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Collective Excitations around Charge Ordered States and Coexistent States with Different Orders

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We study qualitative differences of excited states between a charge ordered state and a charge uniform state. The excited states are calculated in the random phase approximation for the one-dimensional extended Hubbard model at quarter filling. In the charge ordered state, a dominant excitation in the current-current correlation function has no charge density modulation. A dominant excitation in the charge uniform state originates from crossing the dimerization gap and modulates the charge density.

Keywords: charge ordering; linear excitation; intersite Coulomb repulsion; Hartree-Fock approximation; random phase approximation

INTRODUCTION

Some novel ground states have recently been studied in molecular conductors. One is the coexistence of a spin density wave (SDW) and a charge density wave (CDW) in $(TMTSF)_2PF_6$, which is quasi-one-dimensional and is basically a quarter-filled system with dimerization. Seo and Fukuyama[1] and Kobayashi *et al.* [2] have used the Hartree approximation for one-dimensional extended Hubbard models and found coexistence of a $2k_F$ SDW with a $4k_F$ CDW and that

with a $2k_F$ CDW, respectively. They claim that not only the on-site but also the nearest-neighbor and the next-nearest-neighbor repulsive interactions are important, respectively. Another novel state is the charge ordered state (COS) in θ-(BEDT-TTF)$_2$MM'(SCN)$_4$ (M=Rb,Cs,Tl, M'=Zn,Co), which is quasi-two-dimensional and is a quarter-filled system. Pressure changes the dihedral angle between the ET-molecular planes and causes a dimerized phase in addition to a uniform phase. Seo[3] used the Hartree approximation again for a two-dimensional extended Hubbard model and found various charge ordered states depending on anisotropy of the transfer integrals and that of the intersite Coulomb interaction strengths.

In this paper, we study excited states of the COS in one dimension by using the random phase approximation (RPA) and clarify effects of the dimerization and the intersite Coulomb interaction on the excitation spectra. First, we obtain the ground state in the unrestricted Hartree-Fock (UHF) approximation to confirm the previous results. Next, we calculate response functions in the RPA on the basis of the UHF states. We show that the characteristic features of the optical and magnetic excitation spectra depend on the dimerization and the intersite Coulomb interaction.

MODEL HAMILTONIAN AND GROUND STATES

We adopt the one-dimensional extended Hubbard model including the nearest neighbor interaction,

$$H = \sum_{\substack{<i,j> \\ \sigma=\uparrow,\downarrow}} t_{ij}(c^{\dagger}_{i,\sigma}c_{j,\sigma} + H.c.) + U\sum_{i} n_{i,\uparrow}n_{i,\downarrow} + V\sum_{<i,j>} n_i n_j, \quad (1)$$

where $c^{\dagger}_{i,\sigma}(c_{i,\sigma})$ is the electron creation (annihilation) operator with spin σ at site i, $n_{i,\sigma}$ is the electron number density, and $n_i = n_{i\uparrow}+n_{i\downarrow}$. The symbol $\langle i,j\rangle$ stands for nearest neighbor sites. The transfer integrals t_{ij} alternatively take two values, t_1 and t_2. The UHF calculation

is carried out for the system with 16 sites, quarter-filled and with the periodic boundary condition. We obtain different ground states, in

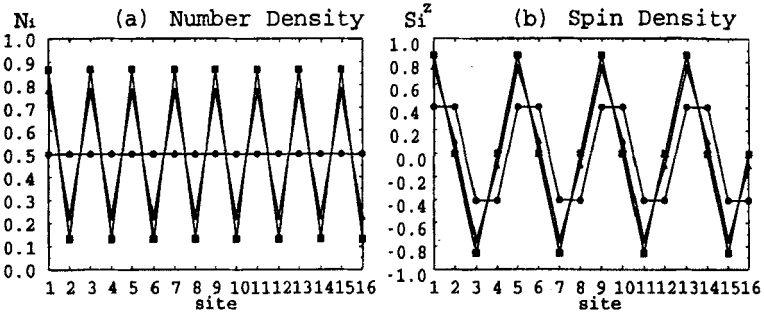

FIGURE 1 (a) Number density and (b) spin density for t_1/t_2=1.5, U=5, V=0 (circle); t_1/t_2=1.5, U=5, V=4 (triangle); and t_1/t_2=1, U=5, V=4 (square).

which the repulsive interaction between nearest neighbor sites causes the charge ordered state and the different magnetic orders as shown in Fig.1.[1, 2] The different ground states have their characteristic excitation spectra, as shown below.

EXCITED STATES AND ABSORPTION SPECTRA

We solve the linearized equation of motion for the particle-hole pairs that describe quantum fluctuations around the HF ground state. Details of the method are found in Refs.[4, 5]. All the energies of the linear excitations are plotted in Fig.2 in increasing order. The low-lying linear excitations in both Figs.2(a) and 2(b) are spin-wave modes that have different spin-wave velocities. In Fig.2(a), a unit of the localized magnetic moment is a dimer of molecules and the spin-wave velocity is estimated as $t_2^2/U = 1/5 = 0.2$, while a molecule is a unit in Fig.2(b) and the spin-wave velocity is estimated as $6t_2^2t_1^2/(U \cdot V^2) = 6/(5 \cdot 4^2) = 0.075$. Excitations modulating both spin and charge densities appear at high energies. Some of them are

