Spin-triplet *f*-wave-like pairing proposed for an organic superconductor $(TMTSF)_2PF_6$

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By examining how the spin- and/or charge-fluctuation exchange can contribute to pairing instabilities, we propose that a spin-triplet *f*-wave-like pairing with a *d*-vector perpendicular to the *b*-axis may be realized in $(TMTSF)_2PF_6$ due to (i) a quasi-one-dimensional Fermi surface, (ii) a coexistence of $2k_F$ charge fluctuations and spin fluctuations, and (iii) an anisotropy in spin fluctuations. Fluctuation-exchange study for the Hubbard model confirms the point (i), while a phenomenological analysis is given for (ii) and (iii). The proposed pairing is consistent with various experiments.

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I. INTRODUCTION

Spin-triplet pairing is conceptually fascinating, but there seem to be few examples. Recently an organic superconductor, $(TMTSF)_2PF_6$, has attracted much attention since a triplet pairing is suggested from an observation of large H_{c2}[1] as well as from a Knight shift experiment,[2] while the absence of Hebel-Slichter peak and the power-law decay of T_1^{-1} below T_c[2] suggest an anisotropic pairing with nodes in the gap.

If triplet pairing is indeed realized in $(TMTSF)_2PF_6$, its mechanism is a challenging theoretical puzzle: in the pressure-temperature phase diagram for this material the superconductivity lies right next to the $2k_F$ spin density wave (SDW), so that if one seeks an electronic origin, a spin-singlet *d*-wave-like pairing mediated by spin fluctuations is most naturally expected as proposed by several authors.[3–5] If, on the other hand, one assumes a phonon-mediated attractive interaction, triplet pairing, with nodes in the gap in general, would seem less favorable compared to singlet *s*-wave pairing without nodes. Recently, Kohmoto and Sato[6] have proposed that this difficulty in the phonon mechanism may be circumvented for TMTSF compounds, where they show that the quasi-one dimensionality of the Fermi surface, along with the presence of spin fluctuations, makes a triplet *p*-wave pairing without nodes on the Fermi surface[7] dominate over *s*-wave pairing. However, it is not clear if such a nodeless gap can be reconciled with the absence of Hebel-Slichter peak and the power-law T_1^{-1} in $(TMTSF)_2PF_6$.

If we turn to another prominent candidate for triplet superconductivity accompanied by SDW fluctuations,[8] Sr_2RuO_4, Takimoto recently proposed that *charge* fluctuations (or more precisely orbital fluctuations) should arise from repulsions between degenerate $4d$ orbitals, and that the coexistence of spin and charge fluctuations may lead to a triplet pairing.[9] This makes us recall an experimental fact that a $2k_F$ charge density wave (CDW) actually *coexists* with the SDW in $(TMTSF)_2PF_6$ as suggested from x-ray diffuse scattering.[10] In another theory for Sr_2RuO_4, Sato and Kohmoto,[11] and independently Kuwabara and Ogata,[12] have proposed that *anisotropy* of the spin fluctuations, known to be present experimentally,[13] may give rise to a triplet *p*-wave pairing. The anisotropy of spin fluctuations is also present in $(TMTSF)_2PF_6$.[14] Moreover, Sr_2RuO_4 has two quasi-1D Fermi surfaces (although they are weakly hybridized to result in two 2D Fermi surfaces), so the ruthenate seems to share several features with the TMTSF compound, although the strong charge fluctuation employed in Takimoto's mechanism is yet to be detected experimentally in Sr_2RuO_4.

