Phase diagram of the two-dimensional extended Hubbard model: Phase transitions between different pairing symmetries when charge and spin fluctuations coexist

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In order to explore how superconductivity arises when charge fluctuations and spin fluctuations coexist, we have obtained a phase diagram against the off-site repulsion V and band filling n for the extended, repulsive Hubbard model on the square lattice with the fluctuation exchange approximation. We have found the existence of (i) a transition between d_{xy} and $d_{x^2-y^2}$ pairing symmetries, (ii) f-pairing in between the $d_{x^2-y^2}$ and CDW phases for intermediate $0.5<n<1.0$ and large V, and (iii) for anisotropic cases the pairing symmetry changing, in agreement with a previously proposed "generic phase diagram," as $d\rightarrow f\rightarrow s$ when V (hence the charge fluctuations) are increased. All these are consequences of the structure in the charge and spin susceptibilities, which have peaks habitating at *common or segregated* positions in k space.

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I. INTRODUCTION

It is gradually becoming clearer that, while spin fluctuations are usually considered for electron mechanisms of high-Tc superconductivity, charge fluctuations may possibly play essential roles in some of the unconventional superconductors. Among these are organic metals such as θ-(BEDT-TTF)$_2X$ (Ref. 1) or (BEDT-TTF)$_3$Cl$_2$2H$_2$O (Ref. 2) that exhibit superconductivity sitting adjacent to the charge density wave (CDW) in the phase diagram. This suggests that charge fluctuations can mediate pairing as well, just as the spin-fluctuation-mediated pairing[3] can appear adjacent to the spin density wave (SDW) phase as in the cuprates and some of the organic superconductors like κ-(BEDT-TTF)$_2X$.

Studies for the pairing mediated by charge fluctuations have been rather scant. Scalapino *et al.* have shown, with the random phase approximation, that $d_{x^2-y^2}$ pairing gives way to d_{xy} in a three-dimensional cubic lattice when charge fluctuations become large with the introduction of the off-site interaction.[4] However, systematic studies are yet to come for the charge fluctuations, in contrast to the spin-fluctuation mediated superconductivity for which favorable situations for its occurrence has been extensively discussed.[5,6]

There is in fact one proposal in the context of the spin-triplet superconductivity in a quasi-one-dimensional (1D) organic metal (TMTSF)$_2$PF$_6$: Three of the present authors have proposed a "generic phase diagram" in which the dominant pairing symmetry changes as $d\rightarrow f\rightarrow s$ as the charge fluctuation becomes stronger.[7] The physical background is as follows: triplet pairing is very hard to be realized to start with, for the reasons identified in Ref. 6, where the primary one is the strength of the pairing interaction for triplets being only 1/3 of that for singlets. The situation can be reverted when charge fluctuations are dominant as discussed in Kuroki *et al.*,[7] but the charge fluctuation was treated phenomenologically there, so a microscopic study is highly desirable. Namely, while spin fluctuations dominate over charge fluctuations when the electron-electron interaction is short-ranged (as in the Hubbard model), charge fluctuations should become more intense as we increase the range of the interaction. The problem becomes especially intriguing when charge and spin fluctuations coexist, since they may induce quantum phase transitions among different pairing symmetries.

This is exactly our motivation here to study the effect of strong charge fluctuations by adopting the extended Hubbard model, as a simplest one in which we can control the relative magnitude of the charge fluctuation by varying the off-site Coulomb repulsion V. The extended Hubbard model has been studied, primarily for specific charge densities n, e.g., half-filling or quarter filling, by means of quantum Monte Carlo method,[8] weak coupling theory,[9] mean-field approximation,[10,11] second-order perturbation,[12] random phase approximation,[4] fluctuation exchange (FLEX) approximation,[13] slave-boson technique,[14] bosonization and renormalization group.[15,16] However, the phase diagram of the *two-dimensional extended Hubbard model against n and V* has yet to be obtained.

