Strong parity mixing in the FFLO superconductivity in systems with coexisting spin and charge fluctuations

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We study the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state of spin fluctuation mediated pairing, and focus on the effect of coexisting charge fluctuations. We find that (i) consecutive transitions from singlet pairing to FFLO and further to $S_z = 1$ triplet pairing can generally take place upon increasing the magnetic field when strong charge fluctuations coexist with spin fluctuations, and (ii) the enhancement of the charge fluctuations lead to a significant increase of the parity mixing in the FFLO state, where the triplet/singlet component ratio in the gap function can be close to unity. We propose that such consecutive pairing state transition and strong parity mixing in the FFLO state may take place in a quasi-one-dimensional organic superconductor (TMTSF)$_2$X.

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The Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state, in which the Cooper pairs formed as $(k+Q, \uparrow, -k+Q, \downarrow)$ have a finite center of mass momentum, is one of the most fascinating superconducting states. [1, 2] One of the interesting aspects of the FFLO state is the parity mixing, i.e., even and odd parity pairings can be mixed. Phenomenological studies have shown that the mixing of the singlet and triplet pairings stabilizes the FFLO state. [3, 4] Recent microscopic studies have shown that the $S_z = 0$ triplet pairing is mixed with singlet pairing in the FFLO state of the Hubbard model on the two-leg ladder-type lattice, [5] and also on the square lattice, where $d$-wave superconductivity is mediated by spin fluctuations. [6, 7] In ref. [6] it has been pointed out that the parity mixing stabilizes the FFLO state, even in the vicinity of the quantum critical point where the quasiparticle lifetime becomes short due to the scattering by spin fluctuations.

Recent experimental indications of possible occurrence of the FFLO state in CeCoIn$_5$, [8] a quasi-two-dimensional (Q2D) organic materials such as $\lambda$-(BETS)$_2$FeCl$_4$ (BETS=bisethylenedithiotetraselenafulvalene) [9] and $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ (BEDT-TTF=bisethylenedithiotetrahiafulvalene), [10] and also in a quasi-one-dimensional(Q1D) one (TMTSF)$_2$ClO$_4$ (TMTSF=tetramethyltetraselenafulvalene) [11, 12] have stimulated extensive studies in this field. For (TMTSF)$_2$ClO$_4$ in particular, the possibility of the spin triplet pairing has previously been suggested experimentally for (TMTSF)$_2$X ($X=PF_6$, $CO_2$, $ClO_4$), [13-16] but a more recent NMR experiment by Shinagawa et al. [11] has revealed that when the magnetic field is low, the pairing occurs in the spin-singlet channel, while when the magnetic field is high, the pairing state is either an FFLO state or a spin-triplet state. Yonezawa et al. [12] have found that the onset $T_c$ exhibits a peculiar magnetic field dependence at high fields, which may be related to the occurrence of the FFLO state, where the direction of the total momentum of the Cooper pairs can play an important role. However, the microscopic origin of the transition between high field pairing states like the FFLO or the triplet state remains unexplored.

Theoretically, various studies have investigated the possibility of triplet pairing [17, 18, 19, 20, 21, 22] and the FFLO state [23, 24]. In particular, three of the present authors have previously shown that the triplet $f$-wave pairing can compete with the singlet $d$-wave pairing in the Q1D system because of the disconnectivity of the Fermi surface when $2k_F$ spin and $2k_F$ charge fluctuations coexist. [25, 26, 27] $2k_F$ spin+$2k_F$ charge fluctuations supported from the fact that diffuse X-ray scattering experiments observe the coexistence of $2k_F$ charge density wave(CDW) and the $2k_F$ spin density wave(SDW) in the vicinity of the superconducting phase in (TMTSF)$_2$PF$_6$. [28, 29] Moreover, we have recently found that this kind of triplet pairing due to $2k_F$ spin+$2k_F$ charge fluctuations is strongly enhanced by the magnetic field. [30]

Then a naive question arises along this line: what happens if magnetic field is applied to a system where spin singlet pairing dominates at zero field but triplet pairing is closely competing? If the FFLO state emerges, what is its nature?

Given this background, in the present Letter, we study the FFLO state of spin fluctuation mediated superconductivity in low dimensional systems, and focus on the effect of the charge fluctuations. We find that (i) consecutive transitions from singlet pairing to FFLO and further to $S_z = 1$ triplet pairing can generally take place upon increasing the magnetic field in the vicinity of the SDW+CDW coexisting phase, and (ii) the enhancement of the charge fluctuations leads to a significant increase of the parity mixing in the FFLO state, where the triplet/singlet component ratio in the gap function can
be close to unity. Based on a calculation on a model for (TMTSF)$_2$X, we propose that such consecutive pairing state transitions and the strong parity mixing in the FFLO state may actually be taking place in this material.

