Order, criticality and excitations in the extended Falicov-Kimball model

S. Ejima, T. Kaneko, Y. Ohta, and H. Fehske

1 Institut für Physik, Ernst-Moritz-Arndt-Universität Greifswald, 17489 Greifswald, Germany
2 Department of Physics, Chiba University, Chiba 263-8522, Japan

(Dated: May 11, 2014)

PACS numbers: 71.35.-y, 71.10.Hf

Using exact numerical techniques we investigate the nature of excitonic (electron-hole) bound states and the development of exciton coherence in the one-dimensional half-filled extended Falicov-Kimball model. The ground-state phase diagram of the model exhibits, besides band insulator and staggered orbital ordered phases, an excitonic insulator (EI) with power-law correlations. The criticality of the EI state shows up in the von Neumann entropy. The anomalous spectral function and condensation amplitude provide the binding energy and coherence length of the electron-hole pairs, which, on their part, point towards a Coulomb interaction driven crossover from BCS-like electron-hole pairing fluctuations to tightly bound excitons. We show that while a mass imbalance between electrons and holes does not affect the location of the BCS-BEC crossover regime it favors staggered orbital ordering to the disadvantage of the EI. Within the BEC regime the quasiparticle dispersion develops a flat valence-band top in accord with the experimental finding for Ta_{2}NiSe_{5}.

The detection of the EI state in Ta_{2}NiSe_{5} has raised and attracted much experimental attention [6]. Most notably, by angle-resolved photoemission spectroscopy (ARPES), an extremely flat valence-band top at 40 K was observed and taken as a strong indication and possible condensation of excitons in 1D systems [7]. The minimal theoretical model in this respect is a candidate for the EI state, quasi one-dimensional (1D) Ta_{2}NiSe_{5}, has raised and attracted much experimental attention [8].

The extended FKM (EFKM) [Eq. (1)] has been studied extensively in the context of EI formation for D > 1, using DMFT [12], random phase approximation [13], slave-boson [14], projective renormalization [15] and variational cluster [16] techniques, or purely numerical diagonalization procedures [17]. At the same time, the problem of electronic ferroelectricity, which is equivalent to the appearance of the EI in some theoretical models, has also attracted much attention [18, 19].

In this paper we present a comprehensive numerical analysis of the 1D EFKM at half filling. At first we determine the ground-state phase diagram from large-scale density matrix renormalization group (DMRG) [20].
calculations and identify—depending on the orbital level splitting—staggered orbital ordered (SOO) and band insulator (BI) phases as well as an intervening critical EI state. Then, within the EI, we detect a crossover between BCS- and Bose-Einstein-type condensates monitoring the exciton-exciton correlation and exciton momentum distribution functions. Note that in our 1D setting we use the term ‘condensate’ to indicate a critical phase with power-law correlation decay. Finally, combining DMRG, Lanczos exact diagonalization (ED) and Green functions techniques [24], we study the anomalous spectral function and extract the correlation length and binding energy of the electron-hole pairs. This allows us to comment on the nature of the excitonic bound states preceding the condensation process and to discuss the effect of a mass imbalance between (c-) electrons and (f-) holes.

Examining the (large-U) strong-coupling regime gives a first hint of which phases might be realized in the 1D EFKM at zero temperature. To leading order the EFKM can be mapped onto the exactly solvable spin-1/2 XXZ-Heisenberg model in a magnetic field \( h = D \) aligned in the z-direction [25], \( H_{\text{XXZ}} = J \sum_j \{ \Delta S_j^z S_{j+1}^z + (1/2)(S_j^x S_{j+1}^x + S_j^y S_{j+1}^y) \} - h \sum_j S_j^z \) with \( J = 4|t_f|t_c/U \) and \( \Delta = (t_f^2 + t_c^2)/(2|t_f|t_c) \). The XXZ model exhibits three phases: the gapped antiferromagnetic (AF) phase, the critical gapless XY phase with central charge \( c = 1 \), and the ferromagnetic (FM) phase, where both transition lines, those between AF and XY phases (\( h_{c2}/J \)) and those between XY and FM phases (\( h_{c2}/J \)), follow from the Bethe ansatz [26]. Correspondingly, increasing the magnitude of the f-c level splitting \( D \) in the EFKM, we expect to find the following sequence of phases: (i) the SOO phase that matches the Ising-like AF phase in the XXZ model, (ii) an intermediate critical EI phase with finite excitonic binding energy, and (iii) a BI state, which is characterized by a filled (empty) \( f \) (c) band and related to the FM phase of the XXZ model. The phase boundary separating the EI and BI states is exactly known to be [27]

\[ D_{c2} = \sqrt{4(|t_f| + |t_c|)^2 + U^2 - U}. \]