However, the triplet pairing mechanism of $(TMTSF)_2PF_6$ cannot be the same with that of Sr_2RuO_4 since $\vec{d}$ (the *d*-vector characterizing the triplet pairing) $\perp \vec{z}$ (easy axis of the spins) is experimentally suggested in the former,[1,2] while $\vec{d} \| \vec{z}$ in the latter.[15] In the present paper, we propose that a triplet *f-wave-like* pairing with $\vec{d} \perp \vec{z}$ can take place in $(TMTSF)_2PF_6$ due to a *combination* of (i) the quasi-one-dimensionality of the Fermi surface, (ii) coexistence of $2k_F$ spin and charge fluctuations, and (iii) the anisotropy in the spin fluctuations. In the first part of the paper, we focus on how the quasi-one-dimensionality works favorably for the triplet pairing, and perform a fluctuation-exchange (FLEX)[16] calculation for the on-site repulsion Hubbard model on a lattice for $(TMTSF)_2PF_6$. Then, in the second part, we discuss phenomenologically how the triplet pairing can become competitive against the singlet when charge fluctuations coexist with spin fluctuations. We finally point out that the anisotropy in the spin fluctuations should further favor triplet pairing with $\vec{d} \perp \vec{z}$.

II. FORMULATION

We first consider the on-site Hubbard model, $\mathcal{H} = \Sigma_{\langle i,j \rangle \sigma} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma} + U \Sigma_i n_{i\uparrow} n_{i\downarrow}$, in the hole picture on a quasi-1D lattice ($|t_S| > |t_I|$) depicted in Fig. 1. There are $n = 0.5$ holes per site. Since sites A and B are inequivalent for $t_{S1} \neq t_{S2}$ and $t_{I1} \neq t_{I2}$ (dimerization of the molecules), we adopt the two-band version of the FLEX[18,19] (although we shall see that the dimerization is not essential in our argument).

For later discussions, we first recapitulate the one-band version of FLEX in a general fashion, where we proceed: (i) Dyson's equation is solved to obtain the renormalized Green's function $G(k)$, where k is a shorthand for the wave vector $\mathbf{k}$ and the Matsubara frequency, $i\epsilon_n$, (ii) the fluctuation-exchange interaction $V^{(1)}(q)$, given as,[20]

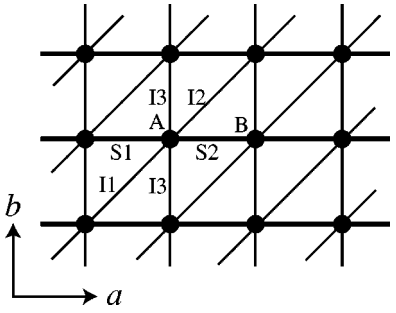

FIG. 1. The lattice considered with the hopping integrals taken to be $t_{S1}=-2.8$, $t_{S2}=-2.5$, $t_{I1}=+0.2$, $t_{I2}=+0.5$, $t_{I3}=-0.5$ in units of a typical energy scale, 100 meV, for organics after Ref. 17.

$$V^{(1)}(q)=\frac{1}{2}V_{\rm sp}^{zz}(q)+V_{\rm sp}^{+-}(q)+\frac{1}{2}V_{\rm ch}(q), \tag{1}$$

consists of the contribution from longitudinal (zz) and transverse $(+-)$ spin fluctuations (sp) and that from charge fluctuations (ch). For the on-site Hubbard model in particular, $V_{\rm sp}^{zz}=V_{\rm sp}^{+-}(\equiv V_{\rm sp})=U^2\chi^{\rm sp}$ and $V_{\rm ch}=U^2\chi^{\rm ch}$, where the spin and the charge susceptibilities are given as $\chi^{\rm sp}(q)=\chi^{\rm irr}(q)/[1-U\chi^{\rm irr}(q)]$ and $\chi^{\rm ch}(q)=\chi^{\rm irr}(q)/[1+U\chi^{\rm irr}(q)]$, respectively, using the irreducible susceptibility $\chi^{\rm irr}(q)=-1/N\Sigma_k G(k+q)G(k)$ (N: number of k-point meshes). (iii) $V^{(1)}$ then brings about the self-energy, $\Sigma(k)=1/N\Sigma_q G(k-q)V^{(1)}(q)$, which is fed back to Dyson's equation, and the self-consistent loop is repeated until convergence is attained.