The anisotropic extended Hubbard model [Fig [1] (a)] that takes into account the Zeeman effect is given by

\[
H = \sum_{i,j} t_{ij} c_i^\dagger c_j + \sum_i U n_i n_{i+1} + \sum_{i,j} V_{ij} n_i n_j.
\]

(1)

Here \( t_{ij} = t_{ij} + h_z \sigma_i \delta_{ij} \), where the hopping \( t_{ij} \) is considered only for intrachain (\( t_x \)) and the interchain (\( t_y \)) nearest neighbors. \( t_x = 1.0 \) is taken as the energy unit. \( U \) is the on-site repulsion, and \( V_{ij} \) are the off-site repulsions: \( V_x, V_{x2}, V_{x3} \) are nearest, next nearest and 3rd nearest neighbor interaction within the chains, and \( V_y \) is the interchain interaction. We ignore the orbital effect, assuming that the magnetic field is applied parallel to the conductor \( x-y \) plane, thus we assume a sufficiently large Maki parameter.

The bare susceptibilities, bubble-type and ladder-type, are written as

\[
\chi_{\alpha\sigma}^0(k) = -\frac{1}{N} \sum_q \frac{f(\xi_\sigma(k+q)) - f(\xi_\sigma(q))}{\xi_\sigma(k+q) - \xi_\sigma(q)},
\]

(2)

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

(3)

where \( \xi_\sigma(k) \) is the band dispersion that takes into account the Zeeman effect measured from the chemical potential \( \mu \), and \( f(\xi) \) is the Fermi distribution function.

Within RPA that takes into account the magnetic field parallel to the spin quantization axis \( \hat{z} \), the longitudinal spin and charge susceptibilities are given as

\[
\chi_{\alpha\sigma}^{zz} = \frac{1}{2} (\chi^{\uparrow\downarrow} + \chi^{\downarrow\uparrow} - \chi^{\uparrow\uparrow} - \chi^{\downarrow\downarrow}),
\]

(4)

\[
\chi_{\alpha\sigma}^{xx} = \frac{1}{2} (\chi^{\uparrow\downarrow} + \chi^{\downarrow\uparrow} + \chi^{\uparrow\uparrow} + \chi^{\downarrow\downarrow}),
\]

(5)

The transverse spin susceptibility, in which we ignore the off-site repulsions for simplicity, is given as

\[
\chi_{\alpha sp}^{++}(k) = \frac{\chi_{\alpha--}^-(k)}{1 - U \chi_{\alpha 0}^0(k)}.
\]

(7)

The pairing interactions from the bubble and ladder diagrams are given as

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

(8)

\[
V_{lad}^{\sigma\sigma}(k) = \frac{U^2}{2} \chi_{sp}^{++}(k),
\]

(9)

\[
V_{bas}^{\sigma\sigma}(k) = V(k) - 2 [U + V(k)] V(k) \chi_{\tilde{\sigma}}^{\tilde{\sigma}}(k)
\]

\[
-V(k)^2 \chi_{\sigma}^{\sigma}(k) - [U + V(k)]^2 \chi_{\tilde{\sigma}}^{\tilde{\sigma}}(k).
\]

(10)

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

(11)

The linearized gap equation for Cooper pairs with the total momentum \( 2Q_c \) (\( Q_c \) represents the center of mass momentum) is given by

\[
\lambda_{Q_c}^{\sigma\sigma} \phi^{\sigma\sigma'}(k) = \frac{1}{N} \sum_q [V_{bas}^{\sigma\sigma}(k+q) + V_{lad}^{\sigma\sigma}(k+q)]
\]

\[
\times \mid f(\xi_{\sigma}(q_\pm)) - f(-\xi_{\sigma'}(-q_\pm)) \phi^{\sigma\sigma'}(q),
\]

(12)

where \( q_\pm = q \mp Q_c \), \( \phi^{\sigma\sigma'}(k) \) is the gap function and \( \lambda_{Q_c}^{\sigma\sigma'} \) is the eigenvalue of this linearized gap equation. The center of mass momentum \( Q_c \), which gives the maximum value of \( \lambda_{Q_c}^{\sigma\sigma'} \), lies along the \( x \)-direction because of the nesting of the Fermi surface [21] and \( \lambda_{Q_c}^{\sigma\sigma'} \) takes its maximum at \( Q_c = (0, 0) \) because the electrons do not scatter between the different directional spins in this pairing channel.

