

Exact-diagonalization study of exciton condensation in electron bilayers

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We report on small-cluster exact-diagonalization calculations which prove the formation of electron-hole pairs (excitons) as a prerequisite for spontaneous interlayer phase coherence in double-layer systems described by the extended Falicov-Kimball model. Evaluating the anomalous Green's function and momentum distribution function of the pairs, and thereby analyzing the dependence of the exciton binding energy, condensation amplitude, and coherence length on the Coulomb interaction strength, we demonstrate a crossover between a BCS-like electron-hole pairing transition and a Bose-Einstein condensation of tightly bound preformed excitons. We furthermore show that a mass imbalance between electrons and holes tends to suppress the condensation of excitons.

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I. INTRODUCTION

The formation of excitonic quantum condensates is an intensively studied continuous problem in condensed matter physics.¹⁻⁴ In a two-component (electron-hole) many-particle system, the attractive Coulomb interaction between oppositely charged electrons and holes can trigger their pairing and—under certain conditions—build up a macroscopic phase-coherent quantum state.

A variety of experimental attempts have been made to observe the condensed state of excitons in quasithermal equilibrium, e.g., in photoexcited semiconductors such as Cu₂O,⁵⁻⁹ or in unconventional semiconductor and bilayer graphene systems subject to electric and/or magnetic fields.¹⁰⁻¹⁴ Quite recently, the emergence of spontaneous coherence in a gas of indirect excitons in an electrostatic trap has been reported.¹⁵ Neutral electron-ion quantum plasmas are other promising candidates for exciton condensates.^{16,17}

From a theoretical point of view, a possible continuous transition between a Bardeen-Cooper-Schrieffer (BCS) electron-hole pair condensate and a Bose-Einstein condensate (BEC) of preformed excitons has been of topical interest.^{4,18-23} However, exact results for the ground-state properties of strongly correlated electron-hole (excitonic) systems are rare. Gas (or fluid) models have recently been studied, e.g., by the diffusion quantum Monte Carlo method.^{24,25} Lattice fermion models with short-range Coulomb interaction, such as multiband Hubbard-like models,²⁶⁻²⁸ should be capable of describing the physics of exciton condensation as well, but they have not yet been thoroughly explored using unbiased numerical techniques.

Motivated by this situation, in this paper we made an attempt to address the problem of exciton condensation in electron-hole bilayers in terms of a minimal lattice fermion model, the so-called extended Falicov-Kimball model (EFKM).²⁹⁻³³ Originally the EFKM described a two-band electron system with local Coulomb interaction between f - and c -band electrons, and it has been used to study electronic ferroelectricity,^{30,31,34} excitonic resonances,³⁵ or the excitonic insulator state.^{33,36-40} Different from these problems, in our double-layer (DL) system, the numbers of f and c particles are separately conserved, however, because charge transfer

between the two layers is assumed to be impossible. This mimics the generic situation in semiconductor electron-hole double quantum wells,^{12,41,42} bilayer quantum antiferromagnets,⁴³ and double-monolayer^{44,45} or double-bilayer graphene.⁴⁶

II. MODEL

The EFKM for an electron-hole DL takes the form

$$\mathcal{H} = -t_f \sum_{\langle i,j \rangle} (f_i^\dagger f_j + \text{H.c.}) - t_c \sum_{\langle i,j \rangle} (c_i^\dagger c_j + \text{H.c.}) - \mu_f \sum_i n_i^f - \mu_c \sum_i n_i^c + U \sum_i n_i^f n_i^c, \quad (1)$$

where f_i^\dagger (f_i) creates (annihilates) an electron in the f orbital at site i of the hole (or valence-band) layer, and $n_i^f = f_i^\dagger f_i$ is the f -particle number operator. The transfer amplitude between f orbitals on nearest-neighbor sites is denoted by t_f . Corresponding definitions apply for the c orbital of the electron (or conduction-band) layer. U (> 0) parametrizes the on-site interlayer (on-site) Coulomb attraction between f holes and c electrons. The spin degrees of freedom have been ignored for simplicity. Furthermore, we assume a band structure with a direct band gap ($t_c t_f < 0$) as shown in Fig. 1.

