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

K Seki¹, T Kaneko¹, S Yamaki¹, R Eder² and Y Ohta¹
¹Department of Physics, Chiba University, Chiba 263-8522, Japan
²Karlsruhe Institute of Technology, Institut für Festkörperphysik, D-76021 Karlsruhe, Germany
E-mail: seki-kazuhiro@graduate.chiba-u.jp

Abstract. Motivated by recent experimental and theoretical studies of excitonic insulators, we use the variational cluster approximation to calculate the single-particle Green’s function and anomalous Green’s function of the Falicov-Kimball model extended by including a finite valence bandwidth. We thus determine the single-particle excitation gap due to the bound-state formation of an electron and a hole. We also evaluate the order parameter of the excitonic insulating state indicating quantum coherence of condensed excitons. We thereby discuss the excitonic insulator that typifies either a BCS condensate of electron-hole pairs (weak-coupling regime) or a Bose-Einstein condensate of preformed excitons (strong-coupling regime). A BCS-BEC crossover of the model thus manifests itself as a function of the Coulombic coupling strength.

1. Introduction
Realization of excitonic insulators (EI) on the proximity of the semimetal-semiconductor transition was suggested more than half a century ago [1, 2]. Because of the weak screening of the Coulomb attraction between the electrons and holes due to a small number of carriers, the electrons and holes may spontaneously form bound states (excitons). As a candidate for EI, 1T-TiSe₂ has been studied both theoretically and experimentally [3, 4]. Ta₂NiSe₅ has also 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 [5]. From the theoretical point of view, the Falicov-Kimball model [6] extended by including a finite valence bandwidth (EFKM) has been extensively studied as a simple lattice model to describe EI; e.g., mean-field theory [7, 8], constrained path Monte Carlo [9], slave-boson [10], and projector-based renormalization [11, 12] methods have been used. The realization of the EI state in some parameter region of this model is thus promising.

In this paper, we study the EI state of the EFKM defined on the two-dimensional square lattice by employing the variational cluster approximation (VCA) [13] based on the self-energy functional theory [14] at zero temperature. We make calculations of the EI order parameter, the single-particle gap, the single-particle Green’s function and the anomalous Green’s function as a function of the Coulomb interaction strength. Thereby, we discuss the BCS-BEC crossover [15] of this model.
2. Model and Method

The Hamiltonian of the EFKM reads

$$\mathcal{H}_{\text{EFKM}} = \sum_{\alpha = c, f} \left( -t_{\alpha} \sum_{\langle ij \rangle} (\alpha_{i}^{\dagger} \alpha_{j} + \text{H.c.}) + (\epsilon_{\alpha} - \mu) \sum_{i} n_{i}^{\alpha} \right) + U \sum_{i} n_{i}^{c} n_{i}^{f} \quad (1)$$

where $\alpha_{i}$ ($\alpha_{i}^{\dagger}$) denotes the annihilation (creation) operator for a fermion 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 two-dimensional square lattice and $\epsilon_{\alpha}$ is the energy level of the $\alpha$-orbitals. $U$ is the inter-orbital Coulomb repulsion. The chemical potential $\mu$ is determined to keep the half-filling condition $\langle n \rangle = 1$, where $\langle n \rangle$ is the average particle density. We set the unit of energy as $t_c = 1$ and we focus on the band parameter values $\epsilon_c = 0, \epsilon_f = -1$, and $t_f = -0.3$.

To analyze the EI state of EFKM, we apply VCA at zero temperature. VCA is based on the variational principle for the grand potential as a functional of the self-energy [14, 16] and can describe the spontaneous symmetry breaking. The trial self-energy for the variation method can be generated from the exact self-energy of a small cluster, so-called reference system. The reference system $\mathcal{H}_{\text{ref}} = \mathcal{H}_{\text{EFKM}} + \mathcal{H}_{\text{pair}} + \mathcal{H}_{\text{local}}$ defined on the small cluster, where $\mathcal{H}_{\text{pair}} = \Delta' \sum_{i} (c_{i}^{\dagger} f_{i} + \text{H.c.})$ and $\mathcal{H}_{\text{local}} = \epsilon' \sum_{i} n_{i}^{c} n_{i}^{f}$, is solved and the trial self-energy (or equivalently the trial single-particle Green’s function) is calculated by the Lanczos exact-diagonalization method. $\Delta'$ and $\epsilon'$ are the variational parameters, which are optimized based on the variational principle. Note that the solution with $\Delta' \neq 0$ corresponds to the spontaneous EI state. The orbital-independent potential $\epsilon'$ is introduced in order to calculate the average particle density $\langle n \rangle$ correctly [17]. The single-particle Green’s function $\mathcal{G}(\mathbf{k}, \omega)$ and anomalous Green’s function $\mathcal{F}(\mathbf{k}, \omega)$ are calculated by use of the cluster perturbation theory (CPT) [18] with the optimized variational parameters. An 8-site (16-orbital) cluster is used as a reference system, thus the effects of electron correlation within the cluster size are taken into account. Details of VCA can be found in Ref. [19].

