

## Quarter-filled extended Hubbard model with alternating transfer integral: Two-dimensional Ising transition in the ground state

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We study the one-dimensional quarter-filled extended Hubbard model with an alternating transfer integral. In the strong-dimerization limit the charge part is described by the quantum Ising model which shows the two-dimensional Ising criticality at the self-dual point, and it is naturally connected to the double-frequency sine-Gordon theory in the weak dimerization. Treating low-lying excitations in finite-size systems, we numerically determine a phase boundary between two types of  $4k_F$  density-wave states and clarify the ground-state phase diagram. Further, we refer to its relevances to the charge-ordered phase observed in the charge-transfer organic salts.

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The organic materials described by the chemical formula  $(\text{TMTSF})_2X$  ( $X=\text{PF}_6, \text{ClO}_4, \text{etc.}$ ) and  $(\text{TMTTF})_2X$  ( $X=\text{PF}_6, \text{AsF}_6, \text{etc.}$ ) form a class of the quasi one-dimensional (1D) conductors; a large number of investigations on these materials have been accumulated in the literature.<sup>1</sup> While the various types of electronic phases, e.g., spin/charge-density-wave (SDW/CDW), the spin-Peierls and the superconducting states, have been observed in the low-temperature region, newly discovered charge-ordered (CO) phase in  $(\text{TMTTF})_2X$  exhibiting an anomaly in the low-frequency dielectric constant<sup>2</sup> and the charge disproportionation in the NMR studies<sup>3</sup> has received intensive current interest. Although the stabilizations of these phases at finite temperature resort to interchain couplings, it is believed that the intrachain interaction effects play a leading role to describe them.

For the study of the CO phase, the 1D quarter-filled extended Hubbard model (EHM) with an alternating transfer integral has been employed:<sup>4-10</sup>  $H=H_1+H_2$  with

$$H_1 = \sum_{j,s} -t[1 - \delta(-1)^j](c_{j,s}^\dagger c_{j+1,s} + \text{H.c.}), \quad (1)$$

$$H_2 = \sum_j (Un_{j,\uparrow}n_{j,\downarrow} + Vn_j n_{j+1}), \quad (2)$$

where  $c_{j,s}$  annihilates an  $s$ -spin electron ( $s=\uparrow$  or  $\downarrow$ ) on the  $j$ th site and satisfies the periodic boundary condition  $c_{L+1,s}=c_{1,s}$  ( $j \in [1, L]$ ;  $L$  is an even number). The number operators are defined as  $n_{j,s}=c_{j,s}^\dagger c_{j,s}$  and  $n_j=n_{j,\uparrow}+n_{j,\downarrow}$ . The parameters  $U$  and  $V$  taking positive values stand for the onsite and nearest-neighbor Coulomb repulsion. The dimerization parameter  $\delta$  shows the alternation in the transfer integral of the molecular chains (we set  $t=1$  in the following). For the theoretical descriptions of the 1D electrons, the Tomonaga-Luttinger liquid (TLL) picture has been widely adopted.<sup>11</sup> Since TLL consists of the massless charge and spin parts both controlled by the Gaussian fixed point [the conformal field theory (CFT) with the central charge  $c=1$ ], it is important to understand its instabilities. In particular, the CO transition may be related to the crossover of the criticality embedded in the

renormalization group (RG) flow,<sup>8</sup> which is one of the typical instability of the  $c=1$  CFT.

In this paper, we present the numerical calculation results of the ground-state phase diagrams of  $H$ . Our method being deeply connected to the instability will be explained briefly. Further, in the strong-dimerization limit ( $\delta=1$ ), we show that the charge part can be described by the so-called quantum Ising chain, which is complementary to the bosonization argument and gives us an exact limiting condition of the phase boundary line. Since the region with sufficiently large Coulomb repulsions is relevant to the CO transition, an occurrence of the phase separation or the transition to the superconducting phases is outside of our research scope.