T_c is the temperature where the eigenvalue λ of the following Éliashberg equation for the superconducting order parameter $\phi(k)$ reaches unity.

$$\lambda_\mu\phi_\mu(k)=-\frac{T}{N}\sum_{k'}\phi_\mu(k')|G(k')|^2V_\mu^{(2)}(k-k'). \tag{2}$$

Here, the pairing interaction $V_\mu^{(2)}(q)$ is given as

$$V_s^{(2)}(q)=\frac{1}{2}V_{\rm sp}^{zz}(q)+V_{\rm sp}^{+-}(q)-\frac{1}{2}V_{\rm ch}(q) \tag{3}$$

for singlet pairing,

$$V_{t\perp}^{(2)}(q)=-\frac{1}{2}V_{\rm sp}^{zz}(q)-\frac{1}{2}V_{\rm ch}(q) \tag{4}$$

for triplet pairing with total $S_z=\pm1$ $(\vec{d}\perp\vec{z})$, and

$$V_{t\|}^{(2)}(q)=\frac{1}{2}V_{\rm sp}^{zz}(q)-V_{\rm sp}^{+-}(q)-\frac{1}{2}V_{\rm ch}(q) \tag{5}$$

for triplet pairing with $S_z=0$ $(\vec{d}\|\vec{z})$. In the on-site Hubbard model, $V_{\rm sp}\gg V_{\rm ch}$ is satisfied, so that $|V_t^{(2)}|\simeq(1/3)|V_s^{(2)}|$ holds with $V_{t\|}^{(2)}=V_{t\perp}^{(2)}\equiv V_t^{(2)}$.

In the two-band version of FLEX, G, χ, Σ, and ϕ all become 2×2 matrices, whose elements are denoted as $G_{\alpha\beta}$ etc. with $\alpha,\beta=$A or B in the site representation, which may be converted to the band representation with a unitary transform. Since the Fermi surface lies in the lower band for quarter filling, we concentrate on Green's function and the order parameter in that band, denoted as G and ϕ_s,ϕ_t, respectively. As for the spin susceptibility, we diagonalize the 2×2 matrix $\chi^{\rm sp}$ and concentrate on the larger eigenvalue, denoted as χ. To ensure convergence at low temperatures in the two-band system we had to take 64×64 k points and ϵ_n from $-(2N_c-1)\pi T$ to $(2N_c-1)\pi T$ with N_c up to 8192.

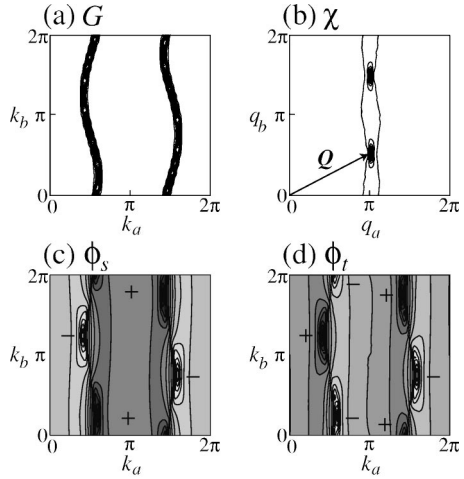

FIG. 2. Contour plots of $|G(\mathbf{k},i\pi k_BT)|^2$ (a), $\chi(\mathbf{q},0)$ (b), $\phi_s(\mathbf{k},i\pi k_BT)$ (c), and $\phi_t(\mathbf{k},i\pi k_BT)$ (d) for $n=0.5$, $U=8$, and $T=0.015$.

