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 the singlet→FFLO→triplet transition upon increasing the magnetic field. 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, the superconductivity occurs as the non-uniform state since the center of mass momentum is finite. When the symmetry breaking in spin space takes place, spin singlet pairing and spin triplet pairing can be mixed. In fact, Matsuo et. al. have shown that mixing of the singlet and the triplet pairing stabilizes the FFLO state. [3] Recent microscopic studies on the Hubbard model have shown that triplet pairing component mixes in the FFLO state of spin fluctuation mediated $d$-wave superconductivity. [4, 5]

Experiments for the quasi one dimensional (Q1D) organic superconductors (TMTSF)$_2$X (X=PF$_6$, ClO$_4$, etc.), 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 field, which supports the possibility of singlet-triplet transition in (TMTSF)$_2$X under magnetic field. [13]

In the present study, we investigate the possibility of the FFLO state from the microscopic point of view by applying random phase approximation(RPA) to a model for (TMTSF)$_2$X. We investigate the competition between triplet, singlet and the FFLO state, and propose the
possibility of singlet→FFLO→triplet transition upon increasing the magnetic field. We also show that the mixing rate of the triplet 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 \( V_1, V_2 \) and \( V_3 \), 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 conduction plane, thus we ignore the orbital effect.

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

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

(2)

\[ V_{\text{lad}}^\sigma (k) = U^2 \chi^\sigma_{sp} (k), \]

(3)

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

(4)

\[ V_{\text{lad}}^\sigma (k) = 0, \]

(5)

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

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

(6)

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

(7)

The transverse spin susceptibility is given as \( \chi^\sigma sp^{-} (k) = \chi^\sigma sp^{-} (k) / [1 - U \chi^\sigma sp^{-} (k)] \). The longitudinal and transverse bare susceptibilities are given as

\[ \chi^\sigma sp^{-} (k) = -\frac{1}{N} \sum_q \frac{\bar{f}(\xi_\sigma (q + k)) - \bar{f}(\xi_\sigma (q))}{\xi_\sigma (q + k) - \xi_\sigma (q)}, \]

(8)

\[ \chi^\sigma sp^{-} (k) = -\frac{1}{N} \sum_q \frac{f(\xi_\sigma (q + k)) - f(\xi_\sigma (q))}{\xi_\sigma (q + k) - \xi_\sigma (q)}, \]

(9)

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.

We solve the linearized gap equation within the weak-coupling theory,

\[ \chi^\sigma_{\sigma \sigma'} (k) = \frac{1}{N} \sum_{k'} V^\sigma_{\sigma \sigma'} (k, k') \frac{f(\xi_\sigma (k' + Q_c)) - f(-\xi_{\sigma'} (-k' + Q_c))}{\xi_\sigma (k' + Q_c) + \xi_{\sigma'} (-k' + Q_c)} \phi_{\sigma \sigma'} (k'), \]

(10)
where $V^{\sigma\sigma'}(k, k') = \left[V^{\sigma\sigma'}_{bb}(k - k') + V^{\sigma\sigma'}_{dd}(k + k')\right]$, $\phi^{\sigma\sigma'}(k)$ is the gap function and $Q_c$ is the center of mass momentum. As for the opposite spin pairing, we define the gap functions as
\[
\phi^{s}(k) = \left[\phi^{1\uparrow}(k) - \phi^{1\downarrow}(k)\right]/2, \quad \phi^{d}(k) = \left[\phi^{1\downarrow}(k) + \phi^{1\uparrow}(k)\right]/2.
\]
(11)

The critical temperature $T_c$ is determined as the temperature where the eigenvalue $\lambda^{\sigma\sigma'}$ 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 $\lambda$ 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 condition $V_2 + V_y \geq U/2$ is satisfied. [10, 11, 12] Applying the magnetic field strongly enhances the spin triplet $f$-wave pairing with $S_z = 1$, so that even if the condition is not satisfied, the triplet pairing still has a chance of taking place for large magnetic field.[13] 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 $1024 \times 64$ sites. Note that $\lambda$ in the FFLO state takes maximum value with the $Q_c = (Q_{xz}, 0)$ in this study.

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 repulsion (Figure 2(a)) and for the case with off-site repulsions ($V_1, V_2, V_3, V_y$) = (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. 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 $S_z = 1$ ($STf^{\uparrow\downarrow}$) 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$_6$.[14, 15]

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 pairing components of the gap in the FFLO state for the two cases are shown in Figure 3(a). Although the triplet component is mixed with the singlet component in the absence of the off-site repulsions, the triplet component mixing is strongly enhanced when the off-site repulsions are present. Figure 3(b) shows the ratio of $\lambda_{FFLO}$ and $\lambda_{SSd}$. It seems from this result that the large mixing of the triplet component results in an enhanced tendency toward the FFLO state over the usual pairing state.

4. Conclusion
To conclude, we have studied the competition between triplet $f$-wave, singlet $d$-wave, and the FFLO state 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 component mixing rate increases in the FFLO state with increasing the magnetic field, and the presence of off-site repulsions (thus the charge fluctuations) 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.
Figure 1. (a) The model and (b) the \( d \)-wave (left) and \( f \)-wave (right) gaps along with the Fermi surface. The arrows is the nesting vector.

Figure 2. The \( h_z \)-dependence of \( x \)-direction of \( Q_x \) (upper) and the \( \lambda \) (lower) in the absence of the off-site repulsions (a) and in the finite off-site repulsions (b). \( SSd \) is spin singlet \( d \)-wave, \( STf^{↑↓} \) is spin triplet \( f \)-wave with \( S_z = 1 \).

Figure 3. The \( h_z \)-dependence of (a) the \( \phi_{STf^{↑↓}}/\phi_{SSd} \) in the FFLO state, where \( STf^{↑↓} \) means spin triplet \( f \)-wave with \( S_z = 0 \), and (b) the \( \lambda_{FFLO}/\lambda_{SSd} \). The red solid (blue dashed) curves are the case in the presence (absence) of the off-site repulsions.

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