Critical Dimerization Strength of the Quarter-Filled $t-J$ Model

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An exact-diagonalization technique on small clusters is used to study the ground state of the dimerized $t-J$ model at quarter filling. The equal-time charge and spin correlations, charge and spin gaps, and binding energy are calculated for the two-dimensional lattice with a spatial dimerization pattern corresponding to organic conductors $\kappa$-(BEDT-TTF)$_2$X. We show that competition between the effects of two types of dimerization, i.e., dimerization in the hopping integral and dimerization in the exchange interaction, leads to the insulator-superconductor transition in the ground state of the model. The phase diagram is thereby presented.

KEYWORDS: $t-J$ model, dimerization, spin gap, charge gap, insulator-superconductor transition, BEDT-TTF

It has been emphasized that quasi-two-dimensional (2D) organic conductors $\kappa$-(BEDT-TTF)$_2$X have some characteristics similar to cuprate superconductors; apart from 2D nature of electron conduction, strong electron correlations, and Mott-insulator to superconductor transition, recent $^{13}$C-NMR and specific-heat measurements have suggested that the superconducting phase is anisotropic with $d$-wave-like nodes in the gap function and the behavior of $(TcT)^{-1}$ is somewhat similar to the spin-gap phenomena observed in cuprate superconductors (see Fig. 1(a)). Also noticeable is the very short coherence length of $\xi_0 \approx 25 \sim 30 \AA$, which is comparable to that for cuprate superconductors.

Motivated by these intriguing experimental data on the materials and some theoretical calculations on the dimerized Hubbard models, we here study the effects of dimerization on the ground state of the quarter-filled $t-J$ model, another representative model for strong electron correlations, by focusing on its insulator-superconductor transition. We will show that due to competition between two types of dimerization the ground-state phase of the model is divided into a Mott-insulating phase and a singlet-pairing phase; i.e., there is a critical dimerization strength. Although applicability of the model to the organic compounds is not yet fully worked out, we believe that the results shown here will help one understand a possible mechanism of superconductivity in strongly correlated electron systems such as $\kappa$-(BEDT-TTF)$_2$X.

The dimerized $t-J$ model we study is defined by the Hamiltonian

$$H = -t_1 \sum_{(ij)\sigma} \langle c^\dagger_{i\sigma} c_{j\sigma} + \text{H.c.} \rangle - t_2 \sum_{(kl)\sigma} \langle c^\dagger_{k\sigma} c_{l\sigma} + \text{H.c.} \rangle + J_1 \sum_{(ij)} \langle S_i \cdot S_j - \frac{n_i n_j}{4} \rangle + J_2 \sum_{(kl)} \langle S_k \cdot S_l - \frac{n_k n_l}{4} \rangle$$

(0.1)

where $c^\dagger_{i\sigma} = c_{i\sigma} (1-n_{i-\sigma})$ is the constrained electron-creation operator at site $i$ and spin $\sigma \in \{\uparrow, \downarrow\}$, $S_i$ is the spin-$1/2$ operator, and $n_i$ is the electron-number operator; we refer to the fermionic particle as 'electron', which corresponds to the hole in the real organic compounds. $(ij)$ stands for nearest-neighbor bonds with parameters $t_1$ and $J_1$ and $(kl)$ for those with parameters $t_2 \geq t_1$ and $J_2 \geq J_1$. We assume the lattice of a dimerization pattern simulating the 2D conducting plane of $\kappa$-(BEDT-TTF)$_2$X compounds (see Fig. 1(a)). Note that the model tends to the usual square-lattice $t-J$ model when there is no dimerization ($t_2 = t_1$ and $J_2 = J_1$), of which much study has been made; whereas in the limit of strong dimerization, the model represents an assembly of isolated dimers. We retain the relations between parameters $t$ and $J$ obtained from perturbation, i.e., $J_1 = 4t_1^2/U$ and $J_2 = 4t_2^2/U$, in order to reduce the number of parameters, where $U$ is the corresponding on-site Hubbard interaction. We thereby keep a relation $J_1/J_2 = (t_1/t_2)^2$.

We thus have three independent parameters, and if we take $t_1$ as a unit of energy, then we have two, for which we will take parameters representing $t$-dimerization and $J$-dimersization (of which a specific definition is given below). Here we restrict ourselves to the case of quarter filling to simulate the situation of the organic compounds.