Let us start with the description of the low-energy physics in the weak-coupling region, where the bosonization method provides a reliable approach, i.e., linearizing the dispersion at Fermi points  $\pm k_F = \pm \pi n/2a$  (electron density  $n=N/L=\frac{1}{2}$ ) and applying the method, we can obtain an effective Hamiltonian. For the present case, according to the recent research results,<sup>1,7,12</sup> we can use the following expression:  $H \rightarrow \mathcal{H} = \mathcal{H}_\rho + \mathcal{H}_\sigma$  with

$$\mathcal{H}_\rho = \int dx \frac{v_\rho}{2\pi} \left[ K_\rho (\partial_x \theta_\rho)^2 + \frac{1}{K_\rho} (\partial_x \phi_\rho)^2 \right] + \int dx \frac{2}{(2\pi\alpha)^2} (-g_\rho \sin \sqrt{8}\phi_\rho + g_{1/4} \cos 2\sqrt{8}\phi_\rho), \quad (3)$$

where the operator  $\theta_\rho$  is the dual field of  $\phi_\rho$  satisfying the commutation relation  $[\phi_\rho(x), \partial_y \theta_\rho(y)/\pi] = i\delta(x-y)$  and parameters  $K_\rho$  and  $v_\rho$  are the Gaussian coupling and the velocity of the charge excitation, respectively.<sup>13</sup> A benefit to use the bosonized expression is now clear, i.e., since the spin-charge separation occurs in  $\mathcal{H}$  and  $\mathcal{H}_\sigma$  is the SU(2) critical Gaussian model in the present case,<sup>7,8</sup> we can concentrate on the charge part  $\mathcal{H}_\rho$  which takes a form of the so-called double-frequency sine-Gordon (DSG) model. In uniform case ( $\delta=0$ ), the  $8k_F$ -Umklapp scattering with  $g_{1/4} \propto U^2(U-4V)$  (Refs. 12 and 14-16) brings about the Berezinskii-

Kosterlitz–Thouless (BKT) transition, and then the charge part becomes massive for large values of the Coulomb interactions. For the BKT transition point, values in the strong coupling limit are known as  $V^*(U \rightarrow \infty) = 2$  and  $U^*(V \rightarrow \infty) = 4$ .<sup>17,18</sup> Further, the estimations for the intermediate region are available.<sup>12,16,19</sup> In the case of nonzero dimerization ( $\delta \neq 0$ ), the scaling dimension of the “half-filled Umklapp scattering” term with  $g_\rho \propto U\delta[1 - A(U - 2V)]$  ( $A$  is a constant)<sup>7,15</sup> on the Gaussian fixed point is small ( $x_{4B} = 2K_\rho$ ) enough to bring about the second-order phase transition for  $V \leq V^*(U)$ , which is accompanied by the divergent correlation length of the form  $\xi \propto \delta^{-1/(2-2K_\rho)}$ .<sup>1,20</sup> For  $V > V^*(U)$ , since the charge gap may survive in a weak-dimerization region, the transition point  $\delta_\rho(U, V)$  takes nonzero values depending on  $U$  and  $V$ , and, more importantly, the universality of the transition is changed. Recently, Tsuchiizu and Orignac,<sup>8</sup> on the basis of the DSG theory,<sup>21</sup> argued that the charge part on  $\delta_\rho(U, V)$  [ $V > V^*(U)$ ] is renormalized to the 2D-Ising fixed point with  $c = \frac{1}{2}$  (i.e., the fixed point with lower symmetry), which is in accord with Zamolodchikov’s  $c$ -theorem<sup>22</sup> (see also Refs. 23 and 24). Then, the critical line corresponds to the phase boundary and satisfies a condition  $\delta_\rho(U, V \rightarrow V^*(U)) \searrow 0$  in the weak-dimerization region. To characterize the phases, we shall use the CDW and the bond-order-wave (BOW) order parameters with the  $4k_F$  wave vector:<sup>12</sup>

$$\mathcal{O}_{4C} \propto \cos \sqrt{8}\phi_\rho, \quad \mathcal{O}_{4B} \propto \sin \sqrt{8}\phi_\rho. \quad (4)$$

