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Excitonic BCS-BEC crossover in double-layer systems

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We investigate electron-hole pair condensation in electron bilayers described by the square-lattice extended Falicov-Kimball model. Using exact diagonalization and variational cluster approximation techniques we first calculate the anomalous Green's function to clarify the character of the excitons in momentum space. We then evaluate the coherence length ξ (in unit of the lattice constant a) from the corresponding condensation amplitude and demonstrate the smooth crossover between a BCS state of weakly paired electrons and holes ($\xi/a \gg 1$) and a BEC state of tightly bound excitons ($\xi/a \ll 1$) as the Coulomb attraction increases. Overcoming the finite-size effects of exact diagonalization while still taking into account the essential correlation effects we show that the variational cluster approximation provides an advantageous description of exciton condensation in strongly correlated electron systems.

KEYWORDS: exciton condensation, BCS-BEC crossover, Falicov-Kimball model, exact diagonalization, variational cluster approximation

1. Introduction

The formation of excitonic quantum condensates are intensively studied during the last half century [1–4]. Experimentally, multifaceted attempts have been made to observe the condensed state of excitons, e.g., in photoexcited semiconductors [5–9], unconventional semiconductor/graphene systems [10–14], electrostatic traps [15], or neutral electron-ion quantum plasmas [16]. Theoretically, a possible crossover between a Bardeen-Cooper-Schrieffer (BCS) electron-hole pair condensate and a Bose-Einstein condensate (BEC) of preformed excitons has been of topical interest [4, 17–23].

In order to get unbiased results for the problem of exciton condensation in electron-hole double-layer systems, in previous work [24], we investigated a minimal lattice fermion model, the so-called extended Falicov-Kimball model (EFKM) [25–29], by exact diagonalization (ED) of small clusters. To pinpoint the finite-size effects and affirm the main conclusions of [24] in the thermodynamic limit, in the present work, we employ the variational cluster approximation (VCA), based on the self-energy functional theory [30–33], to the square-lattice double-layer EFKM. Thereby we will corroborate the excitonic BCS-BEC crossover scenario suggested previously for strongly correlated electron-hole systems [34–36]. Calculating the anomalous excitation spectra and the condensation amplitude, we are able to extract the coherence length and order parameter of the condensate in both limits. Methodically, we will compare the VCA results with the corresponding ED and mean-field (MF) data to point out the range of applicability of the different approaches.

2. Model and Method

2.1 Extended Falicov-Kimball model

The EFKM for an electron-hole double layer is defined by the Hamiltonian

$$\mathcal{H} = -\sum_{\alpha=c,f} t_\alpha \sum_{\langle i,j\rangle} (\alpha_i^\dagger \alpha_j + \mathrm{H.c.}) + U \sum_i n_i^f n_i^c - \sum_{\alpha=c,f} \mu_\alpha \sum_i n_i^\alpha \,, \tag{1}$$

where $\alpha_i^\dagger$ (α_i) creates (annihilates) an electron in the α $(= c, f)$ orbital at site i, and $n_i^\alpha = \alpha_i^\dagger \alpha_i$. The transfer amplitude of electrons between the α orbitals on nearest-neighbor sites is denoted by t_α. We assume a band structure with a direct band gap ($t_c t_f < 0$). Without loss of generality the f orbitals were assigned to the hole (or valence-band) layer and the c orbitals to the electron (or conduction-band) layer. U (> 0) parametrizes the on-site interlayer Coulomb repulsion between f and c electrons that allows for an on-site interlayer Coulomb attraction between f hole and c electrons. Note that in our double-layer system the numbers of f and c particles are separately conserved because charge transfer between the two layers is assumed to be impossible. This mimics the generic situation in semiconductor electron-hole double quantum wells [12, 37, 38], and double-monolayer [39, 40] or double-bilayer graphene systems [41]. We furthermore assume that the excited electrons and holes have infinite lifetime and that the number of excited electrons is equal to the number of excited holes. This is in accord with the experimental situation in the majority of cases [5–13, 23, 40]. In practice, we adjust the chemical potentials μ_f and μ_c to maintain the number of electrons in the f and c layers independently. Throughout this work, we assume that the effective mass of the f band is equal to that of the c band (or $|t_f| = t_c = t$). Then we have the chemical potentials $\mu_f = -\mu_c = \mu$. Let us stress that the exciton condensation state in the double-layer EFKM can be mapped onto the superconducting (superfluid) state in the attractive Hubbard model in the mass-balanced case [42].

