The density-matrix renormalization group method is used to study the ground state of the two-chain zigzag-bond Hubbard model at quarter filling. We show that, with a proper choice of the signs of hopping integrals, the ring exchange mechanism yields ferromagnetic spin correlations between interchain neighboring sites, and produces the attractive interaction between electrons as well as the long-range pair correlations in the spin-triplet channel, thereby leading the system to triplet superconductivity. We argue that this novel mechanism may have possible relevance to observed superconductivity in Bechgaard salts.

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More than two decades have passed since the discovery of superconductivity in so-called Bechgaard salts such as \((\text{TMTSF})_2\text{X}\) with \(\text{X}=\text{PF}_6\), \(\text{ClO}_4\), etc. [1, 2]. The mechanism of superconductivity of this strongly correlated quasi-one-dimensional (1D) electron system [3] is, however, still an open issue. Recently, experimental evidences have been accumulating that the Cooper pairs of this system are in the spin-triplet state: they are from the measurements of the temperature dependence of Knight shift and spin-lattice relaxation ratio [4, 5, 6] as well as that of the upper critical field [7, 8, 9].

A variety of theoretical approaches have so far been made on this unconventional superconductivity [10, 11, 12, 13, 14, 15], but none of them is based upon real-space pairing mechanism possibly relevant due to their strong electron correlations. In this paper, we want to propose a novel mechanism that may have possible relevance to triplet superconductivity of this system. We adopt a numerical approach in order to take fully into account the strong electron correlations.

First, let us point out that the hopping integrals of \((\text{TMTSF})_2\text{X}\) have the unique structure as shown in Fig. 1(a) [16]: they show an alternating sign change along the zigzag bonds connecting two chains, while the sign along the 1D chain is always positive in the electron notation (i.e., for the bands of 3/4 filling of electrons). We hereafter use the hole notation for convenience; i.e., the system is in the quarter filling of holes. The signs of hopping integrals are then always negative along the 1D chain and are changing signs along the zigzag bonds.

Let us then notice that, in Hubbard models defined on such triangular-lattice related structures with proper signs of hopping integrals, the ring exchange mechanism of two spins on a triangle makes the system a ferromagnet with full spin polarization if sufficiently large Hubbard interaction \(U\) acts [17, 18, 19, 20]. The structure of hopping integrals of \((\text{TMTSF})_2\text{X}\) [16], in fact, satisfies the ferromagnetic sign rule \(t_1t_2t_3 > 0\) (in the hole notation) for three hopping integrals of all the triangles.

Our mechanism for triplet superconductivity relies on this ferromagnetic coupling; spin-triplet coupling for ferromagnetism is ‘relaxed’ to short-range spin-triplet correlations in realistic strength of \(U\), thereby leading the system to a metallic state with ferromagnetic spin correlations. We then ask what this state actually is.

We calculate the ground state of the relevant two-chain Hubbard model by the density-matrix renormalization group (DMRG) method [21] and show that indeed the system is metallic with ferromagnetic spin correlations. Furthermore, we show that the attractive interaction acts between holes, being caused by the gain in kinetic energy due to ring exchange of holes. We also show that the superconducting pair correlations in the spin-triplet channel extend long-ranged in power-law length dependence,
while the pair correlations in the singlet channel as well as the spin-density-wave (SDW) correlations decay exponentially, indicating that the system is in the state of spin-triplet superconductivity. We thus propose that the spin-triplet superconductivity is realized in the two-chain Hubbard model by the ring-exchange mechanism. We argue that the proposed mechanism may have possible relevance to triplet superconductivity in (TMTSF)$_2$X.

