Charge order and superconductivity in low-dimensional organic conductors

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We study the coexistent state of the spin density wave (SDW) and the charge density wave (CDW) in a one-dimensional extended Hubbard model. We find that the coexistent state of SDW and CDW is stabilized in various electron-filling cases when band parameters are for organic conductors. The ground state energies have cusp-like minima at $n/4m$-fillings, where n and m are integers. The maxima of the critical temperatures of the coexistent state at $n/4m$-fillings will be observed in X-ray scattering measurements. We discuss the suitable filling for superconductivity of which strong fluctuation in antiferromagnetic ordering may be the origin.

Keywords: stripe-type order; **superconductivity**; **strongly correlated system**; **low-dimensional organic conductors.**

1. Introduction

In quasi-one-dimensional quarter-filled organic conductors such as (TMTSF)$_2X$ and (TMTTF)$_2X$ (X=PF$_6$, Br, etc.), superconductivity and the coexistent phase of CDW and SDW are observed (Ishiguro *et al.*, 1998). Pouget and Ravy (Pouget & Ravy, 1997) observed the coexistence of $2k_F$-SDW and $2k_F$-CDW by the X-ray scattering measurement in (TMTSF)$_2$PF$_6$ at the ambient pressure, where $k_F = \pi/4a$ is the Fermi wave number and a is the lattice constant. Under pressure (12 kbar), the superconducting phase appears at 0.9 K (Jerome *et al.*, 1980).

In (TMTTF)$_2$Br, $4k_F$-CDW accompanied with $2k_F$-SDW is found in X-ray scattering measurements (Pouget & Ravy, 1997). The superconductivity transition occurs at 0.8 K under 26 kbar (Balicas *et al.*, 1994).

Even in quasi-two-dimensional quarter filled organic conductors such as (BEDT-TTF)$_2X$, the coexistent state of CDW and SDW is observed as the stripe-type order (Nakamura *et al.*, 2000). This coexistent state in quasi-two-dimensional systems has been theoretically studied (Seo, 2000). Recently, it has been found that the ground state in X=KHg changes from the charge ordering (Miyagawa *et al.*, 1997), which causes the stripe-type order, to superconductivity under uniaxial pressure (Maesato, 2000). Moreover, in the high-T_c cuprates such as La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ (Tranquada *et al.*, 1997), the stripe-type order is most stabilized when the rate of the doping is $x = 1/8$ and the critical temperature of the superconductivity becomes the highest at near $1/8$. From these experiments, it is expected that the superconductivity appears near the coexistent state of CDW and SDW. This may be due to the fluctuation in antiferromagnetic ordering of the stripe-type order, because it is understood that the superconductivity in high-T_c cuprates is due to strong fluctuation in antiferromagnetic ordering (Anderson, 1996).

In one-dimensional quarter-filled system, these two kinds of coexistent states ($2k_F$-SDW-$2k_F$-CDW and $2k_F$-SDW-$4k_F$-CDW) have been studied theoretically (Seo & Fukuyama, 1997; Kobayashi *et al.*, 1998; Mazumdar *et al.*, 1999; Tomio & Suzumura, 2000; Kishigi & Hasegawa, 2000). From the consideration of inter-site Coulomb interaction (V) in addition to the on-site Coulomb interaction (U), the CDW-SDW coexistent state has been understood as being caused by the interplay between U and V (Seo & Fukuyama, 1997; Kobayashi *et al.*, 1998; Mazumdar *et al.*, 1999; Tomio & Suzumura, 2000; Kishigi & Hasegawa, 2000). Here, V is important, since Mila (Mila, 1995) has estimated $U/t \sim 5$ and $V/t \sim 2$ in quasi-one-dimensional organic conductors, where t is a transfer integral.

In this paper, we study how the electron-filling (f) affects the coexistent state of SDW and CDW due to V. By calculating the condensation energy, we find f values at which the coexistent state in one-dimensional systems is stabilized. The f-dependence of the ground state energy in the one-dimensional extended Hubbard model has never been studied, although the f-dependence of the one in the Hubbard model has been calculated (Carmelo & Baeriswly, 1988). Based on the result, we propose f values which favor superconductivity in strongly correlated one-dimensional systems with V.