2.2 Exact diagonalization and variational cluster approximation

Within the ED investigation of the double-layer EFKM we use finite-size square lattices of $L_\mathrm{c} = 4{\times}4 = 16$ sites (32 orbitals) with periodic boundary conditions (PBC). The particle densities are fixed to be $n_f = 0.75$ and $n_c = 0.25$, i.e., $(N_f, N_c) = (12, 4)$, which realizes a quarter-filled electron and hole band: $n^e = n^h = 0.25$. For the 4×4 lattice considered, the Fermi momenta are $\boldsymbol{k}_\mathrm{F} = (\pm\pi/2, 0)$ and $\boldsymbol{k}_\mathrm{F} = (0, \pm\pi/2)$.

To accomplish the thermodynamic limit, we employ the VCA based on the variational principle for the grand potential as a functional of the self-energy [30–33]. The trial self-energy for the variational method is generated from the exact self-energy of the disconnected finite clusters which act as a reference system. The Hamiltonian of the reference system is defined as $\mathcal{H}' = \mathcal{H} + \mathcal{H}_\mathrm{pair} + \mathcal{H}_\mathrm{local}$ with

$$\mathcal{H}_\mathrm{pair} = \Delta' \sum_i (c_i^\dagger f_i + \mathrm{H.c.}) \quad \text{and} \quad \mathcal{H}_\mathrm{local} = \varepsilon'_f \sum_i n_i^f + \varepsilon'_c \sum_i n_i^c \,, \tag{2}$$

where the Weiss field for the s-wave pairing Δ' and the on-site potentials ε'_α $(\alpha = f, c)$ are variational parameters. Note that the ε'_α, fulfilling for the mass-balanced case $\varepsilon'_f = -\varepsilon'_c = \varepsilon'$, were introduced to determine the particle density correctly. Provided $(\partial\Omega/\partial\Delta', \partial\Omega/\partial\varepsilon'_\alpha) = (0, 0)$ is guaranteed, the particle density n_α follows from $n_\alpha = -\partial\Omega/\partial\mu_\alpha$, and the chemical potential μ_α is determined to maintain $n_f = 0.75$ and $n_c = 0.25$. The Fermi momentum $\boldsymbol{k}_\mathrm{F}$ is defined via $\varepsilon_{\boldsymbol{k}_\mathrm{F}} = \mu$ (at $U = 0$), where $\varepsilon_{\boldsymbol{k}} = -2t(\cos k_x + \cos k_y)$. Note that in the space of the cluster reference system all electronic correlations were exactly taken into account. We use $L_\mathrm{c} = 2 \times 2 = 4$ (8 orbitals) in what follows.