Taking into account the experimental situation,^{5-13,17,45} we assume that the excited electrons and holes have an infinite lifetime, that the number of excited electrons is equal to the number of excited holes, and that the number of bound pairs (excitons) can be viewed as an input parameter, independent of the interaction strength. In practice, we adjust the chemical potentials μ_f and μ_c to maintain the number of electrons in the f and c layer separately, thereby fixing the average f - and c -particle density per site as n^f and n^c , respectively. Due to this simplified description Eq. (1), issues such as the exciton Mott transition and biexciton formation¹⁶ are beyond the scope of this work.

III. THEORETICAL APPROACH

We employ a Lanczos exact-diagonalization technique for a finite square lattice with periodic boundary conditions (see Fig. 1) and calculate the anomalous Green's function for

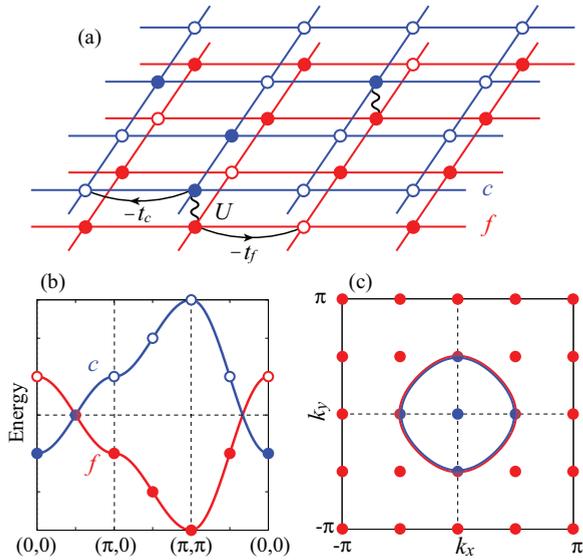


FIG. 1. (Color online) (a) Schematic representation of the DL EFKM cluster model with $N_s = 16$ sites (32 orbitals). (b) Noninteracting tight-binding band structure and (c) square lattice Brillouin zone. Dots indicate the allowed momenta of the 4×4 lattice with periodic boundary conditions. Throughout this work, we assume filling factors $n^f = 0.75$ and $n^c = 0.25$, i.e., $(N_f, N_c) = (12, 4)$, which means $n^h = n^e = 0.25$, irrespective of U . The red and blue lines in (c) show the perfectly matching hole and electron Fermi surfaces, respectively, with finite-lattice Fermi momenta \mathbf{k}_F located at $\mathbf{k} = (\pm\pi/2, 0)$ and $(0, \pm\pi/2)$.

exciton condensation,

$$G^{cf}(\mathbf{k}, \omega) = \langle \psi_0^N | c_{\mathbf{k}}^\dagger \frac{1}{\omega + i0^+ - \mathcal{H} + E_0} f_{\mathbf{k}} | \psi_0^N \rangle, \quad (2)$$

in the momentum (\mathbf{k}) and frequency (ω) space, where $|\psi_0^N\rangle$ is the ground-state wave function and E_0 is the ground-state energy of a system with N electrons. We define the anomalous spectral function

$$F(\mathbf{k}, \omega) = -\frac{1}{\pi} \text{Im} G^{cf}(\mathbf{k}, \omega) \quad (3)$$

and denote its frequency integral by $F_{\mathbf{k}}$. Clearly, the anomalous Green's function vanishes in finite systems without long-range phase coherence. We therefore have to assume the presence of the state $|\psi_0^N\rangle$, which is a coherent superposition of states with different numbers of excited electrons and holes (or excitons) at a given number N , just as for the BCS wave function of superconductors where the number of electrons is also not conserved. To detect particle fluctuations of the exciton condensate, we adopt a technique introduced for the evaluation of the superconducting anomalous Green's function on small clusters,^{47,48} which allows for the calculation of the off-diagonal Green's functions with respect to varying particle numbers [see Eq. (4)]. We thus monitor the excitonic pairing instability via the anomalous excitation spectrum (corresponding to the Bogoliubov quasiparticle spectrum in superconductors). Note that the term ‘‘anomalous’’ is used to indicate that the number of electrons on each of the f and c bands is not conserved in the course of exciton condensation

(or spontaneous c - f hybridization) although the total number of electrons N is conserved.

