Correlation Effects in a One-Dimensional Quarter-Filled Electron System with Repulsive Interactions

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A one-dimensional electron system at quarter-filling has been examined by applying the renormalization group method to a bosonized model on-site (U) and nearest-neighbor (V) repulsive interactions. By evaluating both normal scattering and Umklapp scattering perturbatively, we obtain a phase diagram in which a metallic state with a 2k_F spin density wave (k_F is the Fermi wave number) moves into an insulating state with charge disproportionation of a 4k_F charge density wave with an increase in both U and V. The effect of the next-nearest-neighbor repulsion is also discussed.

KEYWORDS: extended Hubbard model, quarter-filling, commensurability energy, spin density wave, charge density wave.

A one-dimensional (1-D) electron system at quarter-filling is a basic model for understanding the electronic properties of quasi 1-D organic conductors. The 1-D electron system with only on-site Coulomb repulsive interaction, U (1-D Hubbard model), does not exhibit a metal-insulator (M-I) transition as a function of U, and is metallic at any filling, except half-filling. The most dominant state is given by 2k_F spin density wave (2k_F-SDW), where k_F is the Fermi wave number. The presence of the long-range repulsive interaction is expected to enrich the phase at quarter-filling. In fact, numerical diagonalization for the model with both U and the nearest-neighbor interaction, V, shows that the insulating phase appears for a large strength of both U and V and that the superconductivity becomes the most dominant fluctuation for a large V and small U. On the other hand, several properties have been elucidated within the mean-field theory. With increasing V, a transition occurs from a pure 2k_F-SDW state to a coexistent state of 2k_F-SDW and 4k_F charge density wave (4k_F-CDW). Such a coexistence is maintained for the charge ordering in (Dl-DCNQI)$_2$Ag observed by the $^{13}$C-NMR measurement. The transition has been examined by evaluating the commensurability energy corresponding to the 8k_F-Umklapp scattering. The next-nearest-neighbor repulsion results in a coexistence of 2k_F-SDW and purely electronic 2k_F-CDW and it has been proposed to be the origin of the coexistence observed by the X-ray experiment on (TMTSF)$_2$PF$_6$.

At quarter-filling, the 8k_F-Umklapp scattering is crucial to obtain the commensurability energy. Although the above mean-field results may show common features, we need to calculate the Umklapp scattering in the 1-D system by taking into account quantum fluctuation. The existence of commensurability energy of such high order has been pointed out. However, to the best of our knowledge, there are no studies for the phase diagram on the plane of U and V, which is calculated by using the analytical expression of the commensurability energy.

In the present paper, a 1-D system with repulsive interactions at quarter-filling is investigated using the bosonization method and the renormalization group (RG) theory. Based on the commensurability energy, a phase diagram is derived on the plane of U and V. The relevance of the present results to the observation in (Dl-DCNQI)$_2$Ag salt and the effect of the next-nearest-neighbor interaction are discussed.

We consider a 1-D extended Hubbard model given by the Hamiltonian, $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{int}}$,

$$\mathcal{H}_0 = -t \sum_{j,\sigma} \left( a_{j,\sigma}^\dagger a_{j+1,\sigma} + h.c. \right) - \mu \sum_{j,\sigma} n_{j,\sigma}$$

$$= \sum_{K,\sigma} (\epsilon_K - \mu) a_{K,\sigma}^\dagger a_{K,\sigma}, \quad (1)$$

$$\mathcal{H}_{\text{int}} = \frac{U}{2} \sum_{j,\sigma} n_{j,\sigma} n_{j+1,\sigma} + V \sum_{j,\sigma,\sigma'} n_{j,\sigma} n_{j+1,\sigma'}$$

$$= \frac{1}{N_L} \sum_{\sigma'} \sum_{K_1 \sim K_4} \left\{ \frac{U}{2} \delta_{\sigma,\sigma'} + V e^{-i(K_2-K_3)a} e^{iK_4 a} \right\}$$

$$\times \delta_{K_1+K_2-K_3-K_4} G a_{K_1,\sigma}^\dagger a_{K_2,\sigma}^\dagger a_{K_3,\sigma'} a_{K_4,\sigma}, \quad (2)$$