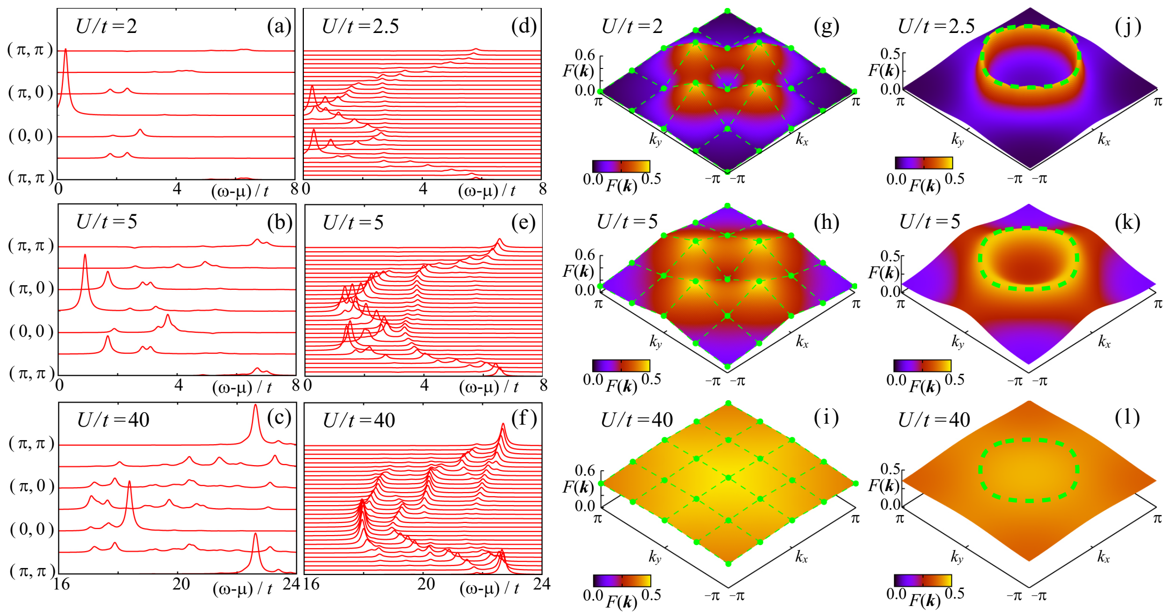

Fig. 1. (Color online) Left panel: Anomalous spectral function $F(\boldsymbol{k},\omega)$ as obtained by ED [(a)-(c)] and VCA [(d)-(f)] with a Lorentzian broadening of $\eta/t = 0.1$. Right panel: Exciton condensation amplitude $F(\boldsymbol{k})$ calculated by ED [(g)-(i)] and VCA [(j)-(l)]. In panels (j)-(l) the green dashed lines indicate the Fermi surface.

3. Numerical results

3.1 Anomalous Green's function

Let us first discuss the anomalous Green's function. Using ED, the anomalous Green's function is obtained from

$$G^{cf}_{\rm ED}(\boldsymbol{k},\omega) = \langle N_f - 1, N_c + 1 | c^{\dagger}_{\boldsymbol{k}} \frac{1}{\omega + i\eta - \mathcal{H} + E_0} f_{\boldsymbol{k}} | N_f, N_c \rangle \,, \tag{3}$$

where $|N_f, N_c\rangle$ is the ground state of the EFKM with fixed numbers of c and f electrons. In Eq. (3), E_0 is the average energy of the states $|N_f, N_c\rangle$ and $|N_f - 1, N_c + 1\rangle$ [24, 43, 44]. Within VCA, the anomalous Green's function is calculated by cluster perturbation theory (CPT) [45], $G^{cf}_{\rm CPT}(\boldsymbol{k},\omega)$, making use of optimized variational parameters. From $G^{cf}(\boldsymbol{k},\omega)$, we can immediately deduce the anomalous spectral function: $F(\boldsymbol{k},\omega) = -\frac{1}{\pi}{\rm Im}G^{cf}(\boldsymbol{k},\omega)$.

Figure 1 (a)-(f) gives an intensity plot of $F(\boldsymbol{k},\omega)$ in the square-lattice Brillouin zone. First of all we note that the VCA spectra basically agree with the ED spectra at the wave vectors allowed for a 4×4 cluster with PBC. In the weak-coupling regime [see Fig. 1 (a) and (d)], $F(\boldsymbol{k},\omega)$ has a sharp peak at the Fermi momentum $\boldsymbol{k}_{\rm F}$ whose intensity rapidly decreases as soon as the momentum deviates from $\boldsymbol{k}_{\rm F}$. With increasing U/t, the lowermost peak of $F(\boldsymbol{k},\omega)$ shifts to higher energies, indicating an enhancement of the exciton's binding energy $|E_B|$, which may also be evaluated by the ground-stete energies [24]. For $U/t = 5$ [see Fig. 1 (b) and (e)], $F(\boldsymbol{k},\omega)$ still exhibits a pronounced peak around $\boldsymbol{k}_{\rm F}$, but to compare to the $U/t = 2.5$ spectrum, $F(\boldsymbol{k},\omega)$ acquires substantial weight at momenta away from $\boldsymbol{k}_{\rm F}$. In the strong-coupling limit [cf. Fig. 1 (c) and (f) for $U/t = 40$], the spectral weight of $F(\boldsymbol{k},\omega)$ is redistributed to higher energies and spread over the entire Brillouin zone.