Here, note that the expectation value of the  $4k_F$ -BOW order parameter is finite,  $\langle \mathcal{O}_{4B} \rangle \neq 0$  and  $\langle \mathcal{O}_{4C} \rangle = 0$  in the upper region ( $\delta \neq 0$ ). While this “mixed” state is basically the  $4k_F$ -CDW phase, we shall use the double quotation marks “ $4k_F$ -CDW” to express this situation.<sup>24</sup>

On the other hand, another condition of the boundary can be found in the strong-dimerization limit ( $\delta = 1$ ). To derive an effective Hamiltonian, it is convenient to work with the orbital operators defined by  $d_{m,\pm,s} \equiv (c_{2m-1,s} \pm c_{2m,s})/\sqrt{2}$ , where  $d_{m,l,s}$  annihilates an  $s$ -spin electron in the  $l$ -orbital ( $l = \pm$ ) on the  $m$ th unit cell ( $m \in [1, L/2]$ ). In this limit,  $H_1$  consists of a sum of the intracell electron hopping, which is diagonalized by using the operators as  $H_1 = \sum_{m,l,s} 2ld_{m,l,s}^\dagger d_{m,l,s}$ . For sufficiently large  $U$  and  $V$ , since the one-electron states  $|l, s\rangle_m = d_{m,l,s}^\dagger |0\rangle$  have a principal role to describe the  $m$ th unit cell in the quarter-filled ground state, and the Hamiltonian does not change the electron number in each cell, we shall introduce the pseudospin operators,

$$T_m \equiv \sum_{l,l',s} \frac{1}{2} d_{m,l,s}^\dagger [\boldsymbol{\tau}]_{l,l'} d_{m,l',s}, \quad (5)$$

acting on the orbital space as, for instance,  $T_m^\dagger |\pm, s\rangle_m = \pm \frac{1}{2} |\pm, s\rangle_m$  [ $\boldsymbol{\tau} = (\tau^1, \tau^2, \tau^3)$ ;  $\tau^i$  is the Pauli matrix]. Using these,  $H_1 = \sum_m -4T_m^3$ . For  $H_2$ , since the intracell Coulomb interactions are absent and the intercell Coulomb repulsion only remains in the restricted Hilbert space spanned by the direct product of one-particle states  $\{\otimes_m |l, s\rangle_m\}$ , a straightforward calculation brings about the expression

$H_2 = \sum_m (-VT_m^1 T_{m+1}^1 + \text{const})$ . Now, since the Hamiltonian acts only on the orbital space, its eigenstate takes a form of the direct product of vectors in the spin and the orbital spaces as  $|\Phi\rangle = |\text{spin}\rangle \otimes |\text{orbital}\rangle$ . Thus, assuming a certain spin configuration belonging to the  $2^{L/2}$ -dimensional space for spins and restricting ourselves to the orbital (or charge) part, we see that the Hamiltonian  $H$  with  $\delta = 1$  is reduced to the quantum Ising chain<sup>25</sup>

$$H_{\rho, \delta=1} = \sum_m (-\Gamma T_m^3 - J T_m^1 T_{m+1}^1) \quad (6)$$

( $\Gamma = 4$ ,  $J = V$ ). Note that this possibility was mentioned qualitatively in Ref. 10. Then, the ground state of Eq. (6) is known to show the 2D-Ising criticality at its self-dual point  $\Gamma = J/2$  ( $V = 8$ ), which separates ordered ( $\langle T_m^1 \rangle \neq 0$ ) and disordered ( $\langle T_m^1 \rangle = 0$ ) phases. The ordered state is realized via the breaking of the  $Z_2$  symmetry ( $\tau^1 \rightarrow -\tau^1$ ), and it is doubly degenerated, e.g.,

$$|\pm \tau^1\rangle = \prod_m \frac{1}{\sqrt{2}} (d_{m,+}^\dagger \pm d_{m,-}^\dagger) |0\rangle = \prod_m c_{2m-1}^\dagger (c_{2m}^\dagger) |0\rangle \quad (7)$$