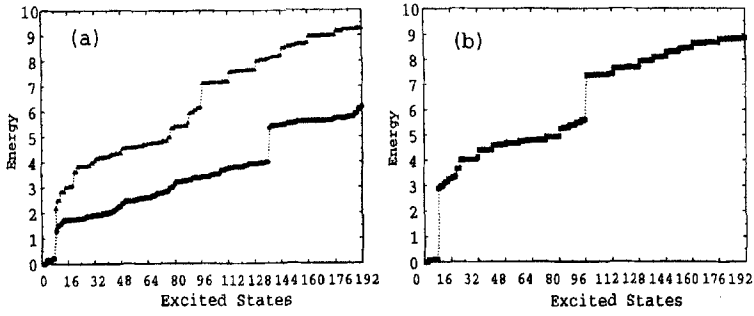

FIGURE 2 Energies of the linear excitations for (a) t_1/t_2=1.5, U=5, V=0 (circle); t_1/t_2=1.5, U=5, V=4 (triangle); and (b) t_1/t_2=1, U=5, V=4 (square), in increasing order.

detected by optical measurements. Then, we plot the imaginary part of the current-current correlation function in Fig.3. In Fig.3(a), the

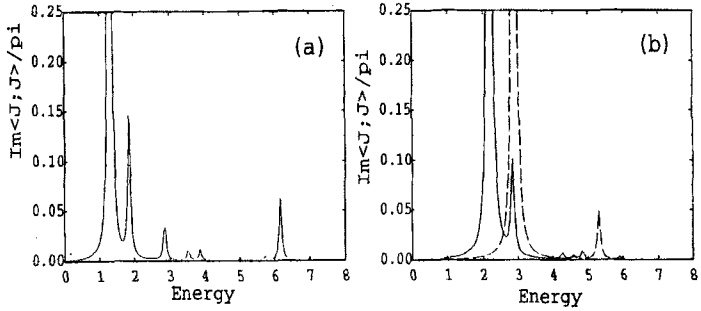

FIGURE 3 Imaginary part of the current-current correlation function for (a) t_1/t_2=1.5, U=5, V=0; (b) t_1/t_2=1.5, U=5, V=4 (solid line); and t_1/t_2=1, U=5, V=4 (dashed line).

peaks around $\omega \sim$1.3 and 1.9 correspond to the excitation from the bonding to the non-bonding orbitals of a dimer crossing the dimerization gap. The small peak around $\omega \sim$6.2 comes from the Hubbard gap due to the on-site repulsion. These characteristic features have

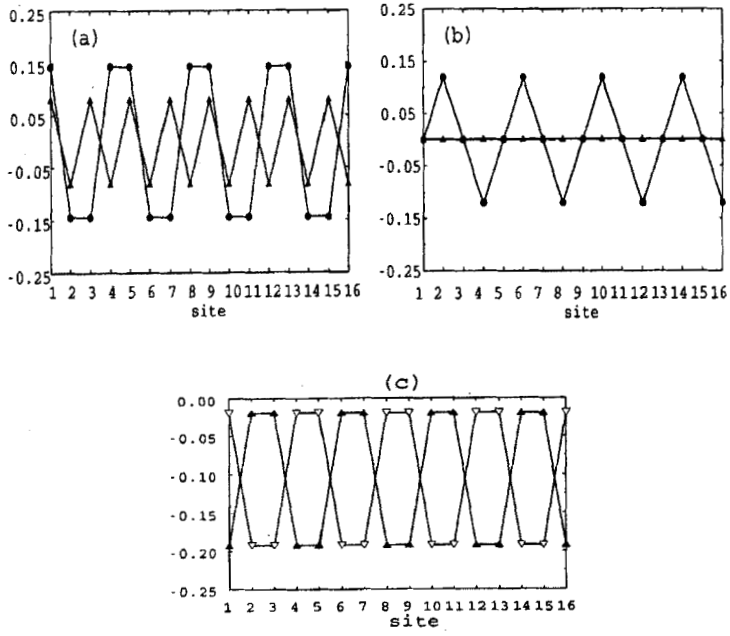

FIGURE 4 Matrix elements of S_i^z (circle) and n_i (triangle) between the ground state and the 9-th excited states for (a) t_1/t_2=1.5, U=5, V=0, and (b) t_1/t_2=1, U=5, V=4. The lower panel (c) shows those of $j_{i\sigma}$ between these two states for t_1/t_2=1, U=5, V=4. The upward and downward triangles indicate the elements of $j_{i\uparrow}$ and $j_{i\downarrow}$, respectively.

been obtained by the exact diagonalization of small clusters.[6] The dominant peaks of the solid line in Fig.3(b) around $\omega \sim 2.2$ and 2.9 also originate from the dimerization gap. The origin of the dominant peak of the dashed line in Fig.3(b) around $\omega \sim 2.9$ is different, since there is no dimerization. In order to see the different characters between the dimerized and uniform systems, the matrix elements of $S_i^z = n_{i\uparrow} - n_{i\downarrow}$ and $n_i = n_{i\uparrow} + n_{i\downarrow}$ between the ground state and the excited state are plotted in Fig.4(a) for the dimerized system and in Fig.4(b) for the uniform system. Both the spin and charge ampli-

tudes are modulated in Fig.4(a), while the charge amplitude is not modulated in Fig.4(b). The character of this excitation in the current channel without the charge modulation is clarified by plotting the matrix elements of $j_{i\sigma} \equiv t_{ij}(c^{\dagger}_{i+1\sigma}c_{i\sigma} - c^{\dagger}_{i\sigma}c_{i+1\sigma})$ in Fig.4(c). It is reasonable that the charge modulation does not occur and the spin modulation appears on the even sites. For example, on the second site, an up-spin electron comes in and a down spin electron goes out.

Acknowledgments

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