Excitonic phases in the two-orbital Hubbard model: Roles of Hund's rule coupling

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Abstract. Excitonic phases described by the quantum condensation of electron-hole pairs (or excitons) are studied in the two-orbital Hubbard model with the Hund's rule coupling J. Using the variational cluster approximation, we calculate the ground-state energy and order parameters, and show that the Hund's rule coupling always stabilizes the excitonic spin-density-wave state and destabilizes the excitonic charge-density-wave state. The pair coherence lengths calculated in the spin-singlet and spin-triplet states indicate that only the spin-triplet excitons are paired more tightly with increasing J. The single-particle spectrum and density of states are also calculated to clarify the characters of these excitonic density-wave states.

1. Introduction

The excitonic phases, which are often referred to as the excitonic insulator or excitonic density-wave states, are described by the quantum condensation of electron-hole pairs (or excitons) and were predicted to occur in a small band-gap semiconductor or a small band-overlap semimetal [1, 2, 3]. The exciton condensation in semimetallic systems can be described in analogy with the BCS theory of superconductors and that in semiconducting systems can be discussed in terms of the Bose-Einstein condensation (BEC) of preformed excitons [4]. The crossover phenomena between the BCS and BEC states are then expected to produce rich physics in the field of quantum many-body systems. A number of candidate materials for the excitonic phases have been discovered so far. Recent examples are the phase transitions of layered chalcogenides $1T$-TiSe$_2$ [5, 6, 7, 8] and Ta$_2$NiSe$_5$ [9, 10, 11, 12], where possible realization of the spin-singlet excitonic condensation has attracted much experimental and theoretical attention. The spin-density-wave state of iron-pnictide superconductors was also argued to be of the excitonic origin [13, 14, 15].

In this paper, motivated by the above development in the field, we study the stability of the excitonic density-wave states in the two-orbital Hubbard model. Using the variational cluster approximation (VCA) [16, 17, 18], we calculate the ground-state energy and order parameters, and show that the Hund's rule coupling always stabilizes the excitonic spin-density-wave (SDW) state and destabilizes the excitonic charge-density-wave (CDW) state. The pair coherence lengths (or the sizes of the electron-hole pair) in the spin-singlet and spin-triplet states are also calculated to show that only the spin-triplet excitons are paired more tightly with increasing J. We also calculate the single-particle spectrum and density of states (DOS) to see the characters of the excitonic density-wave states in detail.

2. Model and method

2.1. Two-orbital Hubbard model

To investigate the excitonic phases, we consider the two-orbital Hubbard model defined by the Hamiltonian

$$\mathcal{H} = -t \sum_{\langle i,j \rangle} \sum_{\sigma} \sum_{\alpha} \alpha_{i\sigma}^{\dagger} \alpha_{j\sigma} - D \sum_{i} (n_{if} - n_{ic}) - \mu \sum_{i} \sum_{\alpha} n_{i\alpha}$$

$$+ U \sum_{i} \sum_{\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} + U' \sum_{i} n_{if} n_{ic} - 2J \sum_{i} (\boldsymbol{S}_{if} \cdot \boldsymbol{S}_{ic} + \frac{1}{4} n_{if} n_{ic}) \qquad (1)$$

where $\alpha_{i\sigma}^{\dagger}$ $(= f_{i\sigma}, c_{i\sigma})$ denotes the creation operator of an electron with spin $\sigma (=\uparrow,\downarrow)$ on the α $(= f, c)$ orbital at site i and $n_{i\alpha} = n_{i\alpha\uparrow} + n_{i\alpha\downarrow} = \alpha_{i\uparrow}^{\dagger} \alpha_{i\uparrow} + \alpha_{i\downarrow}^{\dagger} \alpha_{i\downarrow}$. t is the hoping integral between the neighboring sites and D is the level splitting between the two orbitals. U and U' are the intra- and inter-orbital Coulomb repulsions between electrons. J is the Hund's rule coupling. We set the chemical potential as $\mu = U/2 + U' - J/2$ to maintain the average particle density $n = \sum_{i,\alpha,\sigma} \langle n_{i\alpha\sigma} \rangle / (2N)$ at half filling, $n = 1$. We note here that the excitonic phases in the spinless $(U = J = 0)$ case have been studied in detail using the extended Falicov-Kimball model [19, 20, 21, 22, 23] and that the effects of the intra-orbital Coulomb interaction U on the excitonic phases in the spinfull case have been studied without taking into account the Hund's rule coupling $(J = 0)$ [15, 24]; the effects of the Hund's rule coupling J have only been studied by the dynamical mean-field theory [25, 26].