where $t$ and $\mu$ denote the transfer energy and chemical potential, respectively, and $\epsilon_K = -2t \cos K a$ with lattice constant $a$ and $-\pi/a < K \leq \pi/a$. The quantity $a_{j,\sigma}^\dagger = \frac{1}{\sqrt{N_L}} \sum_k e^{-ikja} a_{K,\sigma}^\dagger$ denotes the creation operator of the electron at the $j$-th site with spin $\sigma$, $n_{j,\sigma} = a_{j,\sigma}^\dagger a_{j,\sigma}$, $G = 0, \pm 2\pi/a$, and $N_L$ is the number of the lattice. For the later convenience, we divide the one-particle states as $d_{K,-,\sigma} = a_{K,\sigma}$ for $-\pi/a < K \leq -\pi/(2a)$, $c_{K,-,\sigma} = a_{K,\sigma}$ for $-\pi/(2a) < K \leq 0$, $c_{K,+,-,\sigma} = a_{K,\sigma}$ for $0 < K \leq \pi/(2a)$, and $d_{K,+,-} = a_{K,\sigma}$ for $\pi/(2a) < K \leq \pi/a$, $k$ being the deviation of the wave

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number from ±k_F for c_{k_±,σ}, and ±3k_F for d_{k_±,σ}. In terms of c_{k,p,σ} and d_{k,p,σ}, H_0 is written as

\[ H_0 = \sum_{p,k,σ} \left\{ (\epsilon_{p,k} - k \cdot c_{k,p,σ}) c_{k,p,σ}^\dagger c_{k,p,σ} + (\epsilon_{p,k} - k \cdot d_{k,p,σ}) d_{k,p,σ}^\dagger d_{k,p,σ} \right\}, \]

and H_int is rewritten as

\[ H_{\text{int}} = \sum_{i=1}^4 \mathcal{H}_{i,i}, \]

where i denotes the number of d_{k,p,σ} and/or d_{k,p,σ}^\dagger in the respective interactions.

When the one-particle states only near the Fermi wave number, ±k_F, are taken into account, the Hamiltonian is reduced to

\[ \mathcal{H} = \sum_{p,k,σ} p v_F k c_{k,p,σ} c_{k,p,σ}^\dagger + \frac{1}{L} \sum_{p k' q k'} \left\{ (g_{1,0}^0 \delta_{σ-σ'} + g_{1,0}^0 \delta_{σ+σ'}) c_{k+q,p,σ}^\dagger c_{k',q',σ'} c_{k,σ} c_{k,σ'} + (g_{2,0}^0 \delta_{σ-σ'} + g_{2,0}^0 \delta_{σ+σ'}) c_{k+q,p,σ}^\dagger c_{k',q',σ'} c_{k,σ'} c_{k,σ} + (g_{4,0}^0 \delta_{σ-σ'} + g_{4,0}^0 \delta_{σ+σ'}) c_{k+q,p,σ}^\dagger c_{k',q',σ'} c_{k,σ'} c_{k,σ'} \right\}, \]

(3)

where L = N_L a and the energy dispersion is linearized at ±k_F as \( \epsilon_{±k_F} = ±v_F k \) with \( v_F = \sqrt{2} t \). Coupling constants are given by \( g_{1,0}^0 = g_0^0 = U a/2 + V a, g_{2,0}^0 = g_0^0 = V a, g_{1,0}^\perp = U a/2, \) and \( g_{1}^0 = 0 \). We note that eq.(3) does not show the MI transition since there is no Umklapp scattering.