III. FLEX RESULTS FOR THE HUBBARD MODEL — ADVANTAGE OF THE QUASI-ONE DIMENSIONALITY

In Fig. 2, we show contour plots of $|G(\mathbf{k},i\pi k_BT)|^2$ (a), $\chi(\mathbf{k},0)$ (b), $\phi_s(\mathbf{k},i\pi k_BT)$ (c), and $\phi_t(\mathbf{k},i\pi k_BT)$ (d) for $T=0.015$. The Fermi surface as identified from the ridge in $|G(\mathbf{k})|^2$ is a pair of warped quasi 1D pieces. The spin susceptibility $\chi(\mathbf{q},0)$ has a peak at $\mathbf{Q}\simeq(\pi,\pi/2)$ (or $(\pi/2,\pi/2)$ in the unfolded Brillouin zone in the absence of dimerization), as expected from the nesting vector and in agreement with experimental results.[21,22] The singlet pairing order parameter is seen to change sign in such a way that (i) $\phi_s(\mathbf{k})=\phi_s(-\mathbf{k})$, and (ii) $\phi_s(\mathbf{k})$ has opposite signs across the nesting vector $\mathbf{Q}$ so that the repulsive $V_s^{(2)}(\mathbf{Q})$ [Eq. (3)] acts as an attractive interaction in the gap equation. We call the singlet pairing a ''d-wave'' in that the sign of $\phi_s(\mathbf{k})$ changes like $+-+-$ if we rotationally scan the Fermi surface, which is consistent with previous studies.[3–5]

For the triplet pairing, by contrast, $V_t^{(2)}(\mathbf{Q})$ is attractive [Eqs. (4) or (5)] with $V_{\rm sp}^{+-}=V_{\rm sp}^{zz}$), so that the order parameter should have the same sign across $\mathbf{Q}$. This requirement, along with the condition for a triplet order parameter $\phi_t(\mathbf{k})=-\phi_t(-\mathbf{k})$, can be satisfied by adding extra nodal lines along $k_a\sim0$ and $k_a\sim\pi$ (mod 2π). We call this pairing an '' f wave'' in that ϕ_s behaves this time like $+-+-+-$ along the Fermi surface.

A virtue of the quasi-one dimensionality is that the magnitudes, $|\phi_s(\mathbf{k})|$ and $|\phi_t(\mathbf{k})|$, are almost identical around the

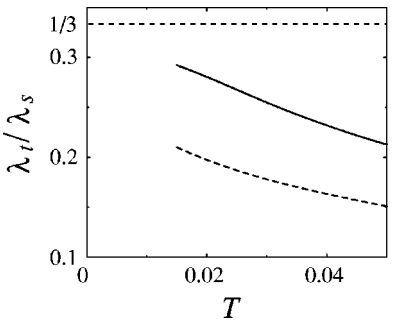

FIG. 3. λ_t/λ_s plotted as a function of temperature for the parameter values adopted in Fig. 2 (solid line), or for $t_{I2}=-t_{I3}=1.2$ (dashed line).

Fermi surface as seen from Figs. 2(a), 2(c), and 2(d). In fact, the ''f wave'' here is to ''d wave'' what p wave is to s wave in Kohmoto-Sato's picture[6] in that a singlet is converted into a triplet by introducing extra nodes that do not affect $|\phi(\mathbf{k})|$ on the Fermi surface. In such a situation, the difference between λ_s and λ_t comes almost entirely from the difference between $|V_s^{(2)}(\mathbf{Q})|$ and $|V_t^{(2)}(\mathbf{Q})|$ in the Éliashberg equation (2).

Our result shows that the ratio λ_t/λ_s indeed tends to 1/3 at low temperatures (Fig. 3; solid line), which reflects the ratio $|V_t^{(2)}(\mathbf{Q})/V_s^{(2)}(\mathbf{Q})|\simeq 1/3$ in the on-site Hubbard model. The ratio approaches to 1/3 as the temperature is lowered, because the ridge in $|G|^2$ sharpens so that the triplet pairing, with the order parameter vanishing around $k_a=0,\pi$, becomes more favorable. We can also confirm that quasi-1D is exploited in realizing $\lambda_t/\lambda_s\simeq 1/3$ by pushing the system toward 2D with larger value of t_{I2} and $|t_{I3}|$, for which the ratio λ_s/λ_t deviates from 1/3 even at low temperatures (Fig. 3; dashed line).

