Crossover from Quarter-Filling to Half-Filling in a One-Dimensional Electron System with a Dimerized and Quarter-Filled Band

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The interplay between quarter-filled and half-filled umklapp scattering has been examined by applying the renormalization group method to a one-dimensional quarter-filled electron system with dimerization, on-site (U) and nearest-neighbor (V) repulsive interactions. The phase diagram on the U-V plane is obtained at absolute zero temperature where the Mott insulator (the charge ordered insulator) is found for smaller (larger) V. By choosing the moderate parameter in the region of Mott insulator, it is shown that the resistivity exhibits a crossover from behavior of quarter-filling to that of half-filling with decreasing temperature.

KEYWORDS: quarter-filling, dimerization, half-filling, umklapp scattering, Bechgaard salts

The organic conductors, Bechgaard salts, which show a crystal structure with a stacking of organic molecules along one-dimensional axis, form a quasi-one-dimensional system with a quarter-filled hole band. Recently, the states above the phase transition temperature have been studied extensively to show several experimental observations which may be relevant to the unconventional non-Fermi liquid. Optical and photoemission experiments exhibit the charge gap in both TMTTF salts and TMTSF salts. The charge ordered (CO) insulating state in (TMTTF)2AsF6 and (TMTTF)2PF6 has been maintained by the NMR measurement and the huge dielectric response in (TMTTF)2PF6 indicates also the CO state. It is necessary to understand these experimental facts in consistent with a general phase diagram of the Bechgaard salts indicating a dimensional crossover.

Two mechanisms for the insulating state have been proposed theoretically. One is of them is the half-filled umklapp scatterings induced by dimerization and the charge gap has been examined for the one-dimensional quarter-filled Hubbard model with the dimerization. The other is the commensurability at the quarter-filling. The quarter-filled extended Hubbard model without the dimerization has been examined by using a method of numerical diagonalization, where the phase diagram shows the insulating state with the charge gap for a large Coulomb repulsion. The similar phase diagram has been obtained analytically by calculating the magnitude of the umklapp scattering at quarter-filling and by using the bosonization method and the renormalization group technique. The model treating both of these two umklapp scatterings on the same footing is required for investigating Bechgaard salts. By using the DMRG method, the charge gap of the model has been estimated where the metallic state is reduced to the insulator in the presence of dimerization.

In the present letter, we investigate the interplay of the half-filled umklapp scattering and the quarter-filled one by extending the previous work to the case including the dimerization. It is demonstrated that the competition between these two scatterings gives rise to two kinds of unconventional insulating states in the ground state and that a crossover between these two states appears in the resistivity at finite temperatures.

We consider a one-dimensional quarter-filled extended Hubbard model with dimerization. The Hamiltonian is given by

\[ H = -\sum_{j,\sigma} \left(t + (-1)^j t_d\right) (c_{j,\sigma}^\dagger c_{j+1,\sigma} + \text{h.c.}) + U\sum_{j} n_{j,\uparrow} n_{j,\downarrow} + V\sum_{j} n_j n_{j+1}, \]

where \( n_{j,\sigma} = c_{j,\sigma}^\dagger c_{j,\sigma} \) and \( n_j = n_{j,\uparrow} + n_{j,\downarrow} \). The fermion operator \( c_{j,\sigma}^\dagger \) denotes a creation of the electron at the \( j \)-th site with spin \( \sigma \). We have two kinds of transfer integrals \( t + t_d \) and \( t - t_d \) due to the dimerization. The quantities \( U \) and \( V \) denote the on-site and nearest-neighbor Coulomb repulsive interaction. In the presence of the dimerization, the unit cell is given by two lattice sites with 2a where \( a \) is the lattice constant. By introducing operators \( c_{R_n,\sigma}^A \) and \( c_{R_{n+a},\sigma}^B \) which are those for the even and odd sites in the \( n \)-th unit cell (\( R_n = 2an \)), respectively, we perform the Fourier transform \( c_{R_n,\sigma}^A = N_0^{-1/2} \sum_{k} e^{-ikR_n} c_{k,\sigma}^A \) and \( c_{R_{n+a},\sigma}^B = N_0^{-1/2} \sum_{k} e^{-ik(R_n+a)} c_{k,\sigma}^B \), where \( N_0 = L/(2a) \) and \( L \) is the length of the system. Then the kinetic term of the Hamiltonian, \( H_0 \), is diagonalized as \( H_0 = \sum_{k,\sigma} \varepsilon_k (d_{k,\sigma}^A c_{k,\sigma}^A + e^{-i\theta_k} d_{k,\sigma}^B c_{k,\sigma}^B) \) with a dispersion relation, \( \varepsilon_k = -2\left|t^2 \cos^2 ka + t_d^2 \sin^2 ka\right|^1/2 \). In a diagonalized basis, the lower and upper band fermion operators are given by \( d_{k,\sigma} = [c_{k,\sigma}^A + e^{-i\theta_k} c_{k,\sigma}^B]/\sqrt{2} \) and