The minimum model to include the effects of interchain coupling may be the two-chain Hubbard model (see Fig. 1(b)) defined by the Hamiltonian

$$\mathcal{H} = \sum_{<ij>} t_{ij} (c_i^\dagger c_j + \text{H.c.}) + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

where $c_i^\dagger$ ($c_i$) is the creation (annihilation) operator of a hole with spin $\sigma$ (= $\uparrow, \downarrow$) at site $i$ ($j$), $n_{i\sigma} = c_i^\dagger c_{i\sigma}$ is the number operator, and $(ij)$ denotes the nearest-neighbor pair. We restrict ourselves to the case at quarter filling; i.e., $n = \sum_{\sigma} \langle n_{i\sigma} \rangle = 0.5$ where $\langle \cdots \rangle$ denotes the ground-state expectation value. $t_{ij}$ is the hopping integral between sites $i$ and $j$: we include $t_1$ along the zigzag chain and $t_2$ along the 1D chains. $U$ is the on-site Hubbard repulsion. We also examine the intersite repulsive term $\sum_{<ij>} V_{ij} n_{i\uparrow} n_{j\downarrow}$ when necessary: we again include $V_1$ along the zigzag chain and $V_2$ along the 1D chains. We hereafter assume $t_2 = -1$ as the unit of energy $|t_2| = 1$; observed small dimerization of hopping integrals along the 1D chains is neglected because the system remains metallic when the interchain hopping integrals are of the zigzag type [22]. We use values $t_1 = \pm 0.25$ and $\pm 0.5$ with the sign alternation. The signs of $t_1$ and $t_2$ can instead be taken all positive because the models where the product of the three hopping integrals of the triangles is positive are equivalent under canonical transformation. We use a value $U = 10$ (in some cases 20) for representing strong electron correlations in real systems [23]. The DMRG method is used for calculating the ground-state energy and correlation functions for clusters of length $L$ (containing $2L$ sites) with open boundary condition. We use clusters of up to $L = 128$ with keeping up to $m \simeq 4500$ density-matrix eigenstates; the discarded weights are typically of the order $10^{-7}$ to $10^{-6}$ to obtain the ground-state energy in the accuracy of $\sim 0.001 |t_2|$. Lanczos exact-diagonalization method on small clusters is also used.

Basic features of the ground state of our model are the following. First, the presence of ferromagnetic spin correlation is evident in Fig. 2, where we show the phase diagram on the $(V_1, V_2)$ plane of an $L=8$ cluster with periodic boundary condition obtained by an exact-diagonalization method. We find that when $U$ is large enough the system is spin polarized with the total spin $S > 0$ (see Fig. 2(a)). When $U$ is not large the total spin of the ground state becomes $S = 0$ but the interchain nearest-neighbor spin correlation is still ferromagnetic (see Fig. 2(b)); these results are confirmed to persist in larger size systems. The mechanism is apparent already in the two-hole ground state of a three-site Hubbard ring (which is spin triplet in a wide parameter region when $t_1 t_2 t_3 > 0$) and persists in higher dimensions as well [15]. We also calculate the charge gap defined as $\Delta_s = \lim_{L \to \infty} \Delta_s(L)$ with $\Delta_s(L) = E_L(N + 2) + E_L(N - 2) - 2E_L(N)$ where $E_L(N)$ is the ground-state energy of a chain of length $L$ with $N$ electrons with equal number of $\uparrow$ and $\downarrow$ spins. We use the DMRG method for clusters of up to $L = 64$ and make a finite-size scaling analysis. The obtained results (not shown) are similar to the results obtained in Refs. [24, 25, 26]; i.e., the charge gap opens due to charge ordering when either $V_1$ or $V_2$ is large, but metallic state appears in a wide parameter region around $V_1 = 2V_2$ (see Fig. 4 below for the case $V_1 = V_2 = 0$). Note that the region where the ferromagnetic correlation is strong agrees well with the metallic region where the ring exchange of holes works well.