IV. EFFECT OF THE CHARGE FLUCTUATION

We have so far seen that the difference in λ between singlet ''d'' and triplet ''f'' can directly reflect the difference between $|V_s^{(2)}(\mathbf{Q})|$ and $|V_t^{(2)}(\mathbf{Q})|$ in a quasi-1D system. The ''f wave'' proposed here is an appealing candidate, because it can account for both experimentally suggested triplet pairing and the nodes in the superconducting gap. However, even for a quasi-1D system, ''f'' is only 1/3 competitive against ''d'' as far as the on-site repulsion Hubbard model is concerned—to make ''f'' dominate over ''d,'' we need to have $|V_t^{(2)}(\mathbf{Q})|>|V_s^{(2)}(\mathbf{Q})|$. So at this stage we depart from the Hubbard model to argue phenomenologically how some factors in the actual $(TMTSF)_2PF_6$ that are not taken into account in the simple Hubbard model can indeed make ''f'' competitive against ''d.''

An important experimental fact for $(TMTSF)_2PF_6$ that cannot be explained by the on-site Hubbard model is that a $2k_F$ CDW actually coexists with the $2k_F$ SDW.[10] The coexistence of spin and charge fluctuations can favor triplet pairing as pointed out for Sr_2RuO_4 by Takimoto mentioned above.[9] This can be seen in Eqs. (3) and (4), where an increase in $V_{\rm ch}$ enhances $|V_t^{(2)}|$ and suppresses $|V_s^{(2)}|$ (as far as $V_{\rm sp}>3V_{\rm ch}$ for isotropic spin fluctuations assumed for the time being). Now, if we take the coexistence of $2k_F$ SDW and $2k_F$ CDW in $(TMTSF)_2PF_6$ to be $V_{\rm sp}(\mathbf{Q})\simeq V_{\rm ch}(\mathbf{Q})$, Eqs. (3) and (4) dictate that $|V_t^{(2)}(\mathbf{Q})|\simeq|V_s^{(2)}(\mathbf{Q})|$, so ''$f$'' does indeed compete with ''d,'' but the competition is still subtle.

V. SPIN FLUCTUATION ANISOTROPY

Is there any mechanism that further favors the triplet pairing? Magnetic anisotropy is, in our view, one. It has actually been revealed experimentally for $(TMTSF)_2PF_6$ that the SDW has an easy axis in the b direction,[14] which implies that $V_{\rm sp}^{zz}(\mathbf{Q})>V_{\rm sp}^{+-}(\mathbf{Q})$ is satisfied for z taken to be $\|b$. In such a situation, the ''f'' pairing is more favorable in the $S_z=\pm 1$ channel since $|V_{t\perp}^{(2)}|>|V_{t\|}^{(2)}|$. The condition for ''$f$'' dominating over ''$d$'' now reads $|V_{t\perp}^{(2)}(\mathbf{Q})|>V_s^{(2)}(\mathbf{Q})$, or

$$V_{\rm ch}(\mathbf{Q})>V_{\rm sp}^{+-}(\mathbf{Q}) \tag{6}$$

from Eqs. (3) and (4). This last condition should be satisfied in $(TMTSF)_2PF_6$ because the spins do not order in the transverse direction even in the SDW phase, while the charges do. We stress that $\vec{d}\perp\vec{z}$ with $\vec{z}\|\vec{b}$ is consistent[23] with the experimental result: it is when the magnetic field is applied *parallel to the b-axis* that (i) H_{c2} becomes largest at low temperatures,[1] and (ii) the Knight shift is unchanged across T_c.[2]