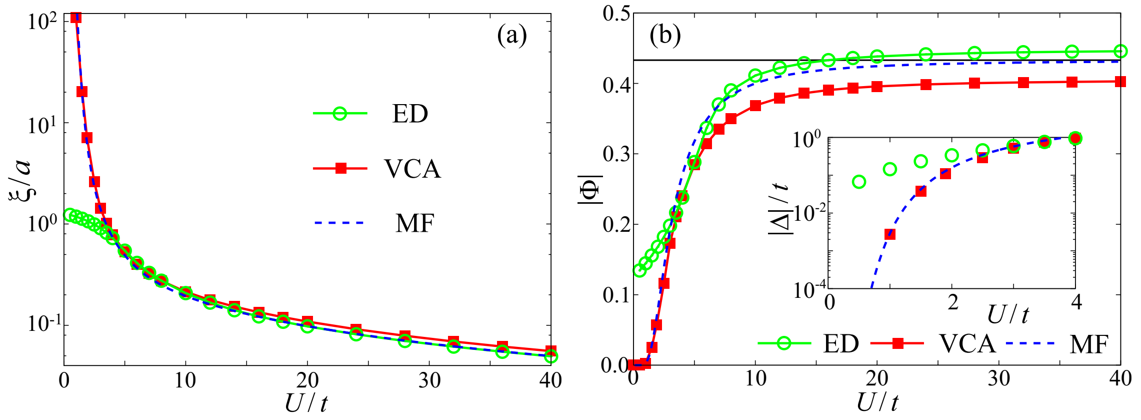

Fig. 2. (Color online) (a) Coherence length ξ in unit of the lattice constant a and (b) anomalous expectation value Φ for the double-layer EFKM, as obtained by ED (open green circles), VCA (full red squares), and MF (dashed blue lines). In (b), the horizontal line indicates $|\Phi| = \sqrt{3}/4$ and the inset gives the order parameter $\Delta = U\Phi$ in the weak-coupling regime.

3.2 Exciton condensation amplitude

To elucidate the nature of excitons in momentum space we now consider the condensation amplitude $F(\boldsymbol{k})$. Within ED the condensation amplitude can be directly calculated from

$$F(\boldsymbol{k}) = \langle N_f - 1, N_c + 1 | c^\dagger_{\boldsymbol{k}} f_{\boldsymbol{k}} | N_f, N_c \rangle \,. \tag{4}$$

In VCA the amplitude emanates from the anomalous CPT Green's function

$$F(\boldsymbol{k}) = \oint_C \frac{\mathrm{d}z}{2\pi\mathrm{i}} G^{cf}_{\mathrm{CPT}}(\boldsymbol{k}, z) \,, \tag{5}$$

where the contour C encloses the negative real axis in frequency space.

The results for $F(\boldsymbol{k})$ are shown in Fig. 1 (g)-(l). Again ED and VCA data agree insofar as comparable. The finite-size limitations of ED become obvious however. At weak couplings [cf. Fig. 1 (g) and (j)], the importance of Fermi surface effects are reflected in the corral-like shape of $\boldsymbol{k}_{\mathrm{F}}$ with $|F(\boldsymbol{k}_{\mathrm{F}})| \simeq 0.5$. The sharply peaked $F(\boldsymbol{k})$ in momentum space indicates that the radius of the exciton is large in real space, i.e., we observe a weakly bound electron-hole pair. Increasing U/t, $F(\boldsymbol{k})$ broadens in momentum space, indicating a spatially more confined exciton. In the strong-coupling regime displayed by Fig. 1 (i) and (l)), $F(\boldsymbol{k})$ is (almost) homogeneously distributed over the whole Brillouin zone. Hence the excitons are tightly bound and small in real space.