The complete phase diagram of the 1D EFKM is presented in Fig. 1. Symbols denote the DMRG BI-EI and EI-SOO transition points, which can be obtained from the energy differences

\[ D_{c2}(L) = E_0(L, 0) - E_0(L - 1, 1) = -E_0(L - 1, 1), \]

and

\[ D_{c1}(L) = E_0(L/2 + 1, L/2 - 1) - E_0(L/2, L/2), \]

respectively, in the course of a finite-size scaling analysis (see the inset). Here \( E_0(N_f, N_c) \) denotes the ground-state energy for a system with \( N_f \) \( f \)- and \( N_c \) \( c \)-electrons at \( D = 0 \). Note that Eq. (3) holds for both, open and periodic boundary conditions (OBC/PBC), whereas Eq. (4) has to be evaluated with PBC (if here OBC were used, an extra factor 2 results: \( D_{c1}^{\text{OBC}} = 2D_{c1} \)). For the DMRG runs performed in this work we keep at least \( m = 3200 \) density-matrix eigenstates which ensures a discarded weight smaller than \( 1 \times 10^{-6} \). The \( D_{c2}(L \to \infty) \) values demonstrate the accuracy of our DMRG calculations. Exact results for \( D_{c1}(L \to \infty) \) can only be obtained numerically, where a comparison with the dotted line reveals the limits of the strong-coupling approach [25]; see Fig. 1. The criticality of the EI phase—corresponding to the critical XY phase in the XXZ model with central charge \( c = 1 \)—can be confirmed by the von Neumann entanglement entropy \( S_L(t) = -\text{Tr}_\ell(\rho_\ell \ln \rho_\ell) \) (with reduced density matrix \( \rho_\ell = \text{Tr}_{L-\ell}(\rho) \)). Numerically, the central charge is best estimated from the entropy difference [28, 29].

\[ c^*(L) \equiv 3[S_L(L/2 - 1) - S_L(L/2)]/\ln[\cos(\pi/L)]. \]

Our results for \( c^* \), displayed in the lower panel of Fig. 1 for \( |t_f| = 0.1 \) at \( U = 5 \), give clear evidence that \( c^* \to 1 \) in the EI, whereas we find \( c^* = 0 \) in the BI and SOO phases.
Regrettably, $e^*(L)$ is strongly system-size dependent near the EI-SOO transition.

Let us now discuss the nature of the EI state in more detail. For simplicity we consider the case $t_f t_e < 0$, where two Fermi points $(\pm k_F)$ exist for $U = 0$ provided $D$ is sufficiently small (otherwise a direct band gap emerges). As a signature of an excitonic Bose-Einstein condensate in 1D one expects (i) a power-law decay of the correlations $(b_i^\dagger b_j)$ with $b_i^\dagger = c_i^\dagger f_i$ and (ii) a divergence of the excitonic momentum distribution $N(q) = \langle b_i^\dagger b_q \rangle$ with $b_q^\dagger = (1/\sqrt{L}) \sum_k c_{k+q}^\dagger f_k$ for the state with the lowest possible energy (in the direct gap case at $q = 0$) due to the absence of true long-range order. Figure 2 supports these expectations: Whereas in the weak-coupling BCS regime ($U = 1$), $(b_i^\dagger b_j)$ decays almost exponentially and $N(q)$ shows only a marginal system-size dependence (for all momenta), in the strong-coupling BEC regime close to the EI-BI transition ($U = 1.9$), $(b_i^\dagger b_j)$ exhibits a rather slow algebraic decay of the excitonic correlations and $N(q = 0)$ becomes divergent as $L \to \infty$.

We note that the $\langle c_i^\dagger f\rangle$-expectation value is always zero for a 1D system in the absence of an explicit $f$-$c$-band hybridization. To examine the BCS-BEC crossover we adopt a technique introduced for detecting the particle fluctuations of Cooper pairs in 2D systems [24]. That is, we consider the off-diagonal anomalous exciton Green function

$$G_{cf}(k, \omega) = \langle \psi_1 | c_k^\dagger f_k | \psi_0 \rangle,$$

where $|\psi_0\rangle$ is the ground state $|N_f, N_c\rangle$ with fixed numbers of $f$- and $c$-electrons, $|\psi_1\rangle$ is the excited state $|N_f - 1, N_c + 1\rangle$, $E_0$ is the averaged energy of $|\psi_0\rangle$ and $|\psi_1\rangle$, and $\eta$ is a broadening, and determine the corresponding spectral function $F(k, \omega) = (-1/\pi) ImG_{cf}(k, \omega)$ that gives the condensation amplitude $F(k) = \langle \psi_1 | c_k^\dagger f_k | \psi_0 \rangle$. $F(k)$ can be directly computed by the ground-state DMRG method taking into account an extra target state $|\psi_1\rangle$.