We introduce two types of dimerization: one is the dimerization of hopping integral, which we call $t$-dimerization, and the other is the dimerization of exchange interaction, which we call $J$-dimerization. We define a parameter

$$\tilde{t}_d = \frac{t_2 - t_1}{t_1}$$

(0.2)

for the strength of $t$-dimerization and a parameter

$$\tilde{J}_d = \frac{J_2 - J_1}{t_1}$$

(0.3)

for the strength of $J$-dimerization. We also take $J_2/t_2$ as a measure of the strength of $J$-dimerization because if we keep $\tilde{t}_d$ constant ($>0$) then $J_2/t_2 \propto \tilde{J}_d$. We note that the $t$-dimerization has the effect leading to a repulsive interaction among electrons that acts when different spins ($\uparrow$ and $\downarrow$) come in a single dimer and that the...
$J$-dimerization has the effect promoting the spin-singlet formation between spins coming in a single dimer. Thus, the competition between the effects of these two dimerization may result in the following situation as illustrated in Fig. 1(b); when $t$-dimerization is dominant, the system can be a Mott insulator (because we have an effective half-filled band with the ‘on-site’ repulsion $U_{\text{dimer}}$), while when $J$-dimerization is dominant, the system can be a singlet-pairing state, and when $J_2/t_2$ is very large, the system will be phase separated.

![Fig. 1](image)

Fig. 1. (a) A 2D lattice structure of $\kappa$-(BEDT-TTF)$_2$X. The nearest-neighbor bonds have either $t_1$ and $J_1$ (thin solid line) or $t_2$ and $J_2$ (bold line). The unit cells, each of which contains four sites, are indicated by dashed lines. (b) Schematic 1D representation of the electronic states: from top to bottom, Mott insulator, singlet-pairing state, and phase separation are illustrated.

In the following, we will present numerical evidences that the competition indeed leads to the insulator-superconductor transition in the ground-state phase diagram of the dimerized $t-J$ model. We employ a numerical exact-diagonalization technique on small clusters. We use clusters of the size $1 \times 4$ and $2 \times 2$ unit cells (16 sites) with periodic boundary conditions (see Fig. 1 (a)). We find that nearly the same results are obtained for these two clusters, so that hereafter we will show the results only for the cluster of $2 \times 2$ unit cells.

Let us first discuss the equal-time charge and spin correlations. In Fig. 2, we show the calculated results for the charge correlation $\langle n_i n_j \rangle$ and spin correlation $\langle S_i^z S_j^z \rangle$, where $\langle \cdots \rangle$ denote the ground-state expectation value. For comparison, we also show the results for the $t-J$ cluster without dimerization ($\tilde{t}_d = \tilde{J}_d = 0$) in the topmost panels of Fig. 2. It is first of all evident in the figures that the effect of dimerizations indeed bring strong impact on the intra-dimer charge and spin correlations. We find that, in the small $J_2/t_2$ region where $J$-dimerization is weak, the intra-dimer charge correlation becomes closer to zero, indicating a tendency that only one electron presents in a dimer. This suggests that the effective repulsive interaction presents in a dimer. With increasing $t$-dimerization ($\tilde{t}_d$) this tendency appears to be much enhanced as clearly seen in Fig. 2. On the other hand, in the large $J_2/t_2$ region where the $J$-dimerization is large, the intra-dimer charge correlation is approaching the value $1/4$ and the intra-dimer spin correlation is decreasing to $-1/16$ as clearly seen in Fig. 2. This result indicates that two electrons with opposite spin come in a single dimer when $J$-dimerization is large. Then, in the region of intermediate strength of $J_2/t_2$, there appears a state where the charge correlations among different sites have nearly the same value $\sim 0.5/4$ and thus there are rather small spatial correlations. We note however that even in this region the nearest-neighbor spin correlations are still significantly antiferromagnetic.

Next let us examine the single-particle gap and charge gap. For the 16-site cluster at quarter filling the single-particle gap may be defined by

$$\Delta_{c1} = \frac{1}{2} \left[ (E_{\text{GS}}(5,4) - E_{\text{GS}}(4,4)) - (E_{\text{GS}}(4,4) - E_{\text{GS}}(3,4)) \right]$$