Having $G^{cf}(\mathbf{k}, \omega)$ determined, we can calculate the condensation amplitude $F_{\mathbf{k}}$ (following Refs. 47 and 48) from

$$F_{\mathbf{k}} = \langle N_f - 1, N_c + 1 | c_{\mathbf{k}}^\dagger f_{\mathbf{k}} | N_f, N_c \rangle, \quad (4)$$

where $|N_f, N_c\rangle$ is the ground state with the fixed numbers of f and c electrons, and subsequently we will be able to determine the order parameter,

$$\Delta = \frac{U}{N_s} \sum_{\mathbf{k}} F_{\mathbf{k}}, \quad (5)$$

and the coherence length,

$$\xi = \sqrt{\frac{\sum_{\mathbf{k}} |\nabla_{\mathbf{k}} F_{\mathbf{k}}|^2}{\sum_{\mathbf{k}} |F_{\mathbf{k}}|^2}}, \quad (6)$$

for the excitonic condensate (N_s denotes the number of lattice sites).

The binding energy of an exciton E_B should be equal to twice the order parameter Δ in the weak-coupling limit and deviate largely from this value in the strong-coupling regime. Within our finite-cluster approach, E_B may be obtained representing the orbital flavor by electron-hole variables, i.e., $f_i^\dagger \rightarrow h_i$ and $c_i^\dagger \rightarrow e_i^\dagger$. As a result, the interaction term of the DL EFKM takes the form $U \sum_i n_i^f n_i^c \rightarrow -U \sum_i n_i^e n_i^h + U \sum_i n_i^e$, where, in addition to the attractive electron-hole interaction, an extra on-site energy term appears. Due to this term, we should first determine the energy for the addition and removal of an electron:

$$E_B^+ = E_0(N_f - 1, N_c + 1) + E_0(N_f, N_c) - 2E_0(N_f, N_c + 1) + U, \quad (7)$$

$$E_B^- = E_0(N_f - 1, N_c + 1) + E_0(N_f, N_c) - 2E_0(N_f - 1, N_c) - U, \quad (8)$$

where $E_0(N_f, N_c)$ is the ground-state energy of the system with (N_f, N_c) electrons. Then, if $|t_f| = t_c$, the exciton binding energy E_B equals $E_B^+ = E_B^-$. For the mass-asymmetric case $|t_f| \neq t_c$, however, $E_B^+ \neq E_B^-$ because $E_0(N_f, N_c + 1) - U \neq E_0(N_f - 1, N_c)$. Hence, E_B should be defined as the average of E_B^+ and E_B^- , i.e., in general the exciton binding energy is given by

$$E_B = E_0(N_f - 1, N_c + 1) + E_0(N_f, N_c) - E_0(N_f - 1, N_c) - E_0(N_f, N_c + 1). \quad (9)$$

Finally, introducing a creation operator $b_{\mathbf{q}}^\dagger = (1/\sqrt{N_s}) \sum_{\mathbf{k}} c_{\mathbf{k}+\mathbf{q}}^\dagger f_{\mathbf{k}}$ of an excitonic quasiparticle with momentum \mathbf{q} , the momentum distribution function of excitons can be obtained from

$$N_{\mathbf{q}} = \langle N_f, N_c | b_{\mathbf{q}}^\dagger b_{\mathbf{q}} | N_f, N_c \rangle. \quad (10)$$

IV. NUMERICAL RESULTS

A. Mass-symmetric case

We now present the results of our exact-diagonalization study. Let us first examine the DL EFKM without mass