Here we have determined the symmetry of the dominant pairing in the V-n space for the extended Hubbard model by focusing on the case of isotropic or anisotropic square lattice, since many of unconventional superconductors are two-dimensional (2D) or quasi-one-dimensional (1D). We adopt the FLEX developed by Bickers *et al.*,[17–20] which is a renormalized perturbation scheme to study pairing instabilities when exchange of spin and charge fluctuations are considered as dominant diagrams. Although this is an approximation, we can explore the tendencies for pairing when the system parameters are varied.

We find that (i) there exists a phase transition between d_{xy} and $d_{x^2-y^2}$ pairing symmetries, (ii) triplet f-pairing appears in between the $d_{x^2-y^2}$ and CDW phases for intermediate $0.5<n<1.0$ and large V, and (iii) for anisotropic cases the pairing symmetry changes, in agreement with the generic phase diagram,[7] as $d\rightarrow f\rightarrow s$ when V (hence the charge fluctuations) are increased. Origin of all these has been identified as the structure in the charge and spin susceptibilities, which can have peaks habitating at segregated positions in k space.

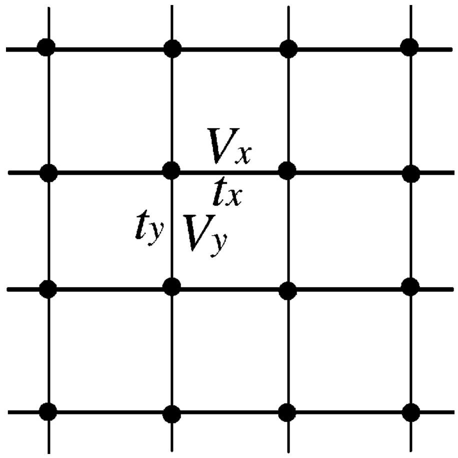

FIG. 1. A tetragonal lattice with hopping integral t_x along the x-axis and t_y along the y, along with the nearest-neighbor Coulomb repulsion V_x along the x-axis and V_y along the y.

II. FORMULATION

Let us start with the extended Hubbard Hamiltonian,

$$\mathcal{H} = -\sum_{i,j}^{\text{nn}} \sum_{\sigma} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow} + \frac{1}{2} \sum_{i,j}^{\text{nn}} \sum_{\sigma\sigma'} V_{ij} n_{i\sigma} n_{j\sigma'}, \tag{1}$$

in the standard notation on a tetragonal lattice depicted in Fig. 1. For the (isotropic) square lattice the unit of energy is taken to be the nearest-neighbor $t_{ij}=1.0$, and lattice constant $a=1$.

To determine the dominant gap function, we solve Éliashberg's equation with the FLEX approximation,

$$\lambda \phi(k) = -\frac{T}{N} \sum_{k'} \Gamma(k,k') G(k') G(-k') \phi(k'), \tag{2}$$

where ϕ is the gap function, G is Green's function, and Γ is the pairing interaction with $\mathbf{k} \equiv (k, \omega_n)$. The eigenvalue λ, a measure of the pairing, becomes unity at $T=T_C$. For the calculation we take an $N=32\times 32$ lattice, the temperature $T=0.02$, and the Matsubara frequency for fermions $-(2N_c-1)\pi T \leqslant \omega_n \leqslant (2N_c-1)\pi T$ with $N_c=1024$.

Esirgen *et al.*[13,21,22] have extended the FLEX method to general lattice Hamiltonians including the extended Hubbard model. Following them we introduce the pairing interaction,

$$\begin{aligned}\Gamma_{\rm s}(k,k') = \sum_{\Delta\mathbf{r},\Delta\mathbf{r}'} \Big\{ & \tfrac{3}{2}[V_m \chi_{\rm sp} V_m](k-k';\Delta\mathbf{r};\Delta\mathbf{r}') e^{i(\mathbf{k}\cdot\Delta\mathbf{r}+\mathbf{k}'\cdot\Delta\mathbf{r}')} \\ & -\tfrac{1}{2}[V_d \chi_{\rm ch} V_d](k-k';\Delta\mathbf{r};\Delta\mathbf{r}') e^{i(\mathbf{k}\cdot\Delta\mathbf{r}+\mathbf{k}'\cdot\Delta\mathbf{r}')} \\ & +\tfrac{1}{2} V_s(0;\Delta\mathbf{r};\Delta\mathbf{r}') e^{i(\mathbf{k}\cdot\Delta\mathbf{r}'-\mathbf{k}'\cdot\Delta\mathbf{r})} \Big\},\end{aligned} \tag{3}$$