The mechanism in which the anisotropy of the spin fluctuations favors triplet pairing is reminiscent of the one proposed in Refs. 11 and 12 for Sr_2RuO_4, but a crucial difference is that Refs. 11 and 12, which do not consider charge fluctuations, conclude a p-wave pairing with $S_z=0$ in agreement with the experimental results suggesting $\vec{d}\|\vec{z}$ in Sr_2RuO_4.[15] Let us see how this would occur in the present context. If $V_{\rm sp}^{zz}>(2V_{\rm sp}^{+-}+V_{\rm ch})$, we can see from Eq. (5) that $V_{t\|}^{(2)}(\mathbf{Q})$ becomes *repulsive*, which will mediate a triplet pairing having an order parameter with opposite signs across $\mathbf{Q}$. This requirement, along with the triplet condition $\phi_t(\mathbf{k})=-\phi_t(-\mathbf{k})$, can be satisfied by putting nodes only at $k_a\simeq 0$ and $k_a\simeq\pi$, thereby making the order parameter nodeless on the Fermi surface as in Refs. 6 and 7. Specifically, if $V_{\rm ch}/V_{\rm sp}^{zz}$ *and* $V_{\rm sp}^{+-}/V_{\rm sp}^{zz}$ are both sufficiently small, the pairing interactions $V_s^{(2)}$ (favoring ''d''), $V_{t\perp}^{(2)}$ (''f''), and $V_{t\|}^{(2)}$ (p) will all have similar magnitudes, so the p-wave pairing, with no nodes on the Fermi surface, should dominate over the others. Thus, the ''f'' pairing is not realized unless $V_{\rm ch}/V_{\rm sp}^{zz}$ is significant even if Eq. (6) is satisfied (see Fig. 4).

In this context, a possibly related problem is the pairing symmetry in $(TMTSF)_2ClO_4$, another candidate for a triplet superconductor. For this compound an NMR experiment suggests a presence of nodes on the Fermi surface,[24] while a recent thermal conductivity measurement suggests a nodeless gap.[25] If we adopt the latter result, the nodeless p-wave pairing should become a strong candidate. Then a compari-

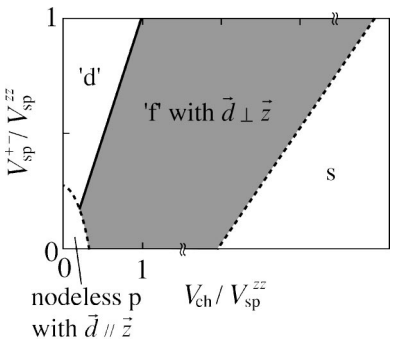

FIG. 4. A phase diagram against the charge/spin axis (V_{ch}/V_{sp}^{zz}) and the spin anisotropy axis (V_{sp}^{+-}/V_{sp}^{zz}). The solid line is according to Eq. (6), while dashed lines are schematic.

son of the magnitude of the charge fluctuation as well as the direction of $\vec{d}$ between $(TMTSF)_2ClO_4$ and $(TMTSF)_2PF_6$ will be a crucial test.

Having discussed the lower bound of V_{ch}/V_{sp}^{zz} for ''f'' pairing, how about an upper bound? When $V_{ch}/V_{sp}^{zz} \gg 1$, *singlet s-wave* pairing with $\phi_s(\mathbf{k}) \sim$ constant should enter as the dominant pairing. This is because $V_s^{(2)}$ becomes *attractive* for $V_{ch} > (V_{sp}^{zz} + 2V_{sp}^{+-})$, so that $\phi_s(\mathbf{k})$ no longer has to change sign. As $V_s^{(2)}/V_{t\perp}^{(2)}$ tends to unity with the increase of charge fluctuations, the ''f,'' with its nodes on the Fermi surface, should thus give way to the nodeless s. All the above reasoning is schematically summarized as a generic phase diagram in Fig. 4.

An additional bonus from the coexistence of strong spin/charge fluctuations and the anisotropic spin fluctuations is that these effects may serve to enhance the transition temperature for triplet pairing. Namely, a flaw in triplet superconductivity mediated by isotropic spin fluctuations is that the absolute value of the triplet pairing interaction $|V_t^{(2)}|$ is only one third of the effective interaction $V^{(1)}$ that determines the normal self-energy as seen from Eqs. (1) and (4) [or Eq. (5)] with $V_{sp}^{zz} = V_{sp}^{+-} \gg V_{ch}$. This is in contrast with the case of singlet pairing, where $V_s^{(2)}$ is nearly identical to $V^{(1)}$. Since a large self-energy correction results in a short quasi-particle lifetime, $V^{(1)}$ suppresses T_c while $V^{(2)}$ enhances it, and $V^{(1)} \simeq 3|V_t^{(2)}|$ in the Hubbard model generally results in a T_c, if any, too low to be detected in FLEX calculations.[26,27] This difficulty is resolved for large V_{ch} and/or small V_{sp}^{+-}, for which $|V_{t\perp}^{(2)}|$ approaches $V^{(1)}$.