From $F(k)$ the coherence length characterizing the excitonic condensate follows as

$$\xi^2 = \sum_k |\nabla_k F(k)|^2 / \sum_k |F(k)|^2.$$  

The binding energy of the excitons, $E_B$, can be also determined from diverse ground-state energies [17]:

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

Figures 3(a) and 3(b) show the anomalous spectral function $F(k, \omega)$ in the weak-coupling ($U = 1$) and strong-coupling ($U = 1.9$) regimes, respectively, where $D = 1$. In the former case the EI arises from a semimetallic phase. As a consequence most of the spectral weight of the quasiparticle excitations is located around the Fermi momentum $k_F = \pm k_F$, again indicating a BCS-type pairing of electrons and holes. Obviously, Fermi surface effects play no role for large $U$ where the Hartree shift drives the system in the semiconducting regime. Here the excitation gap occurs at $k = 0$. Note that the gap between the lowest energy peaks in $F(k, \omega)$ is equal to the binding energy.
$E_B$ given by Eq. (8). Figure 3(c) displays the frequency-integrated quantity $F(k)$. At $U = 1$, $F(k)$ exhibits a sharp peak at the Fermi momentum. Increasing $U$ the peak weakens and shifts to smaller momenta. Close to the EI-BI transition point $U = 1.9 \lesssim U_c = 1.92$, $F(k)$ has a maximum at $k = 0$ but is spread out in momentum space, indicating that the radius of electron-hole pairs becomes small in real space. Panel (d) gives the quasiparticle dispersion $E(k)$ derived from $A(k,\omega)$. Driving the BCS-BEC crossover by increasing $U$, the peaks around $k = \pm k_F$ disappear as well as the notch around $k = 0$. Instead a valence band with a flat top around $k = 0$ develops, just as observed e.g. in quasi-1D Ta$_2$NiSe$_5$ [9].

Figure 4 shows the variation of the coherence length and the binding energy in the EI phase of the 1D EFKM with $|t_f| = 1$ (left panels) and 0.1 (right panels). At small $U$ the excitonic state is composed of electron-hole pairs having large spatial extension, leading to large values of $\xi$. $E_B$, on the other hand, is rather small, but increases exponentially with $U$. This typifies a BCS pairing mechanism. At large $U$, the binding increases linearly with $U$. Here, tightly bound spatially confined excitons acquire quantum coherence (with $\xi \ll 1$) in a Bose-Einstein condensation process.

We finally address the influence of a mass imbalance between $f$- and $c$-band quasiparticles. The EI phase is absent for $t_f = 0$. In the mass-symmetric case $|t_f| = t_c$, the 1D Hubbard model results for $D = 0$. Here we cannot distinguish between the AF (with vanishing spin gap) and EI phases, because both phases are critical. Therefore, in this limit, we have examined the 1D EFKM for $N_f > L/2$. To this end, both the $U$ and $D$ axes in Fig. 4 have been rescaled by $(|t_f| + t_c)$, as suggested by the EI-BI transition lines (2). Indeed we find that EI phase shrinks as $|t_f|$ decreases. That is, the mass anisotropy gets stronger, which is simply a bandwidth effect, however, leading to a stronger Ising anisotropy. This, on their part, enlarges the SOO region, while the EI-BI phase boundary basically is unaffected. Importantly, the location of the BCS-BEC crossover, which can be derived from the intensity plots for $E_B$ and $\xi$, does not change in this presentation. To expose correlation effects, we included in Fig. 4 the semimetallic-to-semiconducting transition line assuming that the EI phase is absent. $U_{BI}(D)$ can be obtained from the band gap $\Delta_c$, that depends linearly on $U$ for fixed $D$: $\Delta_c(D) = U/2(|t_f| + t_c) + U_{BI}(D)$ [i.e., $U_{BI}(D)$ scales again with $|t_f| + t_c$]. Apparently in the BCS-BEC crossover regime a strong renormalization of the band structure due to the incipient $f$-$c$ hybridization takes place.