for singlet pairing, and

$$\begin{aligned}\Gamma_t(k,k') = \sum_{\Delta\mathbf{r},\Delta\mathbf{r}'} \Big\{ & -\tfrac{1}{2}[V_m \chi_{\rm sp} V_m](k-k';\Delta\mathbf{r};\Delta\mathbf{r}') e^{i(\mathbf{k}\cdot\Delta\mathbf{r}+\mathbf{k}'\cdot\Delta\mathbf{r}')} \\ & -\tfrac{1}{2}[V_d \chi_{\rm ch} V_d](k-k';\Delta\mathbf{r};\Delta\mathbf{r}') e^{i(\mathbf{k}\cdot\Delta\mathbf{r}+\mathbf{k}'\cdot\Delta\mathbf{r}')} \\ & +\tfrac{1}{2} V_t(0;\Delta\mathbf{r};\Delta\mathbf{r}') e^{i(\mathbf{k}\cdot\Delta\mathbf{r}'-\mathbf{k}'\cdot\Delta\mathbf{r})} \Big\}\end{aligned} \tag{4}$$

for triplet pairing. Here $\Delta\mathbf{r}(=\mathbf{0}, \pm\hat{\mathbf{x}}, \pm\hat{\mathbf{y}})$ is null or nearest-neighbor vectors,

$$\chi_{\rm sp} = \overline{\chi}/(1 + V_m \overline{\chi}),$$

$$\chi_{\rm ch} = \overline{\chi}/(1 + V_d \overline{\chi})$$

are the spin and charge susceptibilities, respectively, where $\overline{\chi}$ is the irreducible susceptibility,

$$\overline{\chi}(q;\Delta\mathbf{r};\Delta\mathbf{r}') = -\frac{T}{N} \sum_{k'} e^{i\mathbf{k}'\cdot(\Delta\mathbf{r}-\Delta\mathbf{r}')} G(k'+q) G(k'), \tag{5}$$

and $V_d(V_m)$ is the coupling between density (magnetic) fluctuations,

$$V_d(q;\Delta\mathbf{r};\Delta\mathbf{r}') = \begin{cases} U + 4[V_x \cos(q_x) + V_y \cos(q_y)], & \Delta\mathbf{r} = \Delta\mathbf{r}' = \mathbf{0}, \\ -V_x, & \Delta\mathbf{r} = \Delta\mathbf{r}' = \pm\hat{\mathbf{x}} \\ -V_y, & \Delta\mathbf{r} = \Delta\mathbf{r}' = \pm\hat{\mathbf{y}} \end{cases} \tag{6}$$

$$V_m(q;\Delta\mathbf{r};\Delta\mathbf{r}') = \begin{cases} -U, & \Delta\mathbf{r} = \Delta\mathbf{r}' = 0, \\ -V_x, & \Delta\mathbf{r} = \Delta\mathbf{r}' = \pm\hat{\mathbf{x}} \\ -V_y, & \Delta\mathbf{r} = \Delta\mathbf{r}' = \pm\hat{\mathbf{y}}, \end{cases} \tag{7}$$

where $q \equiv (q, \epsilon_n)$ with $\epsilon_n = 2n\pi T$ being the Matsubara frequencies for bosons. We have found that the q dependence of V_m and V_d does not in fact affect Γ_s and Γ_t significantly. Accordingly the peak position of $\chi_{\rm ch}$ is almost the same as that for $V_d \chi_{\rm ch} V_d$ term in the expression for Γ.

