Strong magnetic field enhancement of spin triplet pairing arising from coexisting $2k_F$ spin and $2k_F$ charge fluctuations

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We study the effect of the magnetic field (Zeeman splitting) on the triplet pairing. We show generally that the enhancement of spin triplet pairing mediated by coexisting $2k_F$ spin and $2k_F$ charge fluctuations can be much larger than in the case of triplet pairing mediated by ferromagnetic spin fluctuations. We propose that this may be related to the recent experiment for (TMTSF)$_2$ClO$_4$, in which a possibility of singlet to triplet pairing transition has been suggested.

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Spin triplet superconductivity is one of the most fascinating unconventional superconducting state. The investigation of the mechanism of such a pairing state has been a theoretical challenge. Spin triplet pairing mediated by ferromagnetic spin fluctuations has been studied from the early days in the context of superfluid $^4$He, but another possibility has arisen for the past several years: triplet pairing mediated by coexisting $2k_F$ spin and $2k_F$ charge (or orbital) fluctuations proposed for Sr$_2$RuO$_4$[1] and for organic superconductors (TMTSF)$_2$X (X=PF$_6$, ClO$_4$, etc.) [2-9] and by coexisting $2k_F$ spin and $2k_F$ charge fluctuations. Very recently, an NMR study on (TMTSF)$_2$ClO$_4$ has pointed out a possibility of a transition from a spin singlet pairing at low magnetic fields to triplet pairing or an FFLO state at high fields[20]. In fact, such a possibility of singlet to triplet pairing transition under high magnetic field was pointed out theoretically [21, 22, 23].

In the present Letter, we study the effect of the magnetic field (Zeeman splitting) on the triplet pairing. We show generally that the enhancement of spin triplet pairing mediated by coexisting $2k_F$ spin and $2k_F$ charge fluctuations can be much larger than in the case of triplet pairing mediated by ferromagnetic spin fluctuations. Applying the idea to a microscopic model for (TMTSF)$_2$X in which strong $2k_F$ spin and $2k_F$ charge fluctuations take place, we actually show that the magnetic field enhancement of spin triplet $f$-wave pairing is strong compared to the enhancement of ferromagnetic-spin-fluctuation mediated triplet pairing that occurs in a triangular lattice Hubbard model [24, 25]. Due to this strong effect, we show that even when spin singlet pairing dominates in the absence of the magnetic field, a transition to triplet pairing may take place by applying the magnetic field. This is consistent with the possibility of the magnetic field induced singlet-triplet transition in (TMTSF)$_2$ClO$_4$[20]. Moreover, this strong magnetic field effect may be used as a general probe for identifying the pairing mechanism of triplet superconductors.

The extended Hubbard Hamiltonian that takes into account the Zeeman effect is given as $H = \sum_{i,j,\sigma,\sigma'} t_{ij} c_{i\sigma}^\dagger c_{j\sigma'} + \sum_{i} U n_{i\uparrow} n_{i\downarrow} + \sum_{i,j,\sigma,\sigma'} V_{ij} n_{i\sigma} n_{j\sigma'} + g \mu_B B \sum_{i,\sigma} sgn(\sigma) c_{i\sigma}^\dagger c_{i\sigma}$, where $c_{i\sigma}$ creates an electron with spin $\sigma$ at the $i$-th site. $t_{ij}$ represents the hopping parameters, $U$ is the on-site interaction and $V_{ij}$ are the off-site interactions. $g \mu_B B$ is the Zeeman energy with the spin quantization axis $\hat{z} \parallel \mathbf{B}$. We ignore the orbital effect of the magnetic field.

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

$$V^s(\mathbf{q}) = U + V(\mathbf{q}) + \frac{U^2}{2} \chi^z_{sp}(\mathbf{q}) + U^2 \chi^\pm_{sp}(\mathbf{q}) - \frac{(U + 2V(\mathbf{q}))^2}{2} \chi_{ch}(\mathbf{q}),$$

for spin singlet pairing,

$$V^{t\sigma\sigma}(\mathbf{q}) = V(\mathbf{q}) - 2(U + V(\mathbf{q})) \chi^{\sigma\sigma}(\mathbf{q}) - \frac{V(\mathbf{q})^2}{2} \chi^{\sigma\sigma}(\mathbf{q}) - (U + V(\mathbf{q})) \chi^{\sigma\sigma}(\mathbf{q}) - \frac{(U + 2V(\mathbf{q}))^2}{2} \chi^{\sigma\sigma}(\mathbf{q}),$$