To see the behavior of the spin degrees of freedom further, we calculate the equal-time spin correlation function $S(i,j) = \langle S_i^z S_j^z \rangle - \langle S_i^z \rangle \langle S_j^z \rangle$ where $S_i^z$ is the $z$-component of the spin operator of a hole at site $i$. We also calculate the spin gap defined as $\Delta_s = \lim_{L \to \infty} \Delta_s(L)$ with $\Delta_s(L) = E_L(N \uparrow + 1, N \downarrow - 1) - E_L(N \uparrow, N \downarrow)$ for $N \uparrow = N \downarrow = N/2$. The results are shown in Fig. 3, where and hereafter we assume $V_1 = V_2 = 0$ unless otherwise stated. We first find that the values of $\Delta_s(L)$ are rather small and extrapolated to $\Delta_s(L) \to 0$ within our numerical accuracy. Thus, the spin gap $\Delta_s$ vanishes (or becomes quite small if it exists) in the thermodynamic limit. We also find that the spin correlation decays with nearly exponential length dependence, which is associated with the oscillations of a period of 4 times lattice constant (consistent with the period of observed $2k_F$-SDW state in (TMTSF)$_2$X [16]). We should note that the apparent contradiction between the vanishing spin gap and expo-
The note that the values of $\kappa$ and gradients of the two curves (if the factor 4 is taken into account) for small $1/L$ regions agree well. Also noted is that the gradient is positive, or $\kappa > 0$, indicating that the system is thermodynamically stable against phase separation. We then find that the extrapolated value $\Delta_b$ is negative; i.e., the attractive interaction acts between holes in the thermodynamic limit. The energy gain responsible for the negative value of $\Delta_b$ may come from the motion of two holes running around the triangle, avoiding the on-site repulsion $U$ and exchanging spins for triplet coupling, i.e., from the ring exchange of holes. The pairing mechanism is thus kinetic in origin.

We also calculate the pair correlation function defined as

$$D(l) = (\Delta^\uparrow(\ell + 1)\Delta^\downarrow(\ell))$$

with $\Delta^\uparrow(\ell) = c_{\ell \uparrow}c_{\ell + 1 \uparrow} - c_{\ell \downarrow}c_{\ell + 1 \downarrow}$ for singlet pairs and $\Delta^\downarrow(\ell) = c_{\ell \downarrow}c_{\ell + 1 \uparrow} + c_{\ell \uparrow}c_{\ell + 1 \downarrow}$ for triplet pairs where $i + r$ denotes the neighboring sites of $i$. The results at $L = 128$ are shown in Fig. 5. We find that $D(l)$ shows the power-law length dependence for the interchain triplet pairing but decays exponentially for the singlet pairing as well as for the triplet pairing on the single 1D chain. These are the case also at $L = 64$, indicating that the size of the clusters used is sufficiently large. Quantitatively, the pair correlation function (see Fig. 5(b)) at long distances $r_{ij}$ decays as $\sim 1/r_{ij}^2$, the decay of which is slower by comparison than the decay of the charge correlation function $\sim 1/r_{ij}^0$; the estimation is made by fitting the data at $1 < l < L$ in order to avoid the effects of the edges of the clusters. Then, combined with the attractive interactions of two holes shown above, our results indicate that the system should be in the state of spin-triplet superconductivity where the pairing of holes occurs between the interchain nearest-neighbor sites.

Finally, let us consider possible relevance of our results to Bechgaard salts. The model is a coupled sequence of the 1D chains. We have cut out two neighboring chains as a minimum model to seek for consequences of the interchain coupling. Since the ring-exchange mechanism works also for models of more than two chains [18] where...
the hopping integrals of all the triangles satisfy the ferromagnetic sign rule, we point out that the triplet pairing state obtained in the two-chain model may persist also in quasi-1D (or 2D) systems; future studies will be interesting. Also pointed out is that there is intriguing competition between the SDW and spin-triplet superconducting states in the experimental pressure-temperature phase diagram [24]. Because small intersite Coulombic repulsions exist in the materials [23], we examine their effects; we find that the inclusion of a realistic value $V_2 \simeq 1$ [25] does not change our results. However, if we include $V_1$ between the 1D chains as well, the triplet pair correlation becomes less long-ranged and thus the SDW correlation can be comparable with it. A true long-range order may be selected among these competing correlations when the two (or three) dimensionality is taken into account, although to predict which order is realized is beyond the scope of the present work. We hope that future quantitative analyses will help clarifying the issue.

In summary, we have calculated the binding energy of holes and pair correlation functions in the two-chain zigzag-bond Hubbard model and have shown that the model can be superconducting in spin-triplet channel when we make an appropriate choice of the signs of hopping integrals for the ferromagnetic ring-exchange mechanism to work. We have argued that the mechanism proposed may have possible relevance to the triplet superconductivity in (TMTSF)$_2$X.

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