where $E_{\text{GS}}(N_{\uparrow}, N_{\downarrow})$ is the ground-state energy for the cluster of $N_{\uparrow}$ up-spin and $N_{\downarrow}$ down-spin electrons. The calculated results are shown in Fig. 3 (a). When there is no dimerization, one might expect that the system is metallic, i.e., $\Delta_{c1} = 0$ in the entire region of $J_2/t_2$ (except in the region of phase separation). We see however that the obtained value for $\Delta_{c1}$ is finite and it increases with increasing of $J_2/t_2$. The reason is, apart from an obvious finite-size effect of small clusters, that $\Delta_{c1}$ reflects the effect of electron pairing, even in the charge-gapless region (as discussed below). When there is dimerization, $\Delta_{c1}$ is finite even at $J_2/t_2 = 0$ and rapidly increases with increasing the strength of $t$-dimerization. We see that at constant $\tilde{t}_d$ and with increasing $J$-dimerization $J_2/t_2$,
the singlet-pairing region. When there is dimerization, 
phase separation), of which effect $\Delta$ may however be understood if we assume that the attrac-
tive interaction between pairs. We may also introduce. The calculated results are shown 
as $J_{2}/t_{2}$, because the value agrees with the value of $(J_{2}/t_{2})_{c}$ defined by using $\Delta_{s1}$. Above $(J_{2}/t_{2})_{c}$, the gap decreases as $J_{2}/t_{2}$ increases, which may again be due to the effect of attractive interaction between pairs.

Let us now consider the spin gap, which is defined by
$$\Delta_{s} = E_{GS}(5, 3) - E_{GS}(4, 4).$$

The calculated results are shown in Fig. 3 (c). We find that the spin gap opens at $J_{2}/t_{2} \approx (J_{2}/t_{2})_{c}$ and increases rapidly with increasing $J_{2}/t_{2}$ (if we assume that a small $\Delta_{s} > 0$ at $J_{2}/t_{2} < (J_{2}/t_{2})_{c}$ is a finite-size effect). We also find that with increasing $\tilde{t}_{d}$ the size of the gap decreases and the value $(J_{2}/t_{2})_{c}$ increases. Here we should emphasize that the $(J_{2}/t_{2})_{c}$ values estimated from the behaviors of $\Delta_{s1}$, $\Delta_{s2}$, and $\Delta_{s}$ are all consistent and have the same $\tilde{t}_{d}$ dependence. Thus it seems quite reasonable to assume that the spin gap $\Delta_{s}$ is finite in the entire charge-gapless region, where the electrons form a bound state as shown below.

The binding energy may be estimated by
$$\Delta_{B} = |E_{GS}(5, 5) - E_{GS}(4, 4)| - 2|E_{GS}(5, 4) - E_{GS}(4, 4)|$$

where $\Delta_{B}$ is negative if the state of two electrons minimize its energy by forming a bound state. A possibility of superconductivity may then be indicated. The calculated result for $\Delta_{B}$ is shown in Fig. 3 (d). We find that $|\Delta_{B}|$ is small for $J_{2}/t_{2} < (J_{2}/t_{2})_{c}$, but starts to increase at around $(J_{2}/t_{2})_{c}$, which is consistent with the expectation that the bound state of electrons is formed in the region of charge-gapless and spin-gapful phase. We also note that, with increasing $t$-dimerization, the binding energy is suppressed and the value of $(J_{2}/t_{2})_{c}$ increases accordingly. We should however be careful because when the value of $J_{2}/t_{2}$ is too large, the system will be phase separated. We estimate the parameter region of phase separation by examining if the value of the compressibility becomes negative. It is known that the phase separation actually occurs at around $J_{2}/t_{2} \approx 2.5 - 3$ when there is no dimerization. We thus examine its dependence on the $t$- and $J$-dimerizations; the obtained results are summarized in the phase diagram shown below.

Based on the calculated results given above, we obtain the phase diagram of the model on the parameter space of the two dimerizations. The result is shown in Fig. 4 where the horizontal axis is the $t$-dimerization $\tilde{t}_{d}$ and vertical axis is $J_{2}/t_{2}$, the measure of $J$-dimerization. The value of $\tilde{J}_{d}$ is also plotted. We see that when the $t$-dimerization is predominant over the $J$-dimerization, the system is a Mott insulator (the effective half-filled band with $U_{\text{dimer}}$), whereas when the $J$-dimerization is predominant over the $t$-dimerization, the system is in a singlet-pairing (or superconducting) state.