We define the singlet and the \( S_z = 0 \) triplet component of the gap function in the opposite spin pairing channel as

\[
\phi_{SS}^{0}(k) = |\phi^{\uparrow\downarrow}(k) - \phi^{\downarrow\uparrow}(k)|/2,
\]

\[
\phi_{ST}^{0}(k) = |\phi^{\uparrow\downarrow}(k) + \phi^{\downarrow\uparrow}(k)|/2.
\]

(13)

In our calculation, the spin singlet and the spin triplet component of the gap function in the FFLO state is essentially \( d \)-wave and \( f \)-wave as schematically shown in Fig [1] (b), so we write the singlet (\( S_z = 0 \) triplet) component of the FFLO gap \( \phi_{SS}^{0}(\phi_{ST}^{0}) \) in Eq. [13] as \( \phi_{SSd}^{0}(\phi_{STf}^{0}) \), where SSd(STf) stands for spin singlet \( d \)-wave (spin triplet \( f \)-wave with \( S_z = 0 \) pairing). The eigenvalue of each pairing state is determined as follows. \( \lambda_{Q_c}^{\sigma\sigma} \) with \( Q_c = (0, 0) \) gives the eigenvalue of the singlet \( d \)-wave pairing \( \lambda_{SSd}^{0} \) \( (S_z = 0 \) triplet \( f \)-wave pairing \( \lambda_{STf}^{0} \) \( \phi_{SSd}^{0} = 0 \) \( \phi_{STf}^{0} = 0 \), while \( \lambda_{Q_c}^{\sigma\sigma} \) with \( Q_c \neq (0, 0) \) gives \( \lambda_{FFLO} \). \( \lambda_{Q_c}^{\sigma\sigma} \) with \( Q_c = (0, 0) \) gives the eigenvalue for the spin triplet \( f \)-wave pairing with \( S_z = +1 \) \( (S_z = -1) \lambda_{STf}^{+1} \) \( (\lambda_{STf}^{-1}) \).

**FIG. 1:** (a) The model adopted in this study. (b) The schematic figure of the gap for \( d \)-wave(upper) and \( f \)-wave(lower), where the nodes of gap (blue dashed lines), the disconnected Fermi surface in Q1D lattice (red solid curves).

First, to make the argument general, we concentrate on a simple model with only the on-site \( U = 1.5 \) and the
nearest neighbor repulsion $V_z$ in the $x$-direction. When $V_z$ is large, $2k_F$ charge fluctuations tends to develop for band fillings close to half filling, so we take the band filling $n = 1.1$, where $n =$ number of electrons/number of sites. Here we fix the value of $t_y$ at 0.5, but this value does not have a specific meaning, and qualitatively (although not quantitatively) similar results can be obtained for other values of $t_y$. The temperature is fixed at $T = 0.01$ here. System size is taken as $2048 \times 64$ sites here.

In Fig. 2, we show the magnetic field dependence of $Q_{xx}$ of the FFLO state, the parity mixing ratio $\phi_{ST}/\phi_{SSd}$ and the eigenvalues of the gap equation for (a) $V_x = 0$ and (b) $V_x = 0.65$. Note that we denote the ratio between the maximum value of the $S_z = 0$ triplet component and that of the singlet component of the gap function in the FFLO state as “$\phi_{ST}/\phi_{SSd}$” hereafter. The dominating pairing state changes from singlet $d$-wave to FFLO upon increasing the magnetic field for both $V_x = 0$ and $V_x = 0.65$, but for sufficiently large field, FFLO further gives way to the triplet $f$-wave state with $S_z = 1$ for $V_x = 0.65$, i.e., when the charge fluctuations are present. The reason why $S_z = 1$ triplet $f$-wave dominates at high fields can be explained as follows. The presence of the charge fluctuations suppresses the spin singlet pairing interaction and enhance the triplet one. $^{24, 25, 26}$ Second, an $S_z = 1$ triplet pairing state induced by the coexistence of spin and charge fluctuations is strongly enhanced by the magnetic field applied parallel to the spin quantization axis $\hat{z}$. $^{30}$ In fact, such a possibility of transition from singlet pairing to FFLO, and further to triplet pairing upon increasing the magnetic field has been phenomenologically proposed by Shimahara. $^{18}$