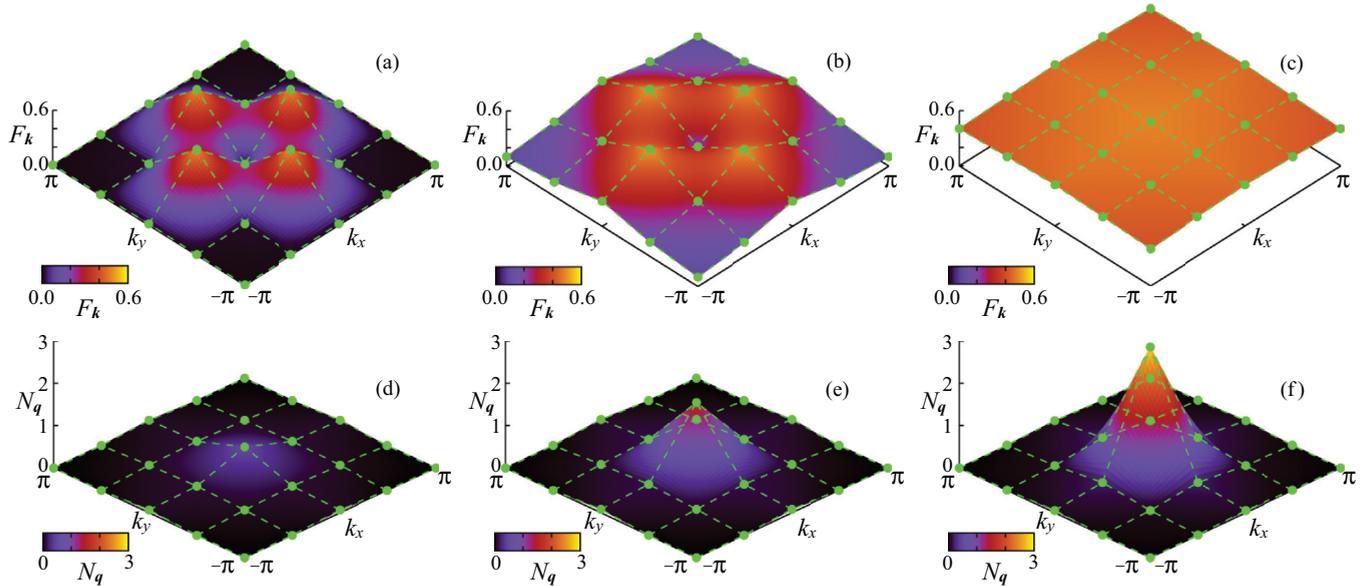


FIG. 2. (Color online) Condensation amplitude F_k (upper panels) and momentum distribution function N_q of excitons (lower panels) in the mass-symmetric DL EFKM with $U/t = 0.5$ (left), 5 (middle), and 50 (right).

imbalance, i.e., $|t_f| = t_c \equiv t$. Figure 2 shows the corresponding data for the condensation amplitude F_k and the exciton momentum distribution N_q , in a wide parameter range of U/t . In the weak-coupling regime [panels (a) and (d)], F_k exhibits pronounced maxima at the Fermi momenta, $\mathbf{k}_F = (\pm\pi/2, 0)$, $(0, \pm\pi/2)$, and decreases rapidly away from the “Fermi surface,” pointing toward a BCS-type instability of weakly bound electron-hole pairs with s -wave symmetry. As U/t increases, F_k broadens in momentum space [panel (b)], indicating that the radius of the bound electron-hole objects becomes smaller in real space. Accordingly, N_q is enhanced at momentum $\mathbf{q} = (0, 0)$; see Fig. 2(e). In the strong-coupling regime [panels (c) and (f)], F_k is homogeneously spread over the entire Brillouin zone, whereas N_q is sharply peaked at $\mathbf{q} = (0, 0)$, which is a sign of a BEC of tightly bound excitons. That is to say, as the attraction between electrons and holes increases in the DL EFKM, we get evidence for a BCS-BEC crossover scenario.

The behavior of the coherence length depicted in Fig. 3 as a function of the Coulomb attraction corroborates this finding. The spatial coherence of the excitonic state decreases with increasing U/t , indicating that the character of the condensate changes from BCS-like to BEC-like. That ξ stays finite as $U/t \rightarrow 0$ is an obvious artifact of our small cluster calculation. Figure 3 also displays the functional dependence of both the exciton order parameter and the exciton binding energy on U/t . The results may be compared with those of the BCS mean-field theory,^{21,22} which gives Δ and E_B as a solution of the self-consistent equations

$$1 = \frac{U}{2N_s} \sum_k \frac{1}{\sqrt{(\varepsilon_k - \bar{\mu})^2 + \Delta^2}}, \quad (11)$$

$$2n = 1 - \frac{1}{N_s} \sum_k \frac{\varepsilon_k - \bar{\mu}}{\sqrt{(\varepsilon_k - \bar{\mu})^2 + \Delta^2}}, \quad (12)$$