The boundary between these two phases approaches $J_{2}/t_{2} = 2$ in the limit of strong $t$-dimerization. This phase diagram

Fig. 3. (a) Single-particle gap $\Delta_{s1}/t_{2}$, (b) charge gap $\Delta_{s2}/t_{2}$, (c) spin gap $\Delta_{s}/t_{2}$, and (d) binding energy $\Delta_{B}/t_{2}$ as a function of $J_{2}/t_{2}$.

the charge gap $\Delta_{s2}$ opens for $\tilde{t}_{d} > 0$ and increases rapidly with increasing $\tilde{t}_{d}$. With increasing $J_{2}/t_{2}$, the gap decreases first and at some $J_{2}/t_{2}$ value the rate of the decrease changes, and at the same time $\Delta_{s2}$ changes sign where the gap closes. We may write this $J_{2}/t_{2}$ value as $(J_{2}/t_{2})_{c}$ because the value agrees with the value of $(J_{2}/t_{2})_{c}$ defined by using $\Delta_{s1}$. Above $(J_{2}/t_{2})_{c}$, the gap decreases as $J_{2}/t_{2}$ increases, which may again be due to the effect of attractive interaction between pairs.

The charge gap $\Delta_{s2}$ is suppressed and the value of $(J_{2}/t_{2})_{c}$ becomes negative. Here we should emphasize that the $(J_{2}/t_{2})_{c}$ values estimated from the behaviors of $\Delta_{s1}$, $\Delta_{s2}$, and $\Delta_{s}$ are all consistent and have the same $\tilde{t}_{d}$ dependence. Thus it seems quite reasonable to assume that the spin gap $\Delta_{s}$ is finite in the entire charge-gapless region, where the electrons form a bound state as shown below.

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is thus quite consistent with the expectation discussed in the beginning of the paper and illustrated in Fig. 1 (b).

It should be noted here that the phase diagram is obtained without finite-size scaling analysis of small-cluster data, as it is not feasible in any 2D-model calculations. We point out however that almost the same results are obtained for the 2×2 and 1×4 unit-cell clusters, that indications for the existence of the critical dimerization strength are robust, and that the proposed physical picture is quite obvious. We also note that, in the 1D model where a finite-size scaling can be done, we have confirmed the same mechanism for the insulator-metal transition actually works. We therefore believe that the numerical results presented in this paper reflects the reality of the model at least qualitatively.

Finally, let us examine the recent experimental data and some other calculations by taking κ-(BEDT-TTF)$_2$X as an example and see if we can find any relevance to our model system. In κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, the hopping integrals $t_1$ and $t_2$ for holes are reported to be $t_1\approx 0.1$ eV and $t_2\approx 0.25$ eV from the extended Hückel band calculation, which leads to the strength of t-dimerization $\tilde{t}_d\approx 1.5$. The Coulomb interaction $U$ on a BEDT-TTF molecule is reported to be ~1.0 eV, which is deduced from the analysis of the pressure dependence of the Knight shift based on the random-phase approximation. Thus the Coulomb interaction may be 4–10 times larger than the hopping integrals. Then if we assume that the exchange interactions can be estimated by the perturbation expression $J=4t^2/U$ we have $J_1/t_1\approx 0.4$ and $J_2/t_2\approx 1.0$. It follows then that the obtained point ($\tilde{t}_d$, $J_2/t_2$) falls on the Mott-insulating region of the phase diagram of Fig. 4. Now it is interesting to note that the effect of pressure is to decrease $t_2/t_1$ (and simultaneously increase $J_2/J_1$); an optimistic expectation would thus be that the system which is located in the Mott-insulating phase at ambient pressure shifts to the upper-right direction in Fig. 4 and can be in the singlet-pairing (or superconducting) phase with increasing pressure, as it actually is in experiment. It is also interesting to note that the critical dimerization strength ($J_2/t_2\approx 1.7$ obtained at $\tilde{t}_d=1.5$) is roughly consistent with the Hartree-Fock estimate of the critical repulsion ($U_{\text{dimer}}/t_2\approx 1.0$ in the dimerized Hubbard model because, if we use the expression $U_{\text{dimer}}=2t-J$ of an isolated $t-J$ dimer, we have $(J_2/t_2)\approx 1.0$. Further studies will however be required to clarify whether there is any continuity between the present dimerized $t-J$ model and more realistic dimerized Hubbard models, and also whether such large $J/t$ values are realized in the organic compounds κ-(BEDT-TTF)$_2$X.

In summary, we have examined the 2D dimerized $t-J$ model at quarter filling by using an exact-diagonalization technique on small clusters and have shown that there is a critical dimerization strength which divides the ground-state phase of the model into the Mott-insulating phase and singlet-pairing phase; either of the two phases is realized depending on the strength of $t$- and $J$-dimerizations. We have thereby obtained the ground-state phase diagram of the model in the parameter space of the two types of dimerization.

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