2. Model

The one-dimensional extended Hubbard model is, $\hat{\mathcal{H}} = \hat{\mathcal{K}} + \hat{\mathcal{U}} + \hat{\mathcal{V}}$; $\hat{\mathcal{K}} = -t\sum_{i,\sigma}(c^{\dagger}_{i,\sigma}c_{i+1,\sigma} + h.c.)$; $\hat{\mathcal{U}} = U\sum_i n_{i,\uparrow}n_{i,\downarrow}$; $\hat{\mathcal{V}} = V\sum_{i,\sigma,\sigma'} n_{i,\sigma}n_{i+1,\sigma'}$, where $c^{\dagger}_{i,\sigma}$ is the creation operator of σ spin electron at i site, $n_{i,\sigma}$ is the number operator, $i = 1, \cdots, N_S$, N_S is the number of the total sites and $\sigma =\uparrow$ and $\downarrow$. We use parameter, $U/t = 5.0$, and $0 \leq V \leq U$.

The interaction terms, $\hat{\mathcal{U}}$ and $\hat{\mathcal{V}}$, are treated in the mean field approximation as $\hat{\mathcal{U}}^{\mathrm{M}}$ and $\hat{\mathcal{V}}^{\mathrm{M}}$ and the self-consistent equation for the order parameter $\rho_{\sigma\sigma}(Q)$ is given by $\rho_{\sigma\sigma}(Q) = I\sum_{k_x} < C^{\dagger}(k_x,\sigma)C(k_x - Q,\sigma) >$, where $I = U/N_S$. We consider the various electron-fillings, $f = q/p$, where q and p are mutually prime numbers. We take the possible wave vectors of the order parameters as $Q = nQ_0$, where $Q_0 = 2k_F = (2\pi/a)\cdot f$ and $n = 1, \cdots, p$.

The condensation energy, E_c, is given by $E_c = E_g(\rho) - E_N$, where E_N and E_g are the normal state energy and the ground state energy for the ordered states.

The electron density and the spin moment at site j are $n(j) = \frac{1}{U}\sum_{Q,\sigma}\rho_{\sigma\sigma}(Q)e^{iQja}$ and $S_z(j) = \frac{1}{2U}\sum_Q(\rho_{\uparrow\uparrow}(Q) - \rho_{\downarrow\downarrow}(Q))e^{iQja}$, respectively.

3. Results

We search for the most stable self-consistent solutions by changing the initial values of the order parameters. Since $n(j)$, $S_z(j)$ and E_c for $f > 0.5$ is the same as those for $f < 0.5$ due to the symmetry between an electron and an hole and we do not focus our attention on the low electron-filling, we calculate at various fillings $(2 \leq p \leq 20, 1 \leq q \leq 9)$ in the region of $0.2 \leq f \leq 0.5$.

At $f = 1/2$, the stable state is $2k_F$-SDW, $(\downarrow,\uparrow)$, $(S_z(1) = -S_z(2) \simeq 1.0$ and $n(1) = n(2) \simeq 1.0)$ for $2V \leq U$ and the state is changed to $2k_F$-CDW, $(0,\downarrow\uparrow)$, $(S_z(1) = S_z(2) \simeq 0$ and $n(1) \simeq 0$ and $n(2) \simeq 2.0)$ for $2V > U$, where the arrows mean the spin moment, 0 means small or zero electron density and $\downarrow\uparrow$ represents that the up and down electrons exist in the same site. For $2V > U$, the period of the CDW becomes two.As this charge order is suitable for the nearest Coulomb repulsion interaction, the large charge gap made by the CDW exists at large V. As a result, E_c becomes lower, which will be shown in Fig. 5. This result at the half-filling has been shown by Cabib and Callen (Cabib & Callen, 1975).