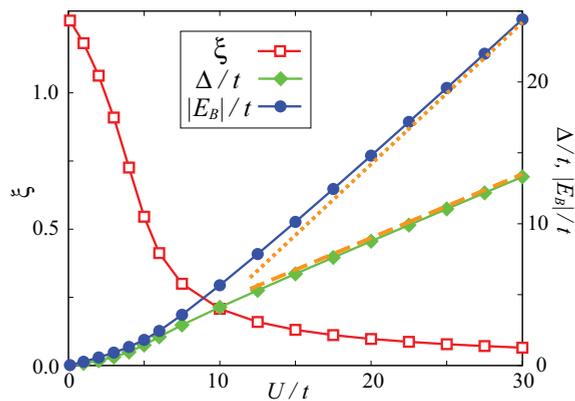


FIG. 3. (Color online) Coherence length ξ (squares), order parameter Δ (diamonds), and exciton binding energy $|E_B|$ (circles) for the mass-symmetric DL EFKM as functions of U/t . For comparison, the asymptotics in the strong-coupling limit $\Delta \propto 0.45U$ (dashed line) and $|E_B| \propto U$ (dotted line) have been inserted.

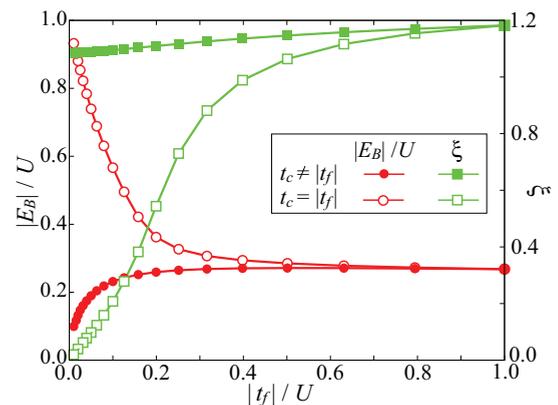


FIG. 4. (Color online) Binding energy $|E_B|/U$ (left ordinate) and coherence length ξ (right ordinate) for the mass-asymmetric (filled symbols) and mass-symmetric (open symbols) DL EFKM as functions of $|t_f|/U$ at $t_c/U = 1$.

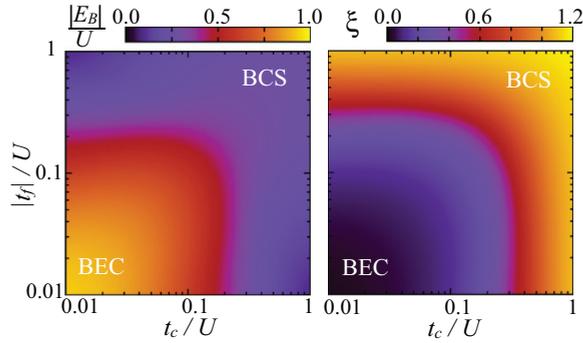


FIG. 5. (Color online) Exciton binding energy E_B/U (left panel) and coherence length ξ (right panel) of the DL EFKM in the t_c/U - $|t_f|/U$ plane.

where $\varepsilon_{\mathbf{k}} = 2t(\cos k_x + \cos k_y)$, $n = n^e = n^h$, $\bar{\mu} = \mu - U(n - 1/2)$, and $\mu_f = -\mu_c = \mu$.

In the weak-coupling limit, we should recover the usual BCS picture. Δ should therefore increase exponentially with U : $\Delta \propto \exp[-1/\rho(\varepsilon_F)U]$, thereby satisfying the relation $|E_B| = 2\Delta$, with $\rho(\varepsilon_F)$ being the density of states at the Fermi level. In the strong-coupling limit, on the other hand, the BCS equations yield the asymptotic behavior: $\Delta = U\sqrt{n(1-n)} = \sqrt{3}U/4 \simeq 0.433U$ and $|E_B| = 2\sqrt{\bar{\mu}^2 + \Delta^2} = U$. The numerical results obtained for Δ and $|E_B|$ show that we find the BCS relation $|E_B| = 2\Delta$ at weak couplings. In the strong-coupling limit, Δ and $|E_B|$ are found to be $\propto 0.45U$ and $\propto U$, respectively, which matches the BEC of composite bosons, where $\Delta = 0.433U$ and $|E_B| = U$ for $U/t \rightarrow \infty$.