VI. INTUITIVE PICTURE IN REAL SPACE

We can give an intuitive picture in real space for the singlet-triplet competition. In the presence of only $2k_F$ SDW, the spins are aligned in the a direction as shown in Fig. 5(a). On the other hand, it has been proposed that the coexisting $2k_F$-SDW-CDW phase should have a spin and charge alignment shown in Fig. 5(b).[28–30] The spin-charge alignment of a pure $2k_F$ CDW is given in Fig. 5(c). Given these spin-charge configurations, we now consider pairing in real space. If we neglect for simplicity the weak dimerization to unfold the Brillouin zone, the singlet ''d-wave'' order

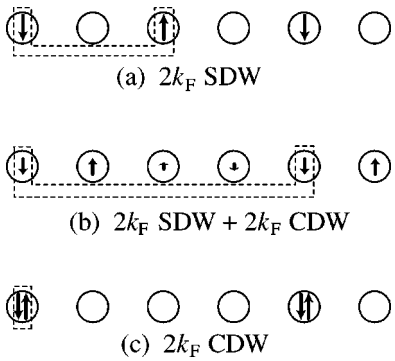

FIG. 5. An intuitive picture of pairing in real space at quarter filling. The arrows point the direction of the spin, while the length of the arrows represent the amount of charge. Cases for pure $2k_F$ SDW (a), coexisting $2k_F$ SDW and CDW (b), and pure $2k_F$ CDW (c) are shown. The pairs are depicted by dashed lines.

parameter will essentially have a $\cos(2k_a)$-like k_a-dependence [$\cos(k_a)$-like dependence with nodes at $k_a = \pi/2, 3\pi/2$ in the folded Brillouin zone], which roughly means that the singlet pairs are mainly formed between sites separated by two lattice spacings in the a direction. This is consistent with the alignment given in Fig. 5(a). Similarly, the triplet ''f-wave'' order parameter has a $\sin(4k_a)$-like k_a-dependence [$\sin(2k_a)$-like in the folded Brillouin zone], which implies that triplet pairs are formed between sites separated by four lattice spacings, which goes along with the configuration in Fig. 5(b). Finally, the singlet s-wave pairing with a constant $\phi_s(\mathbf{k})$ means on-site pairing, which is consistent with Fig. 5(c). We stress here that although the pairing mainly takes place in the a direction as suggested from the fact that the order parameter is nearly constant in the k_b direction, the two-dimensionality of the system is crucial in our mechanism since it is the warping of the Fermi surface that gives rise to the f-wave like sign change of the order parameter along the Fermi surface.

VII. SUMMARY AND FUTURE STUDY

To summarize, we have proposed a possible competition between singlet ''d-wave'' and triplet ''f-wave'' pairings in $(TMTSF)_2PF_6$ due to (i) the quasi-one-dimensionality of the Fermi surface, (ii) the coexistence of $2k_F$ spin and charge fluctuations, and (iii) the anisotropy in the spin fluctuations. The microscopic origin of the charge fluctuation remains to be identified. Since there is no orbital degeneracy in $(TMTSF)_2X$, the origin cannot be the one proposed by Takimoto for Sr_2RuO_4. In fact, the mechanism of $2k_F$ SDW-CDW coexistence in $(TMTSF)_2PF_6$ has been investigated by several authors. Some assume electron-lattice coupling,[28,29] while others envisage a purely electronic origin in terms of off-site repulsions up to second nearest neighbors.[30] It would be an interesting future problem to investigate microscopically the singlet-triplet competition and to evaluate T_c by taking these effects into account.

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