According to a theoretical suggestion by Schulz and calculation of the mean-field theory, Umklapp scattering in the case of quarter-filling appears through the one-particle states with ±3k_F, which are connected to those of the Fermi point by ±2k_F. We take into account the states near ±3k_F systematically by following the procedure in the above discussion, to derive the effective Hamiltonian, which consists of the one-particle states only near ±k_F. Representing c_{k,p,σ} and d_{k,p,σ} in terms of Grassmann algebra, the partition function of eqs.(1) and (2), Z, is given by

\[ Z = \int [D c_{k,p,σ}^\dagger D c_{k,p,σ}] e^{-S}, \]

where \( S = S_0[c_{k,p,σ} c_{k,p,σ}^\dagger] + S_0[d_{k,p,σ} d_{k,p,σ}^\dagger] + \sum_{i=0}^4 S_{\text{int},i}, \) and \( S_{\text{int},i} \) denotes the action corresponding to \( H_{\text{int},i} \).

After integrating c\_\_\_\_\_p,σ, the partition function, Z, is written as

\[ Z = Z_0 \int [D c_{k,p,σ}^\dagger] e^{-S_{\text{eff}}}, \]

where \( Z_0 = \int [D d_{k,p,σ}] e^{-S_0[d_{k,p,σ} d_{k,p,σ}^\dagger]} \) and \( S_{\text{eff}} = S_0[d_{k,p,σ} d_{k,p,σ}^\dagger] - \ln \left( \exp(-\sum_{i=0}^4 S_{\text{int},i}) \right) (\cdot \cdot \cdot) \_d \) is the average by \( S_0[d_{k,p,σ} d_{k,p,σ}^\dagger] \). By calculating \( S_{\text{eff}} \) perturbatively, the effective Hamiltonian is derived from the diagram for the vertex includes only the green functions of d_{k,p,σ}. From the perturbation up to the second order of U and/or V, the correction to the normal processes, \( g_{i,2}^\perp \), in eq.(3) is calculated as

\[ \Delta g_{1,2} = -4D_1 \left( \frac{U a}{2} \right) \left( \frac{U a}{2} - V a \right), \]

(4)

\[ \Delta g_{2,2} = -2D_1 (V a)^2, \]

(5)

\[ \Delta g_{4,2} = -2D_1 \left( \frac{U a}{2} \right)^2 - 2D_1 \left( \frac{U a}{2} - V a \right)^2, \]

(6)

\[ \Delta g_{4,4} = -2D_2 (V a)^2, \]

(7)

\[ \Delta g_{4,4} = -2D_2 \left( \frac{U a}{2} - V a \right)^2, \]

(8)

where

\[ D_1 = (8t a)^{-1} \int_0^{\pi/2} dy \sin y + 1/\sqrt{2} \left( 8t a \right)^{-1} - 1.25/\left( 8t a \right) \]

and

\[ D_2 = -D_2 (8t a)^{-1} \int_0^{\pi/2} dy \sin y + 1/\sqrt{2} \left( 8t a \right)^{-1} - 1.25/\left( 8t a \right) \]

The 8k_F-Umklapp scattering, which comes from the third order expansion, is obtained as

\[ H_{1/4} = \frac{(U a)^3}{8L^2} \sum_{p σ} \int dx (c_{p,σ} c_{p,σ})^3 c_{p,σ} c_{p,σ} c_{p,σ}, \]

(9)

where the first term corresponds to the conventional commensurability energy \( \tilde{\epsilon} \) and \( ψ_{p,σ} = 1/√L \sum_k \epsilon_{k} c_{k,p,σ} \). In the third order, there are other contributions leading to the correction to the normal processes, which are disregarded in the following discussion due to eqs.(4)-(8) being large enough for the present choice of parameters.

Here we utilize the bosonization method by introducing phase variables for the charge (spin) fluctuation, \( \theta_ρ \) and \( ϕ_σ \), which are defined by

\[ \theta_ρ = \sum_q \sqrt{\theta_{ρ}(q)} e^{-α/q^2 - i y_k} \sum_k \sqrt{\theta_{ρ}(q)} e^{-α/q^2 - i x_k} \theta_{ρ}(q) \]

and

\[ \phi_σ = \sum_q \sqrt{ϕ_σ} e^{-α/q^2 - i y_k} \sum_k \sqrt{ϕ_σ} e^{-α/q^2 - i x_k} \phi_σ \]