2.2. Variational cluster approximation

To accomplish the calculations in the thermodynamic limit, we employ the method of VCA, which is based on the variational principle for the grand potential as a functional of the self-energy. The trial self-energy for the variational method is generated from the exact self-energy of the disconnected finite-size clusters, which act as a reference system. To investigate the spontaneous symmetry breaking in VCA, we introduce the Weiss fields as variational parameters. The variational Hamiltonians of the CDW and SDW states are then defined as

$$\mathcal{H}'_{\text{CDW}} = \mathcal{H} + \Delta_0' \sum_{i,\sigma} e^{i\boldsymbol{Q}\cdot\boldsymbol{r}_i} (c_{i\sigma}^{\dagger} f_{i\sigma} + \text{H.c.}), \quad \mathcal{H}'_{\text{SDW}} = \mathcal{H} + \Delta_z' \sum_{i,\sigma} \sigma e^{i\boldsymbol{Q}\cdot\boldsymbol{r}_i} (c_{i\sigma}^{\dagger} f_{i\sigma} + \text{H.c.}) \quad (2)$$

respectively, where the Weiss fields for the spin-singlet pairing Δ_0' and the spin-triplet pairing in the z-component Δ_z' are the variational parameters. The variational parameters are optimized on the basis of the variational principle, i.e., $\partial\Omega/\partial\Delta_0' = 0$ for the CDW state and $\partial\Omega/\partial\Delta_z' = 0$ for the SDW state. The solutions with $\Delta_0' \neq 0$ and $\Delta_z' \neq 0$ correspond to the CDW and SDW states, respectively.

In our VCA calculation, we assume the two-dimensional square lattice and use a $L_c = 2\times2 = 4$ site (8 orbital) cluster as the reference system. We use the value $D/t = 3.2$, so that the noninteracting tight-binding band structure is a small band-overlapped semimetal. The band structure has an electron pocket at $\boldsymbol{k} = (0,0)$ and a hole pocket at $\boldsymbol{k} = (\pi,\pi)$ in the Brillouin zone. The modulation vector of the CDW and SDW states is therefore given by $\boldsymbol{Q} = (\pi,\pi)$. Due to the Hartree shift, a Mott insulator state is realized at $U' \gg (U + J)/2$ and a band insulator state is realized at $U' \ll (U + J)/2$ [15, 24]. Here, we assume $U' = (U + J)/2$ to avoid the Hartree shift.

3. Results of calculation

3.1. Stability of the excitonic density-wave states

Figure 1(a) shows the calculated ground-state energy (per site) $E_0 = \Omega + \mu$ as a function of U. We first note that the excitonic states are stabilized with increasing with U under the assumption
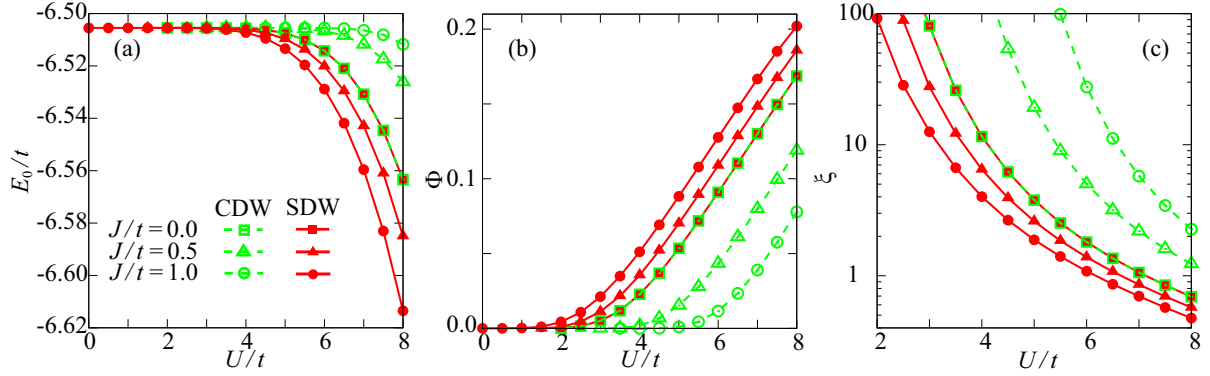
Figure 1. Calculated results for (a) the ground-state energy E_0, (b) order parameter Φ, and (c) pair coherence length (or the spatial size of the electron-hole pair) ξ as a function of U at $J/t = 0.0$ (square), 0.5 (triangle), and 1.0 (circle). Open and solid symbols indicate the CDW (spin-singlet) and SDW (spin-triplet) states, respectively.