$V_s(0;\Delta\mathbf{r};\Delta\mathbf{r}')$, $V_t(0;\Delta\mathbf{r};\Delta\mathbf{r}')$, appearing in the last lines in Eqs. (3) and (4), respectively, are constant terms involving U and V,

$$V_{\rm s}(q;\Delta\mathbf{r};\Delta\mathbf{r}') = \begin{cases} 2U, & \Delta\mathbf{r} = \Delta\mathbf{r}' = 0, \\ V_x, & \Delta\mathbf{r} = \Delta\mathbf{r}' = \pm\hat{x} \\ V_x e^{\pm iq_x}, & \Delta\mathbf{r} = -\Delta\mathbf{r}' = \pm\hat{x} \\ V_y, & \Delta\mathbf{r} = \Delta\mathbf{r}' = \pm\hat{y} \\ V_y e^{\pm iq_y}, & \Delta\mathbf{r} = -\Delta\mathbf{r}' = \pm\hat{y} \end{cases} \tag{8}$$

$$V_{\rm t}(q;\Delta\mathbf{r};\Delta\mathbf{r}') = \begin{cases} V_x, & \Delta\mathbf{r} = \Delta\mathbf{r}' = \pm\hat{x} \\ -V_x e^{\pm iq_x}, & \Delta\mathbf{r} = -\Delta\mathbf{r}' = \pm\hat{x} \\ V_y, & \Delta\mathbf{r} = \Delta\mathbf{r}' = \pm\hat{y} \\ -V_y e^{\pm iq_y}, & \Delta\mathbf{r} = -\Delta\mathbf{r}' = \pm\hat{y}. \end{cases} \tag{9}$$

When the off-site interaction V is introduced all the vertices (V_m, V_d, V_s, V_t) as well as the susceptibilities become $(Z+1)\times(Z+1)$ matrices for the lattice coordination number Z(=4 for the square lattice).

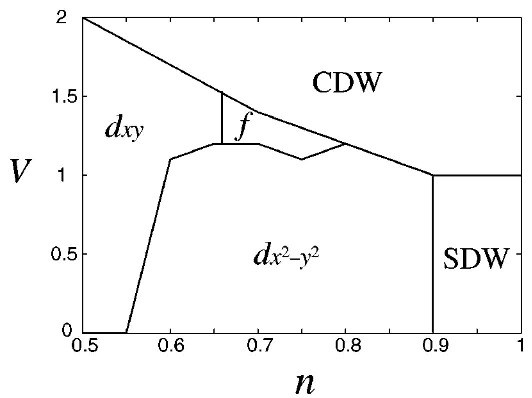

FIG. 2. Phase diagram against V and n with $U/t=4$ for the 2D extended Hubbard model.

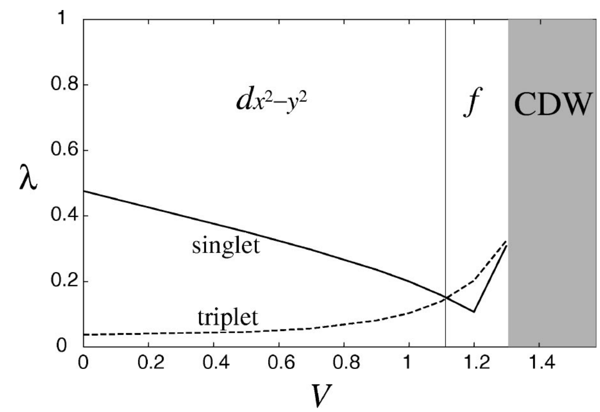

FIG. 3. The maximum eigenvalue, λ, of Éliashberg's equation for the spin-singlet (solid line) and triplet (dotted) channel as a function of V for $n=0.7$ with the dominant orbital symmetry indicated. The CDW phase is identified from the divergence in the charge susceptibility.