(we dropped the spin index). This expresses the  $4k_F$ -CDW state with the perfect microscopic polarization,  $\langle \pm \tau^1 | T_m^1 | \pm \tau^1 \rangle = \langle \pm \tau^1 | \frac{1}{2} (n_{2m-1} - n_{2m}) | \pm \tau^1 \rangle = \pm \frac{1}{2}$ . On one hand, a disordered state is supported by the external field in  $\tau^3$ -direction, and an ideal one is given by

$$|+\tau^3\rangle = \prod_m d_{m,+}^\dagger |0\rangle = \prod_m \frac{1}{\sqrt{2}} (c_{2m-1}^\dagger + c_{2m}^\dagger) |0\rangle, \quad (8)$$

which expresses the  $4k_F$ -BOW state as expected. Here it is worthy of noticing that these states can be distinguished by the expectation value of the twist operator<sup>26</sup>

$$z_\rho \equiv \left\langle \exp \left( \frac{4\pi i}{L} \sum_j j n_j \right) \right\rangle. \quad (9)$$

This quantity takes values  $z_\rho = 1$  for Eq. (7) and  $z_\rho = -[\cos(2\pi/L)]^{L/2}$  for Eq. (8), so the sign of  $z_\rho$  characterizes these two density-wave states (see below). Consequently, in the strong-dimerization limit, the orbital degrees of freedom show the 2D-Ising type transition between the “ $4k_F$ -CDW” and the  $4k_F$ -BOW phases at  $V = 8$ , where  $U$  is irrelevant. Since this pseudospin representation is naturally connected to the bosonization picture in the weak couplings,<sup>8</sup> the phase boundary belongs to the 2D-Ising universality and satisfies the limiting condition  $\delta_\rho(U, V \rightarrow 8) \nearrow 1$ , which provides a solid guide to investigations in the strong-dimerization region.

Here, note that the qualitative estimation of the phase boundary might be possible in the weak- and strong-dimerization region.<sup>7</sup> To evaluate the entire phase diagram precisely, however, a numerical treatment of the 1D electron model is required. For this issue, recently the present authors have numerically treated the same instability observed in the quantum-spin chain and interacting electron systems.<sup>27</sup> Therefore, we shall employ the same approach to the present system (see also Ref. 28). Since there are two critical fixed points connected by the RG flow, a relationship between

lower-energy excitations on these fixed points—the ultraviolet-infrared (UV-IR) operator correspondence—has essential significance in the investigations.<sup>8,24</sup> To see this, let us rescale phase fields and the Gaussian coupling as  $2\phi_\rho \rightarrow \phi$ ,  $\theta_\rho/2 \rightarrow \theta$ , and  $4K_\rho \rightarrow K \approx 1$ , which makes it possible to directly adopt our previous research.<sup>27</sup> With respect to  $\phi$ , the nonlinear potential density is given as  $-g_\rho \sin \sqrt{2}\phi + g_{1/4} \cos \sqrt{8}\phi$ , and the order parameters as  $\mathcal{O}_{4C} \propto \cos \sqrt{2}\phi$  ( $x_{4C}=K/2$ ) and  $\mathcal{O}_{4B} \propto \sin \sqrt{2}\phi$  ( $x_{4B}=K/2$ ). Along the RG flow these operators on the Gaussian fixed point (UV) are transmuted to those on the 2D-Ising fixed point (IR) as

$$\mathcal{O}_{4C} \rightarrow \mu, \mathcal{O}_{4B} \rightarrow I + \epsilon, \quad (10)$$

where  $\mu$  is the disorder field ( $Z_2$  odd), and  $\epsilon$  is the energy density operator ( $Z_2$  even) with scaling dimensions  $x_\mu = \frac{1}{8}$  and  $x_\epsilon = 1$ , respectively. Since the dimerization  $\delta$  couples with  $\mathcal{O}_{4B}$  in the Hamiltonian (3), a deviation from the transition point  $\delta - \delta_\rho(U, V)$  plays a role of the “thermal scaling variable” and brings about  $\xi \propto [\delta - \delta_\rho(U, V)]^{-\nu}$  with  $1/\nu = 2 - x_\epsilon = 1$ . On one hand, the operator  $\mu$  corresponding to  $\mathcal{O}_{4C}$  provides a most divergent fluctuation.