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where \( \phi \) is decoupled into two parts of the charge density wave (CDW) state, shown in terms of phase variable. The sign of \( \phi \) is locked to \( \Omega_{k} \) for \( g_{1/4} < 0 \) or \( g_{1/4} > 0 \) for \( \rho \).

In order to obtain the effective Hamiltonian for the states near \( \pm k_{F} \), one integrates out the contribution from the upper band, which leads to the quarter-filled umklapp scattering. We apply the bosonization method to electrons in the lower band with the dispersion linearized as \( \epsilon_{k} \rightarrow \pm v_{F}|k| - k_{F} \) where \( v_{F} = \sqrt{2a[1 - (t_{d}/t)^{2}]/[1 + (t_{d}/t)^{2}]^{1/2}} \). Then the Hamiltonian, which is rewritten in terms of the charge and spin phase variable, is decoupled into two parts of the charge and spin degrees of freedom. The charge part, \( H_{\rho} \), is expressed as:

\[
H_{\rho} = \frac{v_{F}}{4\pi} \int dx \left[ \frac{1}{K_{\rho}} (\partial_{x} \phi_{\rho})^{2} + K_{\rho} (\partial_{\rho} \phi_{\rho})^{2} \right] - \frac{g_{1/2}}{2\pi^{2}\alpha^{2}} \int dx \sin 2\theta_{\rho} + \frac{g_{1/4}}{2\pi^{2}\alpha^{2}} \int dx \cos 4\theta_{\rho} ,
\]

where \( \alpha \) is a cutoff of the order of lattice constant and \( [\rho(x), \phi_{\rho}(x')] = i\pi \text{sgn}(x - x') \). In eq. (2), the charge velocity and the Tomonaga-Luttinger exponent are given by \( v_{\rho} = v_{F}[(1 + g_{4p}/2\pi v_{F})^{2} - (g_{p}/2\pi v_{F})^{2}]^{1/2} \) and \( K_{\rho} = [(2\pi v_{F} + g_{4p} - g_{p})/(2\pi v_{F} + g_{4p} + g_{p})]^{1/2} \). The magnitude of half-filled umklapp scattering, \( g_{1/2} \), and that of quarter-filled one, \( g_{1/4} \), are given by:

\[
g_{1/2} = B \left[ Ua - \frac{Aa^{2}}{2\pi v_{F}} U(U - 2V) I_{2}(A) \right] ,
\]

\[
g_{1/4} = \frac{1}{(2\alpha \rho)^{2}} A^{4} a^{5} U^{2}(U - 4V) ,
\]

where \( A \equiv [1 - (t_{d}/t)^{2}]/[1 + (t_{d}/t)^{2}] \) and \( B \equiv [2t_{d}/t]/[1 + (t_{d}/t)^{2}] \). The quantity \( g_{1/4} \) has been derived for \( t_{d} = 0 \). The constants \( g_{p} \) and \( g_{4p} \) are given by:

\[
g_{p} = (U + 4V)a
\]

\[
- \frac{Aa^{2}}{4\pi v_{F}} \left( (U - 2V)^{2} + U^{2} + (2V)^{2} \right) I_{2}(A) ,
\]

\[
g_{4p} = (U + 4V)a
\]
two kinds of non-linear terms, i.e., the gap collapses on the Ising transition found in the sine-Gordon system with non-linear terms. We calculate the temperature dependence of the resistivity using the memory functional approach in the presence of non-linear terms. In this case, the resistivity is given by

$$\sigma(\omega) = \frac{2v_p K_\rho}{\pi} \frac{1}{\omega + M(\omega)}$$

where $M(\omega)$ is the memory function defined by

$$M(\omega) = \frac{\omega \chi(\omega)}{\chi(0) - \chi(\omega)}.$$  

The memory function is calculated perturbatively as

$$M(\omega) \simeq -\frac{1}{\omega} \left( \Gamma(\omega, \omega) - \Gamma(\omega, 0) \right) / \chi(0).$$