III. RESULT

A. Square lattice

Let us first display the obtained phase diagram against V and the band filling n for the square lattice in Fig. 2. The phase diagram is drawn by assuming that the pairing instability that has the largest λ in Éliashberg's equation, calculated at $T=0.02$ here, has the highest transition temperature, because, with the present complexity of the model, it is difficult to extend the FLEX calculation to lower temperatures. While the value of λ for $T=0.02$ is still much smaller than unity (see below), it is expected that the order in which the values of λ for various phases appear do not change for $T\to 0$. The density waves (CDW or SDW) are identified as the region where the respective (charge or spin) susceptibility diverges (at $T>0.02$ in the present calculation). The area of the CDW and SDW phases may expand at lower temperatures, but the phase diagram is not expected to change significantly.

Figure 2 is thus obtained, with the on-site Coulomb repulsion fixed at $U=4$ hereafter, and we immediately note that CDW, SDW, singlet superconductivity (SC), and triplet SC all appear on the (V,n) plane. The phase diagram is reminiscent of that for one-dimensional extended Hubbard model obtained with the Tomonaga-Luttinger theory.[23] However, an essential difference is that U or V has to be negative (attractive) to realize SC phases in one dimension, while we are talking about the case when both U and V are repulsive in 2D. Another comment is that in our calculation we cannot treat the Mott insulator, which should appear at n close enough to the half-filling $(n=1)$.

Before elaborating on the superconducting phases, let us make a remark on density waves. Intuitively, CDW should appear for strong enough V, while we should have SDW for the band filling close enough to the half-filling. The boundary between CDW and SDW for $n\to 1$ is seen in the present result to fall upon a line representing $V=1$, which agrees with a mean-field picture: In the CDW state where electrons doubly-occupy every other sites, each electron feels on average an on-site energy $U/2$ per electron, while in the SDW state each electron, singly-occupied, feels an off-site repulsion $V/2\times Z=2V$, so the SDW/CDW boundary corresponds to $V=U/4$.[8] As for the wave vectors describing the CDW (SDW) ordering, they are as indicated by the peaks (around (π,π)) in the charge (spin) susceptibilities, which are displayed below.

A salient feature for superconducting phases in Fig. 2 is that a spin-triplet f-phase appears just below the CDW phase for an intermediate region of n and V. The behavior of λ when V is varied with a fixed $n=0.7$ is depicted in Fig. 3.[24] Figure 4 shows the gap function in k space for the f-wave, which should be called, more precisely, Γ_5^- in the group theoretical representation. This has only one nodal line passing Γ point, but may be called f in that the gap function $(\sim\sin(k_x)+\sin(k_y)+\text{const}[\sin(2k_x)+\sin(2k_y)])$ changes sign as $+-+-+-$ along the Fermi surface.[25]

B. Physical origin: Common vs segregated peaks in χ_{sp} and χ_{ch}

We can keep track of the origin of the spin-triplet instability by looking at the structure (peak intensities and peak positions in k space) of the charge (χ_{ch}) and spin (χ_{sp}) susceptibilities (static with Matsubara frequency=0) in Fig. 5.

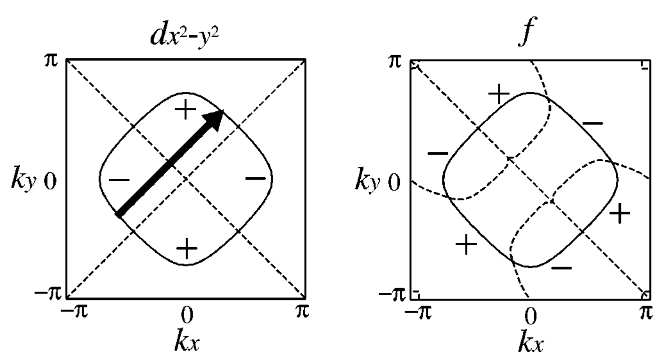

FIG. 4. The f-wave (irreducible representation Γ_5^-) gap function in k space (right panel) for $n=0.7$ and $V=1.3$, as compared with the $d_{x^2-y^2}$ (left) for $n=0.7$, $V=0.5$. Nodal lines are represented by dotted lines, while the Fermi surface by the solid curve on which the sign of the gap function is indicated. The arrow indicates a typical scattering process mediated by spin fluctuations.