for spin triplet pairing with $\mathbf{d} \perp \hat{z}$ which means total $S_z = +1(-1)$ for $\sigma = \uparrow (\downarrow)$,

$$V^{t\sigma\sigma}(\mathbf{q}) = V(\mathbf{q}) + \frac{U^2}{2} \chi^{z\sigma}(\mathbf{q}) - U^2 \chi^\pm_{sp}(\mathbf{q}) - \frac{(U + 2V(\mathbf{q}))^2}{2} \chi_{ch}(\mathbf{q}),$$

for spin triplet pairing with $\mathbf{d} \parallel \hat{z}$ ($S_z = 0$), where $V(\mathbf{q})$ is the Fourier transform of the off-site interactions. The longitudinal spin susceptibility and the charge susceptibility are obtained by $\chi^z_{sp} = (\chi^{11} + \chi^{11} - \chi^{11} - \chi^{11}) / 2$.
and $\chi_{ch} = (\chi^{1+} + \chi^{1+} + \chi^{1+} + \chi^{1+})/2$. Here,
\[
\chi^{\sigma\sigma} = \frac{(1 + \chi^{\sigma\sigma} V_q) \chi^{\sigma\sigma}}{(1 + \chi_0^{\sigma\sigma} V_q) (1 + \chi_0^{\sigma\sigma} V_q) - (U + V_q)^2 \chi_0^{\sigma\sigma} \chi_0^{\sigma\sigma}}
\]
and
\[
\chi^{\sigma\overline{\sigma}} = \frac{-\chi^{\sigma\sigma} (U + V_q) \chi^{\sigma\overline{\sigma}}}{(1 + \chi_0^{\sigma\sigma} V_q) (1 + \chi_0^{\sigma\sigma} V_q) - (U + V_q)^2 \chi_0^{\sigma\sigma} \chi_0^{\sigma\overline{\sigma}}},
\]
where $V_q$ stands for $V(q)$. The longitudinal bare susceptibility is given as
\[
\chi_0^{\sigma\sigma}(q) = -\frac{1}{N} \sum_k f(\xi_\sigma(k + q)) - f(\xi_\sigma(k)),
\]
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. The transverse spin susceptibility is given as $\chi_0^{+\sigma\overline{\sigma}} = \chi_0^{\sigma\sigma}/(1 - U \chi_0^{\sigma\sigma})$, where the transverse bare susceptibility is obtained by
\[
\chi_0^{+\sigma\overline{\sigma}}(q) = -\frac{1}{N} \sum_{k'} V^{\mu}(k - k') \times \frac{1 - f(\xi_\sigma(k')) - f(\xi_{\overline{\sigma}}(k'))}{\xi_\sigma(k') + \xi_{\overline{\sigma}}(k')} \phi^{\mu}(k').
\]
We consider singlet and triplet pairings with $d \parallel \hat{z}$ ($\mu = s, t, \overline{t} \sigma$) for opposite spin pairing ($\sigma \neq \overline{\sigma}$), and triplet pairing with $d \perp \hat{z}$ ($\mu = t \sigma \overline{\sigma}$) for parallel spin pairing ($\sigma = \dagger, \dagger$). $\phi^{\mu}(k)$ is the gap function and the critical temperature $T_c$ is determined as the temperature where the eigenvalue $\lambda$ reaches unity. To give a reference for the values of the magnetic field, we calculate the Pauli limit by $\mu_B B_P = 1.75k_B T_c/\sqrt{2}$. Although RPA may be considered as a quantitative insufficiency for discussing the absolute value of $T_c$, we expect this approach to be valid for studying the competition between different pairing symmetries. In fact, as we shall see, we find very good agreement between the RPA results and the already known results obtained by dynamical cluster approximation (DCA) [22].

