spin triplet pairing takes place more easily than the FFLO state in (TMTSF)$_2$PF$_6$ since 2$k_F$ spin+2$k_F$ charge fluctuations are present as suggested from the experimental observation of the coexisting 2$k_F$ SDW+2$k_F$ CDW. If 2$k_F$ charge fluctuations are smaller than 2$k_F$ spin fluctuations, which corresponds to the condition of $V_x^2 + V_y < U/2$, such as the absence of 2$k_F$ CDW coexisting with 2$k_F$ SDW, the singlet→FFLO(--triplet) transition with increasing the magnetic field arises within very small $V_x^2 + V_y$ parameter regime such as sufficiently small 2$k_F$ charge fluctuations. In (TMTSF)$_2$ClO$_4$, we may propose a possibility that 2$k_F$ charge fluctuations are smaller than 2$k_F$ spin fluctuations since the possibility of the FFLO state is suggested experimentally. Thus, we consider that the actual pairing state is determined by very subtle conditions in (TMTSF)$_2$X compounds.

4. Conclusion
To conclude, we have studied the competition between the triplet $f$-wave, singlet $d$-wave, and FFLO states in the model for (TMTSF)$_2$X. We have pointed out the possibility of the singlet→FFLO→triplet transition, and all of these states have a chance of taking place within a realistic parameters regime. We have also shown that the triplet with $S_z = 0$ component mixing rate increases in the FFLO state with increasing the magnetic field, and the presence of the off-site repulsions which induce the charge fluctuation further increases the mixing rate. We have also shown that the large triplet mixing results in an enhanced tendency toward the FFLO state over the usual pairing state.

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