respectively for \( \rho = (σ) \). The electron operator is expressed as

\[ ψ_{p,σ} = (2πα)^{-1/2} \exp\left( i\rho_σ(ϕ_σ + σ_ρ \phi_σ) \right) \int dϕ_σ \]
2k_F-CDW and 4k_F-CDW are given by
\[
O_{2k_F-SDW} = \sum_{p \sigma} \sigma e^{-i 2k_F p \cdot \mathbf{r}_p} \psi^\dagger_{p,\sigma} \psi_{-p,\sigma} \\
\propto \sin(2k_F x + \theta_\rho) \sin \theta_\sigma, \\
O_{2k_F-CDW} = \sum_{p \sigma} e^{-i 2k_F x} \psi^\dagger_{p,\sigma} \psi_{-p,\sigma} \\
\propto \cos(2k_F x + \theta_\rho) \cos \theta_\sigma, \\
O_{4k_F-CDW} = \sum_{p} e^{-i 4k_F x} \psi^\dagger_{p+} \psi^\dagger_{-p-} \psi_{-p-} \psi_{-p+} \\
\propto \cos(4k_F x + 2\theta_\rho).
\]

We investigate the possible states in the limit of low energy. Since the scaling dimension of H′ given by \(2 - 8K_\rho - 2K_\sigma\) is smaller than that of the other nonlinear terms, the term may be safely neglected for determining the phase diagram. In this case, the Hamiltonian is divided into the charge part and spin part. For the spin part, as long as \(g_{\perp} > 0\), the quantities, \(g_{\perp}\) and \(K_\sigma\) tend toward 0 and 1, respectively, for the low energy limit, due to SU(2) symmetry, and the excitation is gapless. Then the long range correlation functions are given as \(\langle \sin \theta_\sigma(x) \sin \theta_\sigma(0) \rangle \sim x^{-1} \ln^{1/2}(x)\) and \(\langle \cos \theta_\sigma(x) \cos \theta_\sigma(0) \rangle \sim x^{-1} \ln^{-3/2}(x)\). The low energy property for the charge part is determined by the following RG equations,
\[
\frac{d}{dl} K_\rho(l) = -8G_{1/4}^2(l)K_\rho^2(l),
\]
\[
\frac{d}{dl} G_{1/4}(l) = [2 - 8K_\rho(l)]G_{1/4}(l),
\]
with the initial conditions given by \(K_\rho(0) = K_\rho\) and \(G_{1/4}(0) = g_{1/4}/(2\pi v_\rho)\). Here \(l = \ln(\alpha'/\alpha)\) with the new length scale \(\alpha'\) larger than \(\alpha\). The solution of the above RG equations is obtained by
\[
G_{1/4}^2(l) - 1/(2K_\rho(l)) - 2\ln K_\rho(l) = \text{const},
\]
with a decrease of \(K_\rho\) with increasing \(l\).

The RG flows are shown in Fig.1, where the value of \(\alpha \simeq 2a/\pi\) is used. In the regions (I) and (II), the nonlinear term becomes irrelevant, indicating a metallic state. The correlation functions with long distance are given by \(\langle \sin \theta_\rho(x) \sin \theta_\rho(0) \rangle \sim (\cos \theta_\rho(x) \cos \theta_\rho(0) \rangle \sim x^{-K_\rho}(\infty)\) and \(\langle \cos 2\theta_\rho(x) \cos 2\theta_\rho(0) \rangle \sim x^{-4K_\rho}(\infty)\). Therefore, the domain state exhibits a crossover between 2k_F-SDW and 4k_F-CDW at \(K_\rho(\infty) = 1/3\), where \(K_\rho(\infty)\) is determined by \(G_{1/4}(0) - 1/(2K_\rho(0)) - 2\ln K_\rho(0) = -1/(2K_\rho(\infty)) - 2\ln K_\rho(\infty)\). The phase boundary determined by \(K_\rho(\infty) = 1/3\) is shown by the dashed curve. Both regions, (III) and (IV) are insulating states where \(G_{1/4} \rightarrow -\infty\) and \(\theta_\rho\) is locked at 0, \(\pi/2\) mod \(\pi\) in (III), and \(G_{1/4} \rightarrow \infty\) and \(\theta_\rho = \pm \pi/4\) mod \(\pi\) in (IV). The ordered state of 4k_F-CDW leading to charge ordering is realized in (III) and \(\theta_\rho = 0\) (\(\theta_\rho = \pi/2\)) corresponds to the state in which the charge is rich at the even (odd) sites. The closed circle, open circle and closed square correspond to the initial condition of \(V/t = 0, 2\) and 6, respectively, with \(U/t = 5\), showing that the metallic state with 2k_F-SDW in (I) moves to the insulating state with the 4k_F-CDW ordering with increasing \(V\).