Before giving the calculation results, we show generally using the above formula that the effect of the Zeeman splitting on the triplet pairing caused by the coexistence of $2k_F$ spin and $2k_F$ charge fluctuations can be very special. First, let us consider a case where off-site repulsions are not present, so that only the spin fluctuations are relevant, and therefore possibility of triplet pairing superconductivity arises due to ferromagnetic spin fluctuations. In this case, the triplet pairing interactions reduce to $V^{t\sigma\overline{\sigma}}(q) = -U^2 \chi_0^{\overline{t}\sigma} \overline{\chi}_0^{\overline{t}\overline{\sigma}}$ and $V^{t\sigma\overline{\sigma}}(q) = +2U^2 \chi_0^{\overline{t}\sigma} \overline{\chi}_0^{\overline{t}\overline{\sigma}}(q) - U^2 \chi_0^{\sigma\overline{\sigma}}(q)$, where the formula for $\chi^{\sigma\overline{\sigma}}$ also reduces to
\[
\chi^{\sigma\overline{\sigma}} = \frac{\chi_0^{\overline{t}\sigma} \chi_0^{\overline{t}\overline{\sigma}}}{1 - U^2 \chi_0^{\overline{t}\sigma} \chi_0^{\overline{t}\overline{\sigma}}},
\]
Here, we assume without losing generality that $\chi_0^{\sigma\overline{\sigma}}$ is enhanced while $\chi_0^{\overline{t}\sigma}$ is suppressed by the magnetic field. (Whether $\sigma = \dagger$ or $\sigma = \dagger$ depends on the band structure and the band filling of the system as we shall see later.) In the first order of the magnetic field, $\chi_0^{zz} \overline{\chi}_0^{zz}$ and $\chi_0^{\overline{t}z} \overline{\chi}_0^{\overline{t}z}$ are not affected because exchanging $\dagger$ and $\dagger$ do not affect these quantities. $\chi^{\sigma\overline{\sigma}}$ is enhanced because the numerator $\chi_0^{\overline{t}\sigma}$ in eq. (9) is enhanced, but again the term in the denominator, $\chi_0^{\overline{t}\sigma} \chi_0^{\overline{t}\overline{\sigma}}$, is not affected by the magnetic field in its first order. Thus, although $V^{t\sigma\overline{\sigma}}$ should dominate over $V^{t\overline{\sigma}\sigma}$, its enhancement due to the Zeeman splitting occurs only through the direct enhancement of $\chi_0^{\overline{t}\overline{\sigma}}$, which may not be so large for realistic magnetic fields.

When a possibility of triplet pairing arises due to the coexistence of $2k_F$ spin and $2k_F$ charge fluctuations, where the latter are induced by off-site repulsions, the situation can change drastically. To make the discussion simple, let us consider a case with $-(U + 2V(Q_{2k_F})) \simeq U$, or equivalently $U + V(Q_{2k_F}) \simeq 0$, for which, in the absence of magnetic field, $\chi_0^{zz}(Q_{2k_F}) = \chi_0^{zz}(Q_{2k_F})$, and thus $V^{t\sigma\overline{\sigma}}(Q_{2k_F}) \simeq V^{t\overline{\sigma}\sigma}(Q_{2k_F}) \simeq -V^s(Q_{2k_F})$. (We shall see later that our idea works for more general cases). Here, the singlet and the triplet pairing interactions have nearly the same absolute values because the $2k_F$ spin and the $2k_F$ charge contributions work constructively (destructively) in the spin triplet (singlet) pairing interaction $[2, 4, 5, 6, 7]$. In this case, $\chi^{\sigma\overline{\sigma}}$ at $q = Q_{2k_F}$ can be given by a reduced form
\[
\chi^{\sigma\overline{\sigma}}(Q_{2k_F}) \simeq \frac{\chi_0^{\overline{t}\sigma}(Q_{2k_F})}{1 + V(Q_{2k_F}) \chi_0^{\overline{t}\overline{\sigma}}(Q_{2k_F})}.
\]
Here again, we assume without losing generality that $\chi_0^{\sigma\overline{\sigma}}(Q_{2k_F})$ ($\chi_0^{\overline{t}\sigma}(Q_{2k_F})$) is enhanced (suppressed) by the Zeeman splitting. $\chi^{zz}$ and $\chi^{\overline{t}z}$ again are not affected by the magnetic field in its first order, so $V^s$ and $V^{t\overline{\sigma}\sigma}$ are unaffected, while $\chi^{\overline{t}\overline{\sigma}}$ is again affected. The difference from the case with ferromagnetic spin fluctuations lies in that since $V(Q_{2k_F}) < 0$, the denominator of eq. (10) decreases as $\chi_0^{\sigma\overline{\sigma}}$ increases. The enhancement of $\chi^{\sigma\overline{\sigma}}$ due to this effect can be very large in the vicinity of $2k_F$ CDW ordering because $1 + (U + 2V(Q_{2k_F})) \chi_0(Q_{2k_F}) = 0$ signals this ordering, which is the same as $1 + V(Q_{2k_F}) \chi_0(Q_{2k_F}) = 0$ when $U + V(Q_{2k_F}) = 0$. Therefore, when the possibility of triplet pairing arises in the vicinity of coexisting $2k_F$ CDW and $2k_F$ SDW phases, triplet pairing with parallel spins can be strongly favored by the Zeeman splitting. In actual cases, superconductivity is usually degraded
by the orbital effect under magnetic fields, but even in that case, the enhancement of triplet pairing due to the above effect should make the suppression to be moderate. For quasi-1D systems in particular, where the additional node in the SC gap required in the triplet pairing does not intersect the Fermi surface (see Fig.1(b)), the coexistence of $2k_F$ spin and $2k_F$ charge fluctuations already results in a subtle competition between singlet and triplet pairings [4, 5], so that the strong enhancement of the triplet pairing interaction by the magnetic field may easily result in a singlet to triplet pairing transition.