![FIG. 2: (Color online) The $h_z$-dependence of $Q_{xx}$ (upper panels), the $S_z = 0$ triplet/singlet ratio in the FFLO $\phi_{ST}/\phi_{SSd}$ (middle) and the gap equation eigenvalues (lower) for (a) $V_x = 0$ and (b) $V_x = 0.65$. Other parameters are $U = 1.5$, $n = 1.1$, $t_y = 0.5$, and $T = 0.01$.](image)

Let us now look into the nature of the FFLO state. We show the $V_x$ dependence of the ratio $\phi_{ST}/\phi_{SSd}$ in Fig. 3. We see that the mixing ratio increases as $V_x$, namely, the charge fluctuation increases. The strong mixing of the triplet pairing component may be expected from the fact that the presence of charge fluctuations makes triplet pairing more competitive against singlet pairing. $^{20, 21}$

![FIG. 3: (Color online) The $V_x$-dependence of the ratio $\phi_{ST}/\phi_{SSd}$ and the ratio of the eigenvalues between the $S_z = 0$ triplet and the singlet on inset figure. Other parameters are the same with Fig. 2.](image)

We now move on to a realistic model for (TMTSF)$_2$X. We introduce not only $U$ and $V_x$, but also other distant off-site repulsions $V_{x2}$, $V_{x3}$ and $V_y$, since the $2k_F$ charge fluctuations, which becomes competitive against $2k_F$ spin fluctuations when the condition of $V_{x2} + V_{x3} \approx U/2$ is satisfied, are important in this material. $^{22, 23, 26, 30}$ Here, we fix the repulsions as $U = 1.7$, $V_x = 0.9$, $V_{x2} = 0.45$ and $V_{x3} = 0.1$, and vary $V_y$. Other parameters are taken as $t_y = 0.2$, $T = 0.012$, $n = 1.5$ (3/4 filling), and the system size is taken as $1024 \times 64$.

In Fig. 4, the center of mass momentum $Q_{xx}$, the parity mixing ratio $\phi_{ST}/\phi_{SSd}$ and the eigenvalues are plotted as functions of $h_z$ in the (a) absence ($V_x$, $V_{x2}$, $V_{x3}$ and $V_y = 0$) or (b) presence of the off-site repulsions, where for the latter case we set $V_y = 0.35$, for which $2k_F$ charge fluctuations are smaller than $2k_F$ spin fluctuations. As in the previous case for the simple model, FFLO dominates over singlet $d$-wave pairing upon increasing the magnetic field, and the FFLO state further gives way to the $S_z = 1$ triplet $f$-wave state in the presence of the off-site repulsions. As shown in the middle panels in Fig. 4, a strong mixing of singlet and $S_z = 0$ triplet pairing components takes place in the FFLO state, especially when the charge fluctuations are strong.

Finally, we show in Fig. 5 a phase diagram for the pairing competition in the off-site repulsion $V_y$ versus magnetic field $h_z$ space obtained by comparing the eigenvalues of the gap equation for each pairing channel. The size of the symbols denotes the magnitude of the eigenvalues, and “SDW+CDW” means that both spin
The phase diagram shows that for large enough $V_y$, the singlet→FFLO (with strong parity mixing)→triplet transition takes place upon increasing the magnetic field.

To conclude, we find that (i) consecutive transitions from singlet pairing to FFLO and further to $S_2=1$ triplet pairing can generally take place upon increasing the magnetic field in the vicinity of the SDW+CDW coexisting phase, and (ii) the enhancement of the charge fluctuations leads to a significant increase of the parity mixing in the FFLO state, where the triplet/singlet component ratio in the gap function can be close to unity.

We raise (TMTSF)$_2$X as a candidate material for such consecutive pairing state transition and strong parity mixing in the FFLO state to take place. In fact, as mentioned in the introductory part, the experiments suggest the presence of low field and high field pairing states, where the former occurs in the spin-singlet channel. As for the high magnetic field pairing state, Yonezawa et al. have shown that for a magnetic field parallel to the $a$ axis, only the clean sample exhibits an upturn of the $T_c$ curve in the high magnetic field regime above 4T, which suggests the presence of a pairing state sensitive to the impurity content. Between 4T and the Pauli limit of around 2.5T, there seems to be a different high field pairing state, in which superconductivity is stable against the impurities, but is very sensitive to the tilt of the magnetic field out of the $a$-$b$ plane. The bottom line of these experiments is that there may be three kinds of pairing states, i.e., one low field state, and two high field states. The correspondence between these experimental observations and the present study is not clear at the present stage, but the appearance of three kinds of pairing states is indeed intriguing. It would be interesting to further investigate experimentally the possibility and the nature of two kinds of high field pairing states.

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