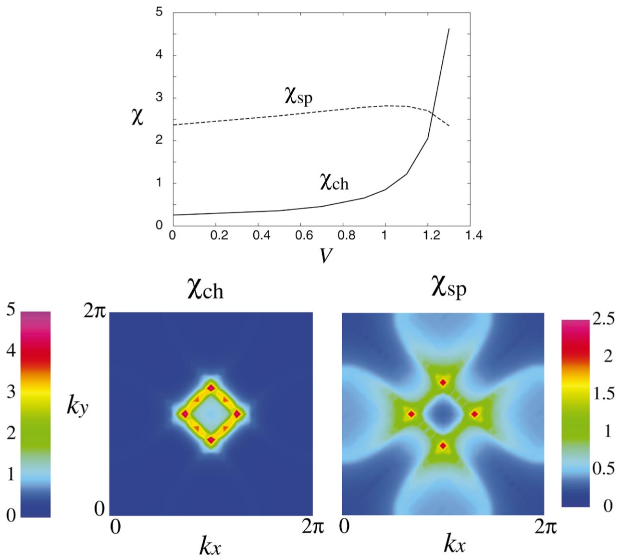

FIG. 5. (Color) (Top) The maximum eigenvalue of the charge (solid curve) and spin (dotted) susceptibilities as a function of V for $n=0.7$. (Bottom) Color-coded plots of the charge (left) and spin (right) susceptibilities in the (k_x, k_y) space for $V=1.3$ and $n=0.7$. The susceptibilities $[\sim(\text{energy})^{-1}]$ in units in which $t=1$.

The peak intensity of $\chi_{\rm ch}$ is seen to exceed that of $\chi_{\rm sp}$ as V is increased. If we turn to the structure in k space, $\chi_{\rm ch}$ and $\chi_{\rm sp}$ have similar peak positions. If we go back to Eqs. (3) and (4), the spin-fluctuation mediated pairing interaction (the first line on the right-hand side) and the charge-fluctuation mediated one (the second) act destructively (with opposite signs for the two terms in Eq. (3)) for singlet pairing, whereas they act *constructively* for triplet pairing (same signs in Eq. (4)). So we can expect the realization of triplet pairing ($|\Gamma_t|>|\Gamma_s|$) for large enough charge-fluctuation mediated pairing interactions (i.e., for large enough V) *provided* $\chi_{\rm ch}$ *and* $\chi_{\rm sp}$ *have common peak positions.*

Physically, $\chi_{\rm sp}=\overline{\chi}/(1-U\overline{\chi})$ while $\chi_{\rm ch}=\overline{\chi}/(1+U\overline{\chi})$ when $V=0$, so the nesting of the Fermi surface (which dominates $\overline{\chi}$) determines the peaks of $\chi_{\rm sp}$ (but *not* those of $\chi_{\rm ch}$). When V is switched on, $\chi_{\rm sp}=\overline{\chi}/(1+V_m\overline{\chi})$ and $\chi_{\rm ch}=\overline{\chi}/(1+V_d\overline{\chi})$ may have common peak positions, which is what is happening here.

How the situation is altered for smaller n for which d_{xy} pairing turns out to be dominant? In Fig. 6, which shows $\chi_{\rm ch}$ and $\chi_{\rm sp}$ for $n=0.6$ with $V=1.6$, $\chi_{\rm ch}$ has a larger peak than $\chi_{\rm sp}$

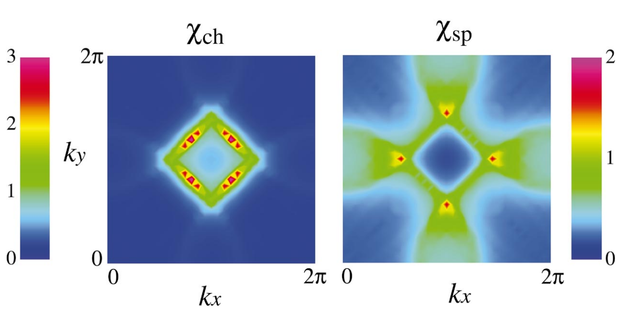

FIG. 6. (Color) A plot similar to Fig. 5, for $n=0.6$ with $V=1.6$.