$U' = (U + J)/2$. Then, we find that the ground-state energies of the CDW and SDW states are exactly degenerate at $J = 0$ and are decreased with increasing U (and U'). On introducing the Hund's rule coupling $J \neq 0$, the degeneracy is lifted, whereby the SDW state is stabilized and the CDW state is destabilized.

Figure 1(b) shows the calculated order parameters of the CDW and SDW states, which are defined as

$$\Phi_0 = \frac{1}{2N} \sum_{\boldsymbol{k}} \sum_{\sigma} \langle c^{\dagger}_{\boldsymbol{k}+\boldsymbol{Q}\sigma} f_{\boldsymbol{k}\sigma} \rangle, \quad \Phi_z = \frac{1}{2N} \sum_{\boldsymbol{k}} \sum_{\sigma} \sigma \langle c^{\dagger}_{\boldsymbol{k}+\boldsymbol{Q}\sigma} f_{\boldsymbol{k}\sigma} \rangle, \tag{3}$$

respectively. We find that the order parameters of the CDW and SDW states are exactly degenerate at $J = 0$ and are increased with increasing U (and U'). On introducing the Hund's rule coupling $J \neq 0$, the degeneracy is lifted, whereby we find that the order parameter of the SDW state becomes larger than that of the CDW state. Using the mean-field approximation, we find that the gap equations for the CDW and SDW states are given, respectively, by

$$1 = \frac{U - 3J}{2} \frac{1}{2N} \sum_{\boldsymbol{k}} \frac{1}{\sqrt{\varepsilon(\boldsymbol{k})^2 + |\Delta_0|^2}}, \quad 1 = \frac{U + J}{2} \frac{1}{2N} \sum_{\boldsymbol{k}} \frac{1}{\sqrt{\varepsilon(\boldsymbol{k})^2 + |\Delta_z|^2}}, \tag{4}$$

where $\varepsilon(\boldsymbol{k}) = -2t(\cos k_x + \cos k_y) - D$, $\Delta_0 = \Phi_0 (U - 3J)/2$ and $\Delta_z = \Phi_z (U + J)/2$. The effective attractive interactions for the spin-singlet and spin-triplet electron-hole pairs are therefore given by $(U - 3J)/2$ and $(U + J)/2$, respectively. Solving the gap equations in the weak-coupling region, we find that the order parameters increase exponentially with respect to the attractive interactions: i.e., $|\Delta_0| \propto \exp[-2/\rho(\varepsilon_{\mathrm{F}})(U - 3J)]$ and $|\Delta_z| \propto \exp[-2/\rho(\varepsilon_{\mathrm{F}})(U + J)]$, where $\rho(\varepsilon_{\mathrm{F}})$ is the density of state at the Fermi energy ε_{F}. We therefore find that the order parameters calculated by VCA increases exponentially with increasing the attractive interactions in the weak-coupling region, just as in the BCS mean-field theory.