3.3 Coherence length

The coherence length ξ gives valuable information as to the nature of the exciton condensate. Coming from $F(\boldsymbol{k})$, this quantity may be defined as

$$\xi^2 = \frac{\sum_{\boldsymbol{r}} \boldsymbol{r}^2 |F(\boldsymbol{r})|^2}{\sum_{\boldsymbol{r}} |F(\boldsymbol{r})|^2} = \frac{\sum_{\boldsymbol{k}} |\boldsymbol{\nabla}_{\boldsymbol{k}} F(\boldsymbol{k})|^2}{\sum_{\boldsymbol{k}} |F(\boldsymbol{k})|^2}, \tag{6}$$

where $F(\boldsymbol{r}) = \frac{1}{\sqrt{L}} \sum_{\boldsymbol{r}'} \langle c^\dagger_{\boldsymbol{r}'+\boldsymbol{r}} f_{\boldsymbol{r}'} \rangle$ is the condensation amplitude in real space for the electron-hole pairs with distance $\boldsymbol{r}$ [24, 35, 42, 44]. The results obtained by ED, VCA, and MF are shown in Fig. 2 (a) in dependence on U/t. We find that ξ calculated by ED stays finite as $U/t \to 0$. This is clearly a finite-size effect caused by the small number of available momenta in the Brillouin zone. In the intermediate-to-strong coupling regime, the coherence lengths calculated by ED, VCA, and MF are even in quantitative agreement. They rapidly decrease as U/t increases. At small U/t, the coherence length is much larger than the lattice constant a, in consequence of the weakly bound electron-hole pairs [cf. the behavior of $F(\boldsymbol{k})$ shown in Fig. 1 (g)-(l)]. This is the BCS limit. Increasing U/t, ξ decreases and firstly becomes comparable and finally much smaller than the lattice constant (for very strong couplings). As a result the excitons become manifest in a BEC. Altogether we observe a smooth crossover from a BCS state of weakly paired electrons and holes ($\xi/a \gg 1$) to a BEC state of tightly bound pairs ($\xi/a \ll 1$). This crossover behavior is consistent with the calculated spectral properties of the system shown in Fig. 1.

3.4 Order parameter

Finally let us discuss the order parameter for exciton condensation $\Delta = U\Phi$, which again can be obtained from the anomalous Green's function since

$$\Phi = \frac{1}{L}\sum_{\boldsymbol{k}} \langle c^{\dagger}_{\boldsymbol{k}} f_{\boldsymbol{k}} \rangle = \frac{1}{L}\sum_{\boldsymbol{k}} F(\boldsymbol{k}). \tag{7}$$

The results for Φ and Δ are shown in Fig. 2 (b). The MF theory predicts that the order parameter $\Delta_{\rm MF}$ increases exponentially with increasing U/t in the weak-coupling limit ($\log \Delta_{\rm MF} \propto -1/U$), and $\Phi_{\rm MF}$ saturates at $\sqrt{3}/4$ for $n^e = n^h = 0.25$ in the strong-coupling limit. Obviously ED fails in reproducing the correct weak-coupling behavior: $\Phi_{\rm ED}$ stays finite as $U/t \to 0$ and $\Delta_{\rm ED}$ does not show the exponential increase at $U/t \gtrsim 0$. Clearly this can be attributed to finite-size effects within our small cluster calculation. Remarkably the VCA yields the exponential increase expected in the weak-coupling limit [see inset of Fig. 2 (b)]. We furthermore note that $\Phi_{\rm VCA}$ is in qualitative accordance with the ED and MF results in the intermediate to strong coupling regime. For $U/t \gtrsim 5$, $\Phi_{\rm VCA}$ is reduced in comparison to the MF result. This may be due to the effects of quantum fluctuations of exciton condensation included in VCA but not in MF.

4. Conclusions

To summarize, we have investigated the formation and condensation of excitons in the mass-balanced double-layer extended Falicov-Kimball model using—besides ED (exact diagonalization)—VCA (variational cluster approximation) and MF (mean-field) based approaches. We have analyzed the nature of excitonic bound states in dependence on the strength of the Coulomb interaction between electrons and holes and showed—evaluating the anomalous Green's function—that the excitonic condensation amplitude, coherence length, and order parameter function signal a smooth BCS-BEC crossover in the condensed phase. Our comparative numerical study reveals the need for taking control of both correlation and finite-size effects. In this respect the VCA turns out to be especially advantageous in the weak-to-intermediate coupling regime.

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