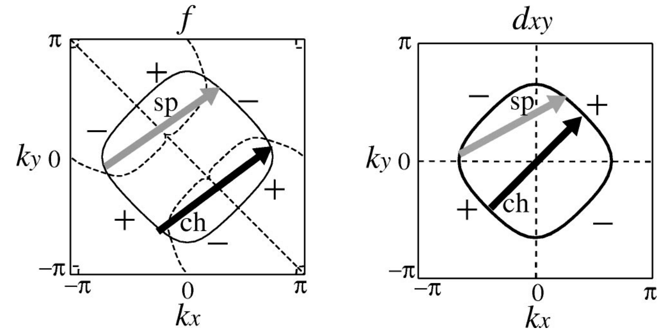

FIG. 7. The Fermi surface for $n=0.6$, $V=1.6$ with d_{xy} pairing (right), and for $n=0.7$, $V=1.3$ with the f pairing (left). The black (gray) arrows indicate typical scattering processes mediated by charge (spin) fluctuations.

does, but the peak positions of $\chi_{\rm ch}$ are *distinct* from those of $\chi_{\rm sp}$. Namely, while $\chi_{\rm ch}$ has peaks at four points around $\boldsymbol{Q}_{\rm ch} \simeq (\pm\pi, \pm\pi)$, $\chi_{\rm sp}$ has peaks that are shifted toward $\boldsymbol{Q}_{\rm sp} = (\pm\pi, 0)$ and $(0, \pm\pi)$. Now, as a basic property of Éliashberg's Eq. (2) with Eq. (3) plugged, to realize a large λ requires that (i) the gap function ϕ must change sign (across the typical pair-scattering momentum transfer $\boldsymbol{Q}_{\rm sp}$) to turn the originally repulsive spin-fluctuation-mediated interaction (i.e., the $V_m\chi_{\rm sp}V_m$ term with a positive coefficient, $3/2$ in Eq. (3)) into an effective attraction in the gap equation, while (ii) ϕ must not change sign (across the pair-scattering momentum transfer $\boldsymbol{Q}_{\rm ch}$) to make the originally attractive charge-fluctuation-mediated interaction (the $V_d\chi_{\rm ch}V_d$ term with a negative coefficient, $-1/2$) remain attractive.

We can see in Fig. 7 that d_{xy} pairing does satisfy the above condition. For n closer to half-filling, by contrast, $\chi_{\rm sp}$ becomes dominant with $\boldsymbol{Q}_{\rm sp}\simeq(\pm\pi,\pm\pi)$ (an arrow in Fig. 4), so $d_{x^2-y^2}$ is favored as usual. So the picture obtained here is that the pairing symmetry can change, even within the spin-singlet channel, when common peaks between $\chi_{\rm sp}$ and $\chi_{\rm ch}$ change into *segregated* peaks (as n and/or V are changed). This is contrasted with the above case of triplet f, for which a transition between singlet and triplet ($d \leftrightarrow f$)) can occur with the common peaks between $\chi_{\rm sp}$ and $\chi_{\rm ch}$ throughout. For $n=0.5$, Merino *et al.*[14] and Kobayashi *et al.*[26] have recently derived similar results using the slave-boson technique and RPA, respectively, which are consistent with the present result.

C. Quasi-one-dimensional lattice

Let us finally discuss the anisotropic (quasi-1D) case. While the original proposal made in Ref. 7 for the triplet "f-wave" pairing in $(\mathrm{TMTSF})_2X$ (where the symmetry refers to the warped Fermi surface; see Fig. 8 below) is for a quarter-filled case, the competition between the charge and spin fluctuations should become more stringent near half-filling, where U introduces $2k_F$ spin fluctuations while V enhances $2k_F$ charge fluctuations. Thus the triplet pairing should dominate over singlet d beyond some value of V. Note that the triplet gap function has to be f rather than p, since, with $\Gamma_t(\boldsymbol{Q})$ negative (attractive), the gap has to have the same sign across the nesting vector $\boldsymbol{Q}$. When V becomes