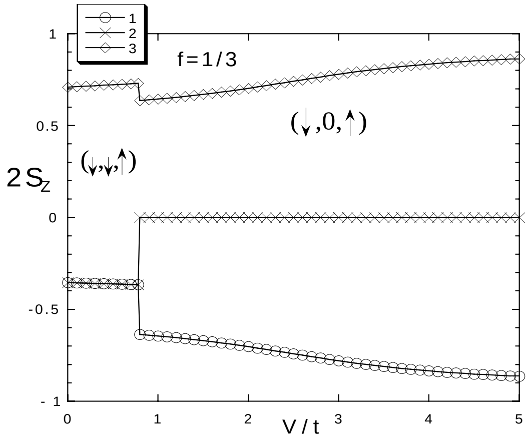

Figure 1
$2S_z(1)$, $2S_z(2)$ and $2S_z(3)$ as a function of V/t.

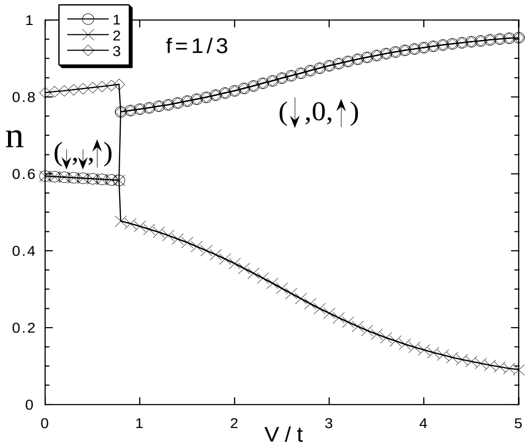

Figure 2
$n(1)$, $n(2)$ and $n(3)$ as a function of V/t.

At $f = 1/3$, the stable state is the coexistence of SDW and CDW, $(\downarrow,\downarrow,\uparrow)$, ($|S_z(1)| = |S_z(2)| < S_z(3)$ and $n(1) = n(2) < n(3)$) for $0 \leq V/t < 0.8$, where the distortion of the charge density is small. The coexistent state of SDW and CDW, $(\downarrow, 0, \uparrow)$ ($S_z(1) = -S_z(3), S_z(2) = 0$, $n(1) = n(3) \simeq 1.0$ and $n(2) \simeq 0$), is stabilized for $V/t \geq 0.8$. These results are shown in Figs. 1 and 2. As the period of n in $(\downarrow, 0, \uparrow)$ is three, there is a frustration for V and the energy gap does not become large. Since the order parameters having $Q = (2\pi/a) \cdot (1/2)$ does not exist when p of $f = q/p$ is an odd number, the period of n does not become two. Thus, when p is an odd number, the order of the CDW is not suitable for V.

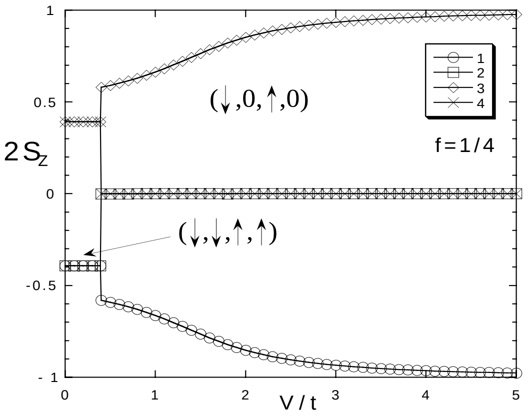

Figure 3
$2S_z(j)$ ($j = 1 \cdots 4$) as a function of V/t.

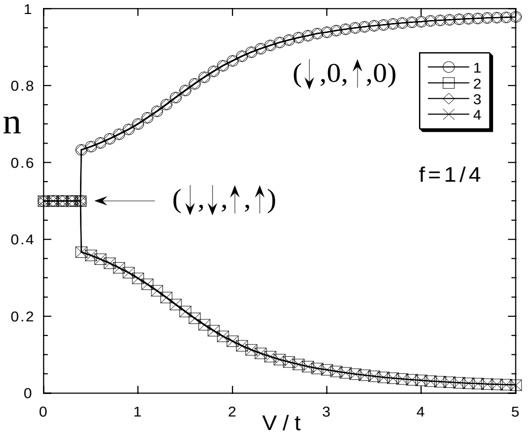

Figure 4
$n(j)$ ($j = 1 \cdots 4$) as a function of V/t.

