Variational cluster approach to the extended Falicov-Kimball model: A BCS-BEC crossover in the Excitonic insulators

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Abstract. We study the Coulomb-interaction induced spontaneous symmetry breaking of the excitonic insulator state in the two-dimensional extended Falicov-Kimball model. Using the variational cluster approximation, we evaluate the order parameter, single-particle excitation gap, momentum distribution functions, and anomalous Green's function as a function of $U$ at zero temperature. We find that in the weak-to-intermediate coupling regime, the Fermi surface is well-defined and the calculated results can be understood in the close correspondence with the BCS theory, whereas in the strong-coupling regime, the Fermi surface is ill-defined and the results are consistent with the picture of BEC.

1. Introduction
Realization of excitonic insulators (EI) in the proximity of the semimetal-semiconductor transition was suggested about half a century ago [1, 2]. Because of the weak screening of the Coulomb attraction between electrons and holes due to a small number of carriers, the electrons and holes may spontaneously form the bound states (excitons), giving rise to the EI state. Recently, TaNi$_2$Se$_5$ has been studied by the angle-resolved photoemission spectroscopy measurements and it was reported that the valence-band top is extremely flat, and the material can be a new candidate for EI of the bound pairs between Ni 3$d$-Se 4$p$ holes and Ta 5$d$ electrons [3]. From the theoretical point of view, the extended Falicov-Kimball model (EFKM) has been studied extensively as a simple lattice model to describe EI by means of various numerical and theoretical approaches. The ground-state phase diagram of EFKM, on one hand, was determined by the constrained path Monte Carlo (CPMC) [4, 5]. On the other hand, the excitation properties of the EFKM are still of great interest. Projector-based renormalization method (PRM) calculation on the one-dimensional EFKM [6] reported incoherent parts of the single-particle excitation spectra, which are related to dissociation of the excitons. Detailed studies on the dynamical excitonic susceptibility at finite temperature calculated by use of the PRM [7] and slave boson technique [8] confirmed that tightly bound excitons exist even above the critical temperature $T_c$ of the exciton condensation. The results strongly support the existence of so-called excitonic halo above $T_c$ suggested by Bronold and Fehske [9], where tightly bound excitons exist without condensation, leading to a scenario of the Bose-Einstein condensation (BEC) of preformed excitons in the strong Coulomb interaction regime.
In this paper, we study the BCS-BEC crossover [10] of the EI state of the EFKM defined on the two-dimensional (2D) square lattice as a function of the Coulomb interaction strength $U$. We employ the variational cluster approximation (VCA) [11] based on the self-energy functional theory [12] at zero temperature. The cluster perturbation theory (CPT) [13] is used to calculate the single-particle and anomalous Green’s functions. Details will be presented elsewhere [14].

2. Model and Method

The Hamiltonian of EFKM reads

$$\mathcal{H} = - \sum_{\alpha = c, f} \left( t_\alpha \sum_{\langle ij \rangle} (\alpha_i \alpha_j^\dagger + \text{H.c.}) + (\epsilon_\alpha - \mu) \sum_i n_{i\alpha} \right) + U \sum_i n_{ic} n_{if}$$

(1)

where $\alpha_i$ denotes the annihilation operator of an electron on the $\alpha$-orbital at site $i$ and $n_{i\alpha} = \alpha_i^\dagger \alpha_i$. $t_\alpha$ is the hopping integral between the neighboring sites of the 2D square lattice and $\epsilon_\alpha$ is the on-site energy level of the $\alpha$-orbitals. $U$ is the inter-orbital Coulomb repulsion between electrons. The chemical potential $\mu$ is determined so as to maintain the average particle density $n$ at half filling $n = 1$. Throughout the paper, we set $h = k_B = 1$. We use $t_\alpha = 1$ as the unit of energy and we focus on the band parameters with values $\epsilon_c = 0, \epsilon_f = -1,$ and $t_f = -0.3$.

We employ VCA in order to study the EI state of EFKM at zero temperature, which enables us to take into account the effects of electron correlations. VCA is based on the variational principle for the grand potential as a functional of the self-energy [12, 15]. The trial self-energy for the variational method is generated from the exact self-energy of the disconnected finite-size clusters, so-called reference system. The Hamiltonian of the reference system is defined as $\mathcal{H}_{\text{ref}} = \mathcal{H} + \mathcal{H}_{\text{pair}} + \mathcal{H}_{\text{local}}$, $\mathcal{H}_{\text{pair}} = \Delta^\prime \sum_i (c_i^\dagger f_i + \text{H.c.})$, $\mathcal{H}_{\text{local}} = \epsilon' \sum_i (n_{ic} + n_{if})$, where the Weiss field for the on-site electron-hole pairing $\Delta^\prime$ and the orbital-independent potential $\epsilon'$ are the variational parameters optimized based on the variational principle. $\epsilon'$ is introduced in order to calculate the average particle density $n$ correctly [16]. The single-particle Green’s function for $\alpha$-electron $G^{\alpha\alpha}(\mathbf{k}, \omega)$ and anomalous Green’s function $G^{cf}(\mathbf{k}, \omega)$ are calculated by CPT [13]. An 8-site (16-orbital) cluster is used as a reference system, thus the effects of static and dynamical electron correlation within the cluster size are taken into account. Details of VCA can be found in Ref. [17].

3. Results of calculation

We first calculate the $U$ dependence of the order parameter for the exciton condensation $2\Delta = U \sum_i (c_i^\dagger f_i + \text{H.c.})$ and single-particle excitation gap $\Delta_{\text{gap}} = \mu^+ - \mu^-$, where $L$ is the number of the lattice sites, $\langle \cdots \rangle$ denotes the ground-state expectation value, and $\mu^+(-)$ is the upper (lower) bound of the chemical potential. The factor 2 for the order parameter is introduced in order to compare with the single-particle gap, by analogy with the BCS mean-field theory. Calculated results are shown in Fig. 1.