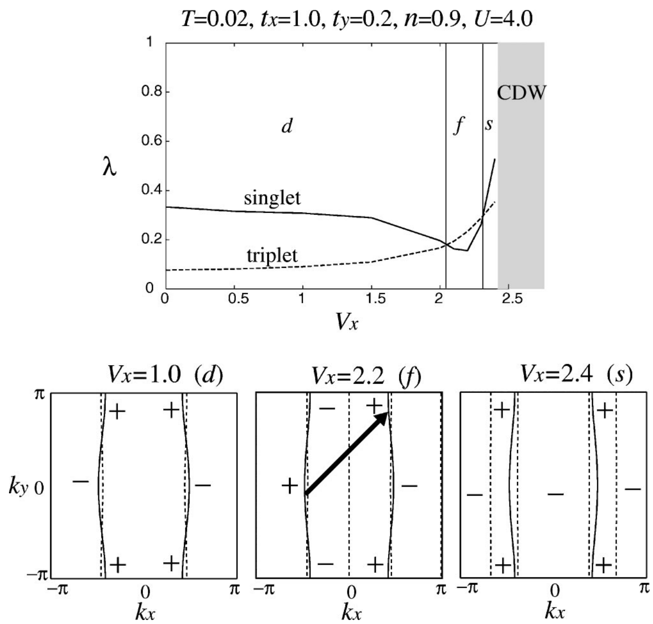

FIG. 8. Result for a quasi-1D system with $t_x=1.0$, $t_y=0.2$ for $n=0.9$. (Top) A plot similar to Fig. 3. (Bottom) The dominant gap function (with nodal lines represented by dotted lines) for $V_x=1.0$ (d wave), $V_x=2.2$ (f), and $V_x=2.4$ (s), along with the Fermi surface (solid curves) on which an arrow represents the nesting vector.

even larger, $\Gamma_s(\boldsymbol{Q})$ turns negative (attractive), so that the singlet s with no nodes on the Fermi surface will take over.

In order to show that this prediction is indeed realized, we have performed a FLEX calculation on a quasi-1D lattice with $t_x=1.0$, $t_y=0.2$. To represent the quasi-1D system we have here assumed that the off-site repulsion only acts between nearest neighbors along x (the conductive direction), although this assumption does not qualitatively alter the result. Figure 8 plots λ as a function of V_x for $n=0.9$, along with the forms of d, f, and s gap functions. We can see that the dominant pairing changes as $d\to f\to s$ with V, in an exact agreement with Ref. 7.

IV. CONCLUSION

We have obtained the phase diagram for the 2D extended Hubbard model with the FLEX approximation. We have found that f-wave pairing is favored for intermediate n and large V. In the more dilute regime ($n\sim 0.5$) the peak position of $\chi_{\rm ch}$ and $\chi_{\rm sp}$ is separated, and $d_{x^2-y^2}$ pairing gives way to d_{xy}. These are the consequences of how each pairing symmetry can exploit charge and spin fluctuations, and we end up with a picture that the pairing symmetry can change, even within the spin-singlet channel ($d_{x^2-y^2}\leftrightarrow d_{xy}$), when common peaks between $\chi_{\rm sp}$ and $\chi_{\rm ch}$ change into segregated peaks (as n and/or V are changed), which is contrasted with the case of common peaks throughout with a transition triplet $f\leftrightarrow d$. In the anisotropic (quasi-one-dimensional) case, the dominant pairing gap function changes as $d\to f\to s$ with V, which agrees with the phenomenological theory.[7]

In a broad context it should be interesting to examine how V (hence the charge fluctuation mediated interaction) can dominate the pairing symmetry in unconventional superconductors, although the nature of the pairing may be sensitively affected by the underlying band structure of each material.

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[24] A kink in λ for singlet pairing at $V \simeq 1.2$ corresponds to a change in the functional form in the singlet pairing channel.

[25] The true ground state may be $f + if$, since Fig. 4 rotated by 90° exists as a degenerate state.

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