Now, we shall explain our numerical procedure to determine the transition point. We shall focus our attention on the level  $\Delta E$  in finite-size systems which corresponds to the operator  $\mathcal{O}_{4C}$  (taking the ground-state energy as zero). According to the finite-size-scaling argument based on CFT,  $\Delta E \approx 2\pi x_{4C}/L$  on the UV fixed point;<sup>29</sup> we can numerically obtain the level by using discrete symmetries of the lattice Hamiltonian in the diagonalization calculations. Various excitations observed in TLL are characterized by a set of quantum numbers for symmetry operations. With respect to  $\mathcal{O}_{4C}$ , it can be found in the subspace of the total spin  $S_T^z = 0$  and the space inversion  $P = -1$  (the boundary condition is the same as that for the ground state).<sup>16</sup> Suppose that  $\Delta E(U, V, \delta, L)$  is a level corresponding to  $\mathcal{O}_{4C}$  in the  $L$ -site system. Then, we numerically solve the phenomenological renormalization-group (PRG) equation  $(L+2)\Delta E(U, V, \delta, L+2) = L\Delta E(U, V, \delta, L)$  with respect to  $\delta$  for given values of  $U$  and  $V$ , where the gap behaves as  $\Delta E \propto 1/L$  [i.e., an  $L$ -dependent transition point  $\delta_\rho(U, V, L+1)$  (see Fig. 1)].<sup>27</sup> After evaluating  $\delta_\rho(U, V, L+1)$ , we extrapolate them to the limit  $L \rightarrow \infty$  using the formula  $\delta_\rho(U, V, L) = \delta_\rho(U, V) + aL^{-3}$ ,<sup>30</sup> where  $\delta_\rho(U, V)$  and  $a$  are determined by the least-square-fitting condition.

From the data of  $L = 12-20$ , we obtain the phase boundary  $\delta_\rho(U, V)$  as shown in Fig. 1. We can check that, for all values of  $U$  used here, the phase boundary lines converge to the point  $(V, \delta) = (8, 1)$  with the 2D-Ising criticality. On the other hand, while the finite-size corrections to the boundary may be large in weak-dimerization region, the boundaries also show convergences to the BKT-transition points  $(V^*(U), 0)$ . Next we demonstrate the  $\delta$  dependence of  $z_\rho(L)$  in Fig. 2. With the increase of  $\delta$ ,  $z_\rho(L)$  decreases and becomes negative [we denote the zero point of  $z_\rho(L)$  as  $\delta'_\rho(U, V, L)$ ]; this corresponds to the change of the center of mass as demonstrated in the above.<sup>26</sup> However, unlike, for instance, the Gaussian transition,  $z_\rho(\infty)$  can take a finite value on the Ising transition point, so  $\delta'_\rho(U, V, L)$  may not give an estimation of the tran-

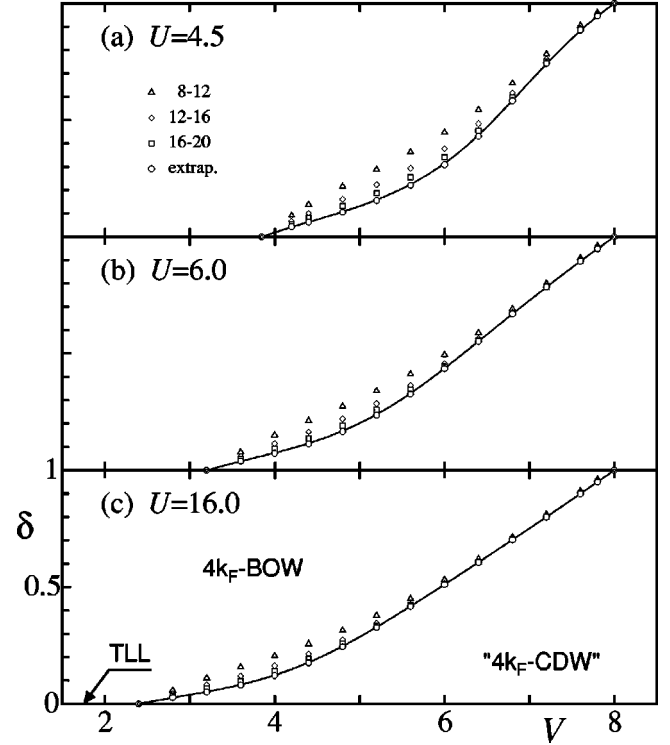


FIG. 1. The ground-state phase diagram of the quarter-filled EHM with an alternating transfer integral. The correspondence between marks and system sizes is given in the figure. The double circles show the limiting values, i.e.,  $(V^*(U), 0)$  at which criticality changes from the Gaussian to the 2D-Ising type, and  $(8, 1)$  the self-dual point.