The phase diagram on the plane of \(V/t\) and \(U/t\) is shown in Fig.2. The insulating phase appears for a large strength of \(U\) and \(V\). In addition, for large \(V\) and small \(U\), \(K_\rho(\infty)\) approaches unity indicating that the superconducting fluctuation is enhanced in this region. Figure 2 is qualitatively the same as that derived from the numerical diagonalization. In the insulating phase, which corresponds to the region (III) in Fig.1, the 4k_F-CDW, i.e., charge disproportionation, is realized. The state is consistent with the mean-field result. Thus, the insulating state obtained by the numerical diagonaliza-
tion is due to the formation of charge disproportionation, which originates in both $U$ and $V$ interactions. Here, we discuss the effects of the nonlinear term, $\cos 4\theta_\rho \cos 2\theta_{\rho'}$ of eq.(12), which may give minor changes on the phase boundary. In the insulating region, the term seems to give the correction of $g_{21}$ as $g_{21} \to g_{21} + g_{21}' \langle \cos 4\theta_\rho \rangle / 2$ with $\langle \cos 4\theta_\rho \rangle > 0$. Such a coupling constant gives rise to the spin gap. However, the procedure breaks the $SU(2)$ symmetry obviously. Therefore, it is expected that the third order correction of $K_{2\sigma}$, which has been discarded in the present treatment, would restore the symmetry, and the spin excitation remains gapless in the insulating region.

Within the present analysis, the insulating state exhibits the fluctuation of $2k_F$-SDW, while $4k_F$-CDW exists as the true long range order. When three dimensionality is introduced by the interchain coupling, the fluctuation of $2k_F$-SDW becomes the true order. However, the characteristic temperature of $2k_F$-SDW is expected to be smaller than that of $4k_F$-CDW. This conclusion may be relevant to the following experimental observation in (Dl-DNQI)$_2$Ag. The magnetic order of $2k_F$-SDW is observed at 5.5 K, whereas the resistivity shows insulating behavior at least below room temperature. In addition, charge disproportionation is observed below 220 K.

Finally, the effects of the next-nearest-neighbor repulsion, $V_2$, are briefly discussed. Up to the second order perturbation in the present formulation, the properties of the charge fluctuation do not show a qualitative change. On the other hand, the coefficient of the nonlinear term of the spin fluctuation, $g_{1\perp}$, is changed as $(U a/2 - V_2 a)\{1 - 4D_1 (U a/2 - V a + V_2 a)\}$. Since the nonlinear term with positive (negative) $g_{1\perp}$ favors $2k_F$-SDW ($2k_F$-CDW), it is obvious that the repulsive next-nearest-neighbor interaction stabilizes $2k_F$-CDW. In fact, the spin excitation becomes gapped and $2k_F$-CDW is realized when $U/2 < V_2$, independent of the strength of $V$ as seen in the mean-field result. Thus the quantity $V_2$ stabilizes (suppresses) $2k_F$-CDW ($2k_F$-SDW). However, the coexistence of two kinds of $2k_F$ density waves, which is predicted by the mean-field theory, cannot be understood from such a calculation, which needs further exploration.

In conclusion, we investigated both normal scattering and Umklapp scattering analytically for one-dimensional quarter-filled electron systems with on-site and nearest-neighbor interactions. We have shown that the increase of both $U$ and $V$ leads to the insulating state with charge disproportionation and that the next-nearest-neighbor interaction suppresses (enhances) the fluctuation of $2k_F$-SDW ($2k_F$-CDW).

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