3. Results of calculation

Figure 1 shows the $U$ dependence of the single-particle excitation gap $\Delta_{\text{gap}}$ and the exciton condensation order parameter $2\Delta = U \sum_{i} (c_{i}^{\dagger} f_{i} + f_{i}^{\dagger} c_{i})/N$, where $N$ is the number of lattice sites and $\langle \cdots \rangle$ denotes the expectation value. In weak-coupling regime ($U \lesssim 5.5$), both the order parameter and the single-particle gap increase with increasing $U$. In strong-coupling regime ($U \gtrsim 5.5$), the order parameter rapidly decreases with increasing $U$ and finally vanishes at $U = 6.6$, but the single-particle gap remains large. The vanishing of the order parameter in the
Figure 2. Calculated imaginary part of the single-particle Green’s function $A(k, \omega)$ (top) and the anomalous Green’s function $F(k, \omega)$ (bottom) at $U = 2$ (left panel) and $U = 6.5$ (right panel). The artificial Lorentzian broadening $\eta = 0.1$ is used.

The imaginary part of the single-particle Green’s function $A(k, \omega) = \frac{1}{G(k, \omega)}$ and the anomalous Green’s function $F(k, \omega) = \frac{1}{F(k, \omega)}$ at $U = 2$ (BCS regime) and $U = 6.5$ (BEC regime) are shown in Fig. 2. At $U = 2$, $A(k, \omega)$ shows a small gap near the “Fermi momentum” $k_F$ where the single-particle gap is the smallest. $F(k, \omega)$ shows sharp peak near $k_F$ and its intensity rapidly decreases as the momentum goes away from $k_F$. At $U = 6.5$, $A(k, \omega)$ shows a semiconductor-like dispersion mainly due to the Hartree potential. The momentum dependence of the intensity of $F(k, \omega)$ is weaker than that at $U = 2$.

The $c$-electron, $f$-electron and anomalous momentum distribution functions, i.e., $n^c(k)$, $n^f(k)$ and $d(k) = -\frac{1}{\pi} \text{Im} \int^\mu_{-\infty} d\omega F(k, \omega)$, respectively, are shown in Fig. 3. At $U = 2$, $d(k)$ shows the sharp peaks near $k_F$, indicating the existence of weakly bound electron-hole pairs. At $U = 6.5$, $k$-dependence of the peak intensity of the anomalous Green’s function is weak and $d(k)$ is spread out in momentum space. Thus, in real space, small electron-hole pairs exist.

4. Summary
The EI state of the EFKM has been analyzed by VCA at zero temperature. In the weak coupling regime, we found that the magnitude of the single particle gap $\Delta_{\text{gap}}$ is almost comparable to

large $U$ regime seems to contradict the case of the pairing in the attractive Hubbard model. The reason for this is that, in $U \geq 6.6$, the Hartree potential makes the $c$-band empty and $f$-band fully occupied, so that there is no Coulomb interaction between $c$- and $f$- electrons. Thus the system is simply a band insulator and the order parameter vanishes in $U \geq 6.6$.

If we can assume that the energy scale of the single-particle gap and the order parameter $2\Delta$ may correspond to that of the characteristic temperature for the exciton formation ($T_{\text{ex}}$) and the critical temperature for the condensation of excitons ($T_c$), respectively, then the two temperatures should be comparable ($T_{\text{ex}} \approx T_c$) in the weak-coupling (BCS) regime but should 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.
that of the order parameter $2\Delta$ and that the anomalous momentum distribution function $d(k)$ shows the sharp peaks near $k_F$. This indicates that the system is in the BCS-like weakly-bound exciton condensation state. In the strong coupling regime, the single-particle gap $\Delta_{\text{gap}}$ remains large with increasing $U$ but the order parameter decreases rapidly and $d(k)$ is widely spread in momentum space, indicating that the system is in the BEC-like condensation state of strongly bound electron-hole pairs. Further details will be reported elsewhere [20].

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