In terms of $M(\omega)$, the resistivity is expressed as

$$\rho(T) = \lim_{\omega \to 0} \frac{\sigma^{-1}(\omega)}{\omega} = \frac{\pi}{2v_p K_\rho} \lim_{\omega \to 0} M(\omega).$$

Equation (10) indicates the power-law behavior,

$$\rho(T) \propto \sum_{n=1,2} G_{1/2n}^2 T^{4n^2 K_\rho - 3}.$$  

In order to obtain the precise behavior of the resistivity, we use the solutions of the renormalization group equations, in which the umklapp scattering becomes relevant at low temperature. By putting $e^{-t} = 2\pi T/W$ with $W \equiv v_p \alpha^{-1}$, the resistivity is obtained as

$$\rho(T) = \frac{4n^2 \pi^2}{\alpha} G_{1/2n}(l)^2 \cos^2 \left( n^2 \pi K_\rho(l) \right) B^2 \left( n^2 K_\rho(l), 1 - 2n^2 K_\rho(l) \right) e^{-1}.$$  

where $B(x, y)$ is the beta function given by $B(x, y) = \Gamma(x) \Gamma(y) / \Gamma(x + y)$. In Fig. 4, the temperature dependence of the resistivity is shown for $U/t = 6$ and $t_{d4}/t = 0.001$ with fixed $V/t = 4.60$ (a) and 4.75 (b). The curve (a) shows the behavior of the Mott insulator, while the curve (b) represents that of the CO insulator. For the CO insulator (curve (b)), the resistivity is determined by the quarter-filled umklapp scattering, where the resistivity exhibits the power-law dependence given by $\rho(T) \propto T^{16K_\rho - 3}$ around the temperature $T/W \gtrsim 1 \times 10^{-1}$. The temperature $T/W \approx 5 \times 10^{-2}$,
corresponding to a minimum of the resistivity, is of the order of the charge gap $\Delta_p$ below which the resistivity increases rapidly. It is expected that the resistivity for $T \ll \Delta_p$ behaves as $\exp(\Delta_p/T)\propto T^{-3}$. At higher temperatures ($T/W > 4\times 10^{-2}$), the contribution of the quarter-filled umklapp scattering becomes dominant at which the resistivity is similar to the case of curve (b). Such a crossover in the region of CO insulator can be obtained by the moderate choice of parameter with $|G_{1/4}| > G_{1/2}$, i.e., close to the boundary in Fig. 3. The similar result is found in the frequency dependence of the conductivity at $T = 0$. While the optical conductivity of the CO insulating phase corresponding to curve (b) shows the power-law dependence with $\sigma(\omega) \propto \omega^{2K_r-5}$ above the frequency of the charge gap, the conductivity of the Mott insulator corresponding to curve (a) shows a crossover from $\sigma(\omega) \propto \omega^{6K_r-5}$ to $\sigma(\omega) \propto \omega^{4K_r-5}$ as decreasing frequency. Thus, the crossover from quarter-filling to half-filling is found with decreasing temperature or frequency when parameters are located in the Mott insulating region and close to the boundary.

Finally we comment on observation in the Bechgaard salts, (TMTTF)$_2$X and (TMTSF)$_2$X. The optical measurement in the normal states of these salts shows that the optical conductivity has a power-law dependence as $\sigma(\omega) \propto \omega^{-1.3}$ at high frequency, indicating a fact that the quarter-filled umklapp scattering is dominant. Our calculation suggests that the quarter-filled umklapp scattering becomes always dominant at high frequency when the parameter is close to the boundary. Therefore the power-law dependence of the optical conductivity $\sigma(\omega) \propto \omega^{6K_r-5}$ could be compatible with the charge gap, which originates in the Mott insulator and/or the CO insulator.

In conclusion, we found two kinds of insulator, the Mott insulator and the CO insulator, for the ground state of a one-dimensional quarter-filled system with dimerization, where the half-filled umklapp scattering induced by the dimerization competes with the quarter-filled umklapp scattering for large nearest-neighbor Coulomb interaction $V (>U/4)$. In the Mott insulating phase close to the boundary, the half-filled umklapp scattering is dominant at low $T$ but the quarter-filled umklapp scattering becomes dominant at high $T$, and then the resistivity could exhibit a crossover from a quarter-filled behavior to a half-filled one as decreasing temperature.

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