We now apply the above idea to actual systems. First, we consider a case where the possibility of triplet pairing occurs due to ferromagnetic spin fluctuations induced by the on-site repulsion. As a typical example, we consider a case on a triangular lattice with dilute band filling as shown in Fig. 1(a). In this case, possibility of spin triplet $f$-wave pairing due to ferromagnetic spin fluctuations has been pointed out previously [24, 25].

The band dispersion is given as $\xi_\sigma(k) = 2t\cos k_x + 2t\cos k_y + 2t\cos(k_x + k_y) - \mu + g\mu_B B sgn(\sigma)$. We take the transfer energy as the unit of the energy, i.e., $t = 1.0$. The on-site interaction is $U = 3.0$, and the band filling is taken as $n = 0.2$. We take $128 \times 128$ $k$-point meshes in the RPA calculation. When the magnetic field is absent, we obtain $k_BT_c \approx 0.014$. The Pauli limit corresponding to this $T_c$ is $\mu_B B_F \approx 0.017$, which should be considered as a reference for the values of the magnetic field. When the Zeeman splitting is introduced, $\chi^{\uparrow\uparrow}_0$ becomes slightly larger than $\chi^\downarrow\downarrow_0$ because of the increase of the density of states (DOS) at the Fermi level of the down-spin due to the Zeeman splitting (see Fig.1(a)). Thus, this corresponds to the case with $\sigma = \downarrow$ in our general argument for the case with ferromagnetic fluctuations, so that we should focus on the enhancement of $\chi^{\downarrow\downarrow}_0$. In order to measure how the magnetic field enhancement of $\chi^{\downarrow\downarrow}_0$ is reflected to the enhancement of $\chi^{\uparrow\uparrow}_0$, we introduce a parameter $\alpha_\sigma(q, B) = \frac{\chi^{\sigma\sigma}(q, B)}{\chi^{\sigma\sigma}(q, 0)}$. In Fig. 2, $\alpha_\sigma$ at $\mu_B B = 0.03$ is plotted as a function of $q$. $\alpha_\parallel \approx 1$ means that the effect of the denominator in eq. (10) is small, as expected from our argument above. Effect of the magnetic field on the strength of the triplet pairing is shown in Fig. 3(a), where we plot the eigenvalues of the linearized gap equation for each triplet pairings. As expected, due to the enhancement in $\chi^{\downarrow\downarrow}_0$, triplet $f^{\uparrow\uparrow}$-wave dominates in the presence of the magnetic field. We note that this result for $\lambda$ closely resembles the field dependence of the pairing susceptibility calculated by DCA for the same system [27], suggesting the reliability of the present approach.

We now turn to the case where the possibility of triplet pairing arises due to the coexistence of $2k_F$ spin and $2k_F$ charge fluctuations. As a typical example, we consider the case of (TMTSF)$_2$X. [3, 4, 5]
We adopt a 3/4-filled Q1D extended Hubbard model as shown in Fig. 1(b). The band dispersion is given by
\[ \xi (\mathbf{k}) = 2t_x \cos k_x + 2t_y \cos k_y - \mu + g_1 B \sin(\sigma). \]
We take \( t_x = 1.0 \) as the unit of the energy, and \( t_y = 0.2 \).

As for the interaction parameters, we consider not only the on-site interaction \( U \) but also off-site interactions: nearest-neighbor (n.n.) interaction \( V_1 \), 2nd n.n. \( V_2 \), 3rd n.n. interaction \( V_3 \) in the \( x \)-direction and n.n. interaction \( V_{pl} \) in the \( y \)-direction, where the Fourier transformed off-site interaction is given as
\[ V(\mathbf{q}) = 2V_1 \cos(k_x) + 2V_2 \cos(2k_x) + 2V_3 \cos(3k_x) + 2V_{pl} \cos(k_y). \]
We set the on-site and the off-site interactions as \( U = 1.7, V_1 = 0.9 \) and \( V_2 = 0.1 \), and \( V_3 \) and \( V_{pl} \) are varied. The band filling is taken as \( n = 1.5 \) in accord with \((\text{TMTSF})_2\text{X}\). We take 256 \( \times \) 128 k-point meshes in the actual calculation.