Figure 1(c) shows the calculated pair coherence length ξ, which corresponds to the spatial size of the electron-hole pair. The pair coherence lengths of the spin-singlet and triplet exciton may be defined by [27]

$$\xi_0^2 = \frac{\sum_{\boldsymbol{k}} |\nabla_{\boldsymbol{k}} F_0(\boldsymbol{k})|^2}{\sum_{\boldsymbol{k}} |F_0(\boldsymbol{k})|^2}, \quad \xi_z^2 = \frac{\sum_{\boldsymbol{k}} |\nabla_{\boldsymbol{k}} F_z(\boldsymbol{k})|^2}{\sum_{\boldsymbol{k}} |F_z(\boldsymbol{k})|^2}, \tag{5}$$
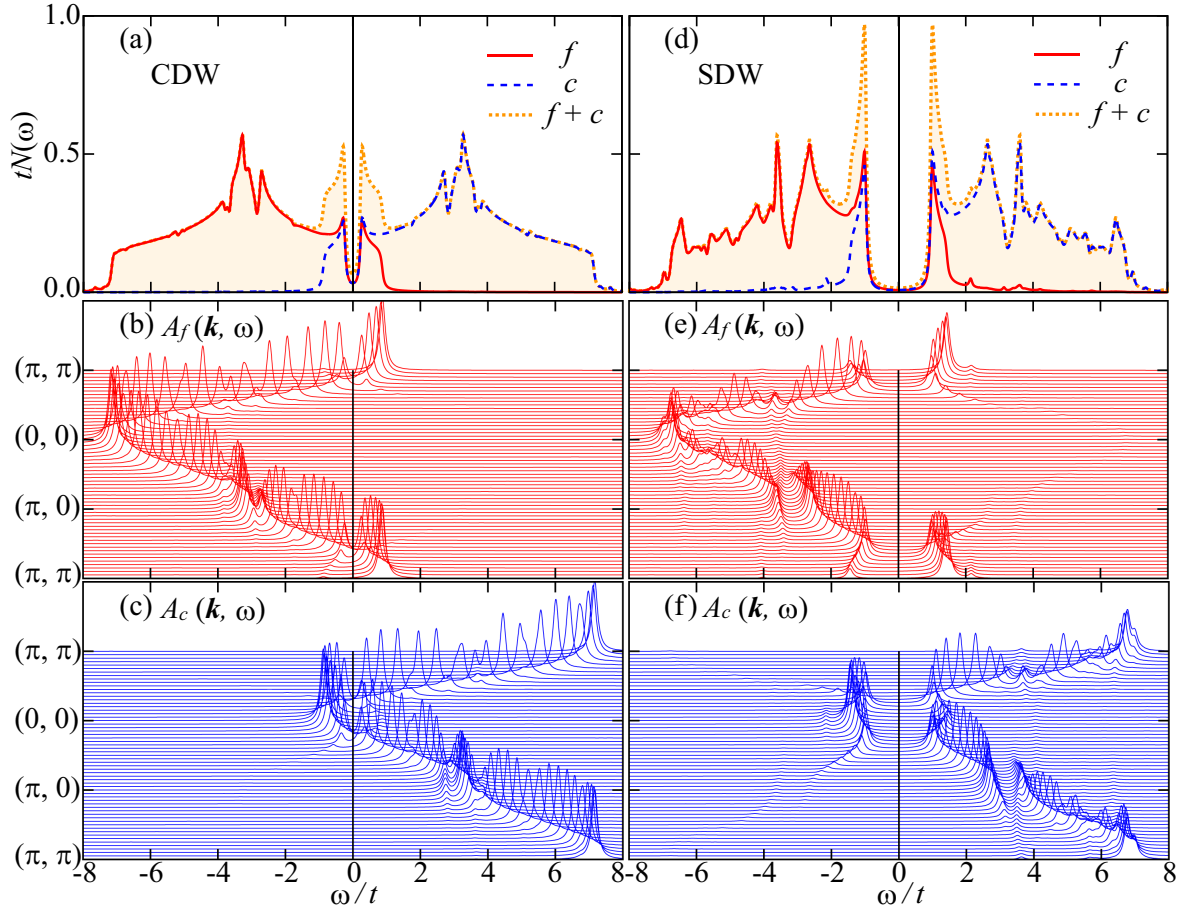
Figure 2. Calculated results for the DOS and single-particle spectrum of (a)-(c) the CDW state and (d)-(f) SDW state at $U/t = 8$ and $J/t = 1$. In (a) and (d), the solid, dashed, and dotted lines indicate the f-orbital, c-orbital, and total DOSs, respectively. Also shown are the single-particle spectra of the f-orbital [in (b) and (e)] and c-orbital [in (c) and (f)]. The Lorentzian broadening of $\eta/t = 0.05$ is used for the DOSs and $\eta/t = 0.1$ is used for the single-particle spectra.