sition point. In fact, the inset of Fig. 2 exhibits that  $\delta'_\rho(U, V, L)$  may be extrapolated to a value different from the PRG result. On the other hand, Fig. 2 also shows that there is a point  $\delta \approx 0.12$  at which  $z_\rho(L)$  is almost independent of  $L$ . This crossing point is expected to be a good estimator for the Ising transition point because this is quite close to the PRG result even for small  $L$ . However, this issue remains as a future problem.

Lastly, we shall refer to some implications of our study to the real materials. Besides the quantum-chemistry

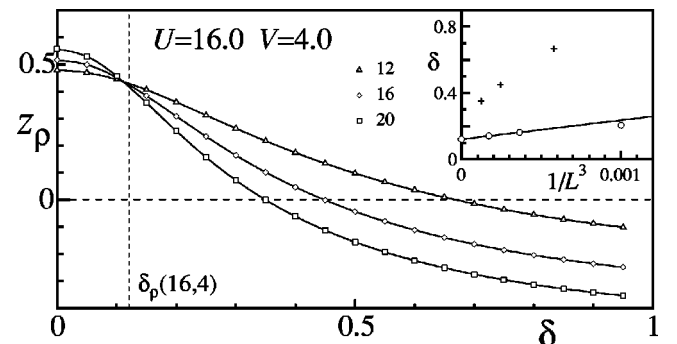


FIG. 2. The  $\delta$  dependence of  $z_\rho(L)$  at  $U=16$  and  $V=4$ . The vertical dotted line indicates the transition point (i.e., the PRG result)  $\delta_\rho(16, 4) \approx 0.12$ . Inset plots  $L$  dependences of  $\delta'_\rho(U, V, L)$  (crosses) and the PRG data  $\delta_\rho(U, V, L)$  (circles) with the fitting line.

calculations,<sup>31</sup> the numerical estimations of the model parameters have been performed based upon the experimental data.<sup>4,9</sup> For example, the realistic values of the dimerization parameter and the onsite Coulomb repulsion of  $(\text{TMTTF})_2\text{PF}_6$  have been estimated as  $t_2/t_1 \approx 0.7$ ,  $U/t_1 \approx 7.0$  ( $t_{1,2} = 1 \pm \delta$ ), but the value of  $V$  is still controversial (an uncertainty exists also in the value of  $t_2/t_1$ <sup>2,32</sup>). Our numerical estimation of the transition point using these values is  $V_c/t_1 \approx 4.0$ , while generally the mean-field-type calculations tend to predict somewhat smaller values due to an overestimation of  $V$  effects.<sup>5,10</sup> On the other hand, several values have been reported for this material, e.g.,  $V/t_1 \approx 2.8$  (1.4) in Ref. 4 (Ref. 9), which is much smaller than the critical value, and thus predicts a uniform charge distribution (this conclusion may not be changed even in smaller dimerization cases). However,  $(\text{TMTTF})_2\text{PF}_6$  has the CO phase in the region above the lower-temperature spin-Peierls phase, and further it was theoretically suggested that a huge anomaly in the dielectric constant may reflect a nature of systems in the critical region.<sup>8</sup> This discrepancy may be attributed to many

other interaction effects not included in the Hamiltonian. However, we think that since experimental findings seem to support the spin-charge separation with respect to the CO transition,<sup>2,3</sup> the 1D electron models are to provide a primary description of real materials.

To summarize, we investigated the ground-state phase diagram of the 1D quarter-filled extended Hubbard model with alternating transfer integral. Especially, the criticality on the phase boundary and the implication to the CO transition observed in the charge-transfer organic salts were mainly argued.

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