B. Mass-asymmetric case

We finally address the effects of a mass imbalance between f holes and c electrons. Since $|t_f| \neq t_c$, it makes sense to use U as the unit of energy and determine the exciton binding energy E_B and coherence length ξ in dependence on $|t_f|/U$. Figure 4 shows the results for $t_c/U = 1$ in comparison to the mass-symmetric case where a BCS-to-BEC transition occurs with decreasing $|t_f|/U$. In contrast, ξ is not reflective of such a crossover for $t_c \neq |t_f|$, and the exciton binding energy even weakens at strong couplings, $|t_f|/U \ll 1$.

In the strong-coupling region, where both $|t_f|/U$ and t_c/U are small, the EFKM can be mapped onto the XXZ quantum spin-1/2 model in a magnetic field,³⁰

$$\mathcal{H}_{\text{eff}} = J \sum_{\langle i,j \rangle} [\boldsymbol{\tau}_i \cdot \boldsymbol{\tau}_j + \delta \tau_i^z \tau_j^z] - B_z \sum_i \tau_i^z \quad (13)$$

with $\boldsymbol{\tau}_i = (1/2) \sum_{\alpha,\beta} \alpha_i^\dagger \boldsymbol{\sigma}_{\alpha\beta} \beta_i$ ($\alpha, \beta = f, c$; $\boldsymbol{\sigma}$ is the vector of Pauli matrices), $J = 4|t_f|t_c/U$, and $\delta = (|t_f| - t_c)^2/(2|t_f|t_c)$. $B_z = 2\mu$ is determined in order to maintain $\sum_i \tau_i^z = 1/4$. The

effective model is isotropic in spin space for the case of $|t_f| = t_c$, and it exhibits antiferromagnetic order in the x - y plane at zero temperature. This long-range ordered state corresponds to an exciton condensate in the original EFKM. Different hopping parameters $t_c \neq |t_f|$ give rise to an Ising anisotropy δ , which tends to suppress the x - y antiferromagnetic order. Accordingly, the exciton binding energy $|E_B|$ (excitonic condensate) is suppressed as $|t_f|/U \rightarrow 0$.

Figure 5 compiles our E_B (left panel) and ξ (right panel) data by two contour plots in the t_c/U - $|t_f|/U$ plane. For the mass-symmetric case $t_c = |t_f|$, i.e., on the diagonals of Fig. 5, both $|E_B|/U$ and ξ indicate a smooth crossover from BCS to BEC as U increases. On the other hand, at sufficiently weak Coulomb interactions, $t_c/U \gtrsim 0.3$, we stay in the BCS-like state as $|t_f|/U$ is varied by changing the absolute value of t_f/t_c . Note that a strong mass imbalance between electrons and holes acts in a “pair-breaking” way in both the BCS⁴⁹ and BEC^{26,50} limits.

V. SUMMARY

Based on unbiased exact-diagonalization calculations for the two-dimensional extended Falicov-Kimball model, we have studied the formation of excitons in both mass-symmetric and mass-asymmetric electron-hole double-layer systems (bilayers) and provided, most notably, strong evidence for exciton condensation and a BCS-BEC crossover scenario at zero temperature. Thereby, the properties of the excitonic quasiparticles and the nature of the condensation process were analyzed, exploiting the anomalous Green’s function in order to determine the order parameter of the condensate and coherence length, as well as the binding energy and momentum distribution function of excitons. The weak and strong correlation limits are discussed and put into perspective to approximative analytical approaches. We corroborated previous analytical^{26,49,50} and numerical¹⁷ findings to the effect that a mass imbalance between electrons and holes might suppress the condensation of excitons. This holds even in the strong-coupling regime. We hope that the presented results will stimulate further experimental studies of exciton condensation in bilayer systems with strong electronic correlations.

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