respectively, where $F_0(\boldsymbol{k}) = \sum_\sigma \langle c^\dagger_{\boldsymbol{k}+\boldsymbol{Q}\sigma} f_{\boldsymbol{k}\sigma}\rangle/2$ and $F_z(\boldsymbol{k}) = \sum_\sigma \sigma \langle c^\dagger_{\boldsymbol{k}+\boldsymbol{Q}\sigma} f_{\boldsymbol{k}\sigma}\rangle/2$ are the condensation amplitudes for the spin-single and spin-triplet excitonic condensations, respectively. We find that the pair coherence lengths of the single and triplet excitons have the same values at $J = 0$, which are decreased with increasing U (and U'). The pair coherence length ξ is much larger than the lattice constant $a = 1$ in the weak-coupling region, which decreases smoothly to much smaller values than the lattice constant in the strong-coupling region, indicating that a smooth crossover occurs from the weakly paired BCS-like state ($\xi \gg 1$) to the BEC state of tightly bound pairs ($\xi \ll 1$). We find that, with increasing J, the spin-triplet excitons are paired more tightly and the spin-singlet excitons are paired more weakly, which are in accordance with the results for the ground-state energies and order parameters.

3.2. Single-particle spectrum

Next, we calculate the Green's function at the optimized values of the variational parameters using the cluster perturbation theory (CPT) [28]. Using the CPT Green's function $\mathcal{G}$, the single-

particle spectrum $A(\boldsymbol{k}, \omega)$ and DOS $N(\omega)$ of the $\alpha (= f, c)$ orbital are defined, respectively, as

$$A_\alpha(\boldsymbol{k}, \omega) = -\frac{1}{\pi} \sum_\sigma \mathrm{Im}\, \mathcal{G}_\sigma^{\alpha\alpha}(\boldsymbol{k}, \omega + i\eta), \quad N_\alpha(\omega) = \frac{1}{N} \sum_{\boldsymbol{k}} A_\alpha(\boldsymbol{k}, \omega), \tag{6}$$

where η gives the artificial Lorentzian broadening to the spectrum.

Figure 2 shows the calculated single-particle spectra and DOS of the metastable CDW state and stable SDW state at $J/t = 1$. Despite the fact that the noninteracting band structure is semimetalic, we find that the hybridisation gap opens between the valence band top at $\boldsymbol{k} = (\pi, \pi)$ and conduction band bottom at $\boldsymbol{k} = (0, 0)$, which is due to the excitonic condensation. We also find that, in agreement with the order parameters shown in Fig. 1(b), the introduction of the Hund's rule coupling suppresses the single-particle gap of the CDW state and enhances the single-particle gap of the SDW state. We note that the sharp coherence peak appears at the edges of the gap; the coherence peak of the SDW state is sharper than that of the CDW state, indicating that the spontaneous c-f hybridization in the SDW (CDW) state is enhanced (suppressed) by the Hund's rule coupling.

4. Summary
We have studied the stability of the excitonic density-wave states in the two-orbital Hubbard model. We have used the VCA to calculate the ground-state energy and order parameters and show that the Hund's rule coupling always stabilizes the excitonic SDW state and destabilizes the excitonic CDW state. The pair coherence lengths in the spin-singlet and spin-triplet states have also been calculated to show that only the spin-triplet excitons are paired more tightly with increasing J. We have also calculated the single-particle spectrum and density of states to see the characters of the excitonic density-wave states in detail.

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