We first consider the case with \( V_2 = 0.45 \), \( V_{pl} = 0.4 \), for which \( U + V(Q_{2k_F}) = 0 \) \((Q_{2k_F} \simeq (\pi/2, \pi))\) and thus singlet \( d \)-wave and triplet \( f \)-wave pairings are nearly degenerate in the absence of the magnetic field. As discussed in the previous studies \([4, 5, 6, 7]\), the degeneracy is because the triplet and singlet pairing interactions are nearly equal at \( Q_{2k_F} \), and the additional gap node in the \( f \)-wave pairing does not intersect the Fermi surface, so that the nodal structure on the Fermi surface is the same between \( d \) and \( f \), as shown in Fig. 1(b). Introduction of the Zeeman splitting enhances \( \chi^{\uparrow \uparrow}_0(Q_{2k_F}) \) and suppresses \( \chi^{\uparrow \downarrow}_0(Q_{2k_F}) \) because the up-spin band becomes close to half filling and the electron-hole symmetry is more restored. Thus, this corresponds to \( \sigma = \uparrow \) in our general argument for the case with coexisting \( 2k_F \) spin and \( 2k_F \) charge fluctuations, so we should look at the enhancement of \( \chi^{\uparrow \uparrow} \) due to the magnetic field. In Fig. 2(b), \( \alpha_\uparrow \) is plotted at \( \mu_B B = 0.03 \), which largely exceeds unity at \( Q_{2k_F} \) as expected from our previous argument that the denominator in eq. 10 becomes close to 0. In Fig. 3(b), we show the magnetic field dependence of the eigenvalue \( \lambda \). It can be seen that triplet \( f^{\uparrow \uparrow} \)-wave is strongly enhanced due to the strong enhancement of \( \chi^{\uparrow \uparrow}(Q_{2k_F}) \). In Fig. 4(c), we compare the \( B \) dependence of \( \lambda \) normalized by its value at \( B = 0 \) for the two cases. We can see that the enhancement of the triplet pairing mediated by coexisting \( 2k_F \) spin and \( 2k_F \) charge fluctuations is much larger.

In the above, we considered a case where \( \chi_{SP}(Q_{2k_F}) = \chi_{SP}(Q_{2k_F}) \) and thus the singlet \( d \)-wave and triplet \( f \)-wave pairings are nearly degenerate at \( B = 0 \), but even when singlet pairing dominates at \( B = 0 \) a transition to triplet pairing can take place within realistic values of \( B \) due to this strong enhancement of the triplet pairing interaction. To see this in detail, we consider a case with \( V_2 = 0.4 \), \( V_{pl} = 0.4 \), for which \( d \)-wave dominates over \( f \)-wave for \( B = 0 \). Here, \( T_c = 0.012 \), which corresponds to the Pauli limit of \( \mu_B B \sim 0.015 \). We obtain in Fig. 4(a) a pairing “phase diagram” in the \((V_2 + V_{pl}) - B\) space obtained by comparing \( \lambda \) at \( k_B T = 0.012 \). The phase diagram for the superconducting state (spin singlet \( d \)-wave and triplet \( f^{\uparrow \uparrow} \)-wave) and the normal state in the temperature-magnetic field space is shown in Fig. 4(b).

Applying the magnetic field, the spin singlet \( d \)-wave (indicated by SC-SSd) gives way to the spin triplet \( f \)-wave pairing with \( S_z = +1 \) (SC-STMf). Note that the \( T_c \) in the spin triplet \( f^{\uparrow \uparrow} \)-wave channel increases with \( B \) because we ignore the orbital effect. We expect that the orbital effect actually suppresses the \( T_c \), but even in that case, the effect of the Zeeman splitting should strongly favor the occurrence of triplet pairing over singlet pairing.

In conclusion, we have generally shown that the magnetic field enhancement of the spin triplet pairing due to the coexistence of \( 2k_F \) spin and \( 2k_F \) charge fluctuations can be extremely large compared to that mediated by ferromagnetic spin fluctuations. Thus, even when spin singlet pairing dominates in the absence of the magnetic field, a transition to spin triplet pairing can take place by applying (not unrealistically large) magnetic field. This is consistent with the possibility of the magnetic field induced singlet-triplet transition in \((\text{TMTSF})_2\text{ClO}_4\).

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