First, we can see from the results that there is the lower (upper) bound of the Coulomb interaction strength $U_{c1}$ ($U_{c2}$) for the EI state. The obtained values are $(U_{c1}, U_{c2}) = (0.65, 6.6)$. The existence of the upper bound $U_{c2}$ seems to contradict to the case of the s-wave superconducting state in the attractive Hubbard model, which has no $U_{c2}$ [18]. What happened at $U = U_{c2}$ is that the Hartree potential makes the $f$-band fully occupied and $c$-band empty, so that there are no Coulomb interactions between $c$-electrons and $f$-holes and thus the system becomes simply a band insulator above $U_{c2}$.

Then, in the weak-to-intermediate coupling regime ($U \lesssim 5$), both the order parameter and single-particle gap increase with increasing $U$ with the relation $2|\Delta| \approx \Delta_{\text{gap}}$. This result is consistent with the relation $2|\Delta| = \Delta_{\text{gap}}$ of the BCS mean-field theory. In the strong-coupling
Figure 1. $U$ dependence of the order parameter $2|\Delta|$ (filled symbol) and the single-particle gap $\Delta_{\text{gap}}$ (open symbol). The dashed straight line indicates the single-particle gap in the normal state.

Figure 2. The momentum distribution functions $n_c(k)$ and $2F(k)$ along $k = (0,0) \rightarrow (\pi, \pi)$ for various $U$. The $f$-electron momentum distribution functions $n_f(k) = 1 - n_c(k)$ are not shown.

regime ($U \geq 5$), the order parameter rapidly decreases with increasing $U$ but the single-particle gap remains large. If we can assume that the energy scale of the single-particle gap $\Delta_{\text{gap}}$ and order parameter $2|\Delta|$ may correspond to that of the characteristic temperature for the exciton formation ($T_{\text{ex}}$) and critical temperature for the condensation of excitons ($T_c$), respectively, then the two temperatures should be comparable ($T_{\text{ex}} \simeq T_c$) in the weak-coupling (BCS) regime but may be well separated ($T_{\text{ex}} \gg T_c$) in the strong-coupling (BEC) regime. The BCS-BEC crossover may then be expected in this model although our calculations are done at zero temperature.

We next calculate the $c$-electron and anomalous momentum distribution functions defined as $n_c(k) = -\frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \Im G^{cc}(k,\omega+i\eta^+)$, $F(k) = -\frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \Im G^{cf}(k,\omega+i\eta^+)$, respectively. The results are shown in Fig. 2. We define, by analogy with the BCS mean-field theory, the “Fermi momentum” $k_F$ as $n_c(k_F) = n_f(k_F) = 0.5$. We can see that, in the weak-coupling regime, $n_c(k)$ sharply drops across the $k_F$ and $F(k)$ is peaked at $k_F$. With increasing $U$, $k_F$ approaches to $(0,0)$ because the Hartree potential for $c$-electrons reduces the $c$-electron density. Accordingly, $F(k)$ becomes broad in the momentum space, indicating that the radius of electron-hole pairs becomes small in real space. When $U$ reaches the crossover regime ($U \sim 5$), we have no $k_F$ and $|F(k)|$ decreases in all momentum with increasing $U$. This behavior is consistent with the rapid decrease of $|\Delta|$ with increasing $U$ in the strong-coupling regime (see Fig. 1).

We also calculate the anomalous spectral function $F(k,\omega) = -\frac{1}{\pi} \Im G^{cf}(k,\omega+i\eta)$ with $\eta = 0.1$ at $U = 2, 5,$ and $6.5$. The results are shown in Fig. 3. At $U = 2$, $F(k,\omega)$ shows sharp peak near $k_F$ and its intensity rapidly decreases as the momentum goes away from $k_F$ and frequency goes away from the Fermi energy $\mu$. At $U = 5$, the incoherent continua appear in the spectral function. We can see that both the single particle gap and intensity of $F(k,\omega)$ are large. At $U = 6.5$, $F(k,\omega)$ shows a semiconductor-like dispersion mainly due to the Hartree potential. The frequency dependence of the intensity of $F(k,\omega)$ is weaker than that at $U = 2$ and $5$.

4. Summary

We have analyzed the EI state of EFKM using VCA as a function of Coulomb interaction strength $U$. In the weak-to-intermediate coupling regime ($U \lesssim 5$), the order parameter $\Delta$ and single-particle gap $\Delta_{\text{gap}}$ behave like the BCS theory, i.e., $\Delta_{\text{gap}} \simeq 2|\Delta|$, the Fermi momentum $k_F$ can be defined from the momentum distribution function, and the anomalous Green’s function...
Figure 3. Calculated anomalous spectral function $F(k, \omega)$ at (a) $U = 2$, (b) $U = 5$, and (c) $U = 6.5$.

is peaked near Fermi energy $\mu$ at $k_F$. In the crossover regime ($U \sim 5$), both $\Delta$ and $\Delta_{\text{gap}}$ take large values and they are close to the maximum. Correspondingly, $k_F$ becomes ill-defined. In the strong-coupling regime ($U \geq 5$), $\Delta$ rapidly decreases with increasing $U$, while $\Delta_{\text{gap}}$ remains open, $k_F$ is ill-defined, and the frequency and momentum dependence of the intensity of the anomalous Green’s function becomes weak. Thus, the Fermi surface plays no role in the strong coupling regime, which is consistent with the picture of BEC.

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