At $f = 1/4$, the stable state of $2k_F$-SDW, $(\downarrow,\downarrow,\uparrow,\uparrow)$ ($S_z(1) = S_z(2) = -S_z(3) = -S_z(4)$ and $n(1) = n(2) = n(3) = n(4) = 0.5$), is changed to $2k_F$-SDW and $4k_F$-CDW coexistent state, $(\downarrow, 0, \uparrow, 0)$ ($S_z(1) = -S_z(3), S_z(2) = S_z(4) = 0$, $n(1) = n(3) \simeq 1.0$ and $n(2) = n(4) \simeq 0$), for $V/t > 0.39$, as shown in Figs. 3 and 4. This result is consistent with the previous study (Seo & Fukuyama, 1997). For $V/t > 0.39$, as the period of the charge density is two, E_c becomes large due to the large energy gap, which will be shown in Fig. 5. The CDW-SDW coexistent state is stabilized except for $f = 1/2$.

We show E_c in Fig. 5 as a function of f for various V. At $V = 0$, E_c monotonically decreases as f increases, because E_c is mainly determined by the density of states on the Fermi surface, $N(0) = N_S/(4\pi t \sin ak_F)$, which decreases as f increases. We can see that $|E_c|$ for $f = 1/2, 1/4, 3/8, 5/12$, 7/16, 7/20 and 9/20 become large upon increasing V. It is found that the cusp-like minima of E_c appear at $f = n/4m$, where n and m are integers. In these fillings, the period of n accompanying the order of the SDW becomes two, resulting in the large energy gap. The local minimum at $f = 5/16$ may be too small to see in Fig. 5.

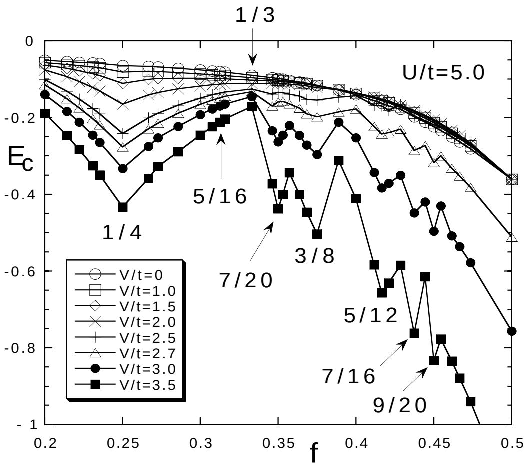

Figure 5
E_c as a function of f for various V.

4. Discussions

In the coexistent state of CDW and SDW, the commensurability of f and V play an important role in the condensation energy. At $f = n/4m$, the coexistent state of CDW and SDW is compatible with the favored real-space order for both U and V. At $f = 1/3$, E_c changes little as V increases. This feature can be understood by considering that frustration occurs in the order of period three; double occupancy should be reduced by U and electrons at nearest sites should be avoided for large V.

The maxima of the critical temperatures of the coexistent state at $f = n/4m$ can be confirmed in X-ray scattering measurement when the electron-filling is changed in quasi-one-dimensional organic conductors. Because the charge ordered temperature can be estimated by X-ray scattering measurements.

In the case of one-dimensional systems with V where the superconductivity is attributable to fluctuation in the antiferromagnetic ordering, the superconductivity may appear at the fillings near $f = n/4m$. If the origin of the superconductivity of quasi-one-dimensional organic conductors is so, the superconductivity will be obtained by changing electron-filling slightly from the quarter-filling.

5. Conclusion

We study the coexistent state of CDW and SDW in the one-dimensional system with V by changing f. We find that the coexistent state is stabilized at $f = n/4m$ due to V, which will be observed in X-ray scattering measurements. Since strong fluctuation in antiferromagnetic ordering is expected in the region close to the coexistent state, the superconductivity will appear near $f = n/4m$.

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