To conclude, adopting the numerically exact density matrix renormalization group technique, we examined the one-dimensional (1D) extended Falicov-Kimball model (EFKM) and, most notably, proved the excitonic insulator (El) state shown to be critical. The complete ground-state phase diagram was derived, and put into perspective with the Bethe ansatz results obtained in the strong-coupling limit for the spin-1/2 XXZ chain. Besides the Ei to band insulator transition, the boundary between the EI and a phase with staggered orbital ordering was determined with high accuracy. The whole phase diagram of the 1D EFKM could be scaled by $|t_f| + t_c$: staggered orbital ordering appears only for small mass-imbalance ratios $|t_f|/t_c$. The absence of an order parameter prevents addressing the problem of excitonic condensation in 1D systems by usual mean-field approaches. That is why we exploited the off-diagonal anomalous Green function. The related anomalous spectral function elucidates the different nature of the electron-hole pairing and condensation process at weak and strong couplings. At fixed level splitting the binding energy between $c$ electrons and $f$ holes is exponentially small in the weak-coupling regime. It strongly increases as the Coulomb attraction increases. Concomitantly the coherence length of the electron-hole pair condensate shortens. This unambiguously demonstrates a crossover from BCS-like electron-hole pairing to a Bose-Einstein condensation of preformed excitons. The quasiparticle band dispersion in the BEC regime exhibits a rather dispersionless

FIG. 4. (Color online) Intensity plots of the binding energy $E_B$ (upper panels; $L = 128$, OBC) and the coherence length $\xi$ (lower panels; $L = 64$, PBC) in the rescaled $U/(|t_f| + t_c)$–$D/(|t_f| + t_c)$ plane. Data were calculated by the DMRG for $N_f > L/2$ (to avoid the AF state in the Hubbard model limit $|t_f| = 1$, $D = 0$). Solid lines denote the SOO-EI and EI-BI transition points in the thermodynamic limit (in the lower panels the small uncolored slot just above the SOO-EI appears because $|E_B|$ and $\xi$ are obtained here for a fixed finite system size). The dashed line [$U_{BI}(D)$] would separate the semimetallic and semiconducting phases if the EI is assumed to be absent.
valence band near $k = 0$, despite the fact that the expectation value $\langle c^f \rangle$ is zero because of the 1D setting. This result further supports the EI scenario for quasi-1D $\text{Ta}_2\text{NiSe}_5$, where the flat valence-band top was detected by ARPES experiments.

The authors would like to thank Y. Fuji, F. Göhmann, S. Nishimoto, K. Seki, T. Shirakawa, and B. Zenker for valuable discussions. S.E. and H.F. acknowledge funding by the DFG through SFB 652 Project B5. T.K. was supported by a JSPS Research Fellowship for Young Scientists. Y.O. acknowledges the Japanese Kakenhi Grant No. 22540363.

[1] N. F. Mott, Philos. Mag. 6, 287 (1961); L. V. Keldysh and H. Y. V. Kopaev, Sov. Phys. Sol. State 6, 2219 (1965); D. Jérome, T. M. Rice, and W. Kohn, Phys. Rev. 158, 462 (1967).
[2] J. Neuenschwander and P. Wachter, Phys. Rev. B 41, 12693 (1990); P. Wachter, B. Bucher, and J. Malar, ibid. 69, 094502 (2004).
[3] H. Cercellier, C. Monney, F. Clerc, C. Battaglia, L. Despont, M. G. Garnier, H. Beck, P. Aebi, L. Patthey, H. Berger, and L. Forró, Phys. Rev. Lett. 99, 146403 (2007); C. Monney, C. Battaglia, H. Cercellier, P. Aebi, and H. Beck, ibid. 106, 106404 (2011).
[4] H. Min, R. Bistritzer, J. J. Su, and A. H. MacDonald, Phys. Rev. B 78, 121401(R) (2008); T. Stroucken, J. H. Grönqvist, and S. W. Koch, J. Opt. Soc. Am. B 29, A86 (2012).
[5] F. X. Bronold and H. Fehske, Phys. Rev. B 74, 165107 (2006).
[6] Y. Wakisaka, T. Sudayama, K. Takubo, T. Mizokawa, M. Arita, H. Namatame, M. Taniguchi, N. Katayama, M. Nohara, and H. Takagi, Phys. Rev. Lett. 103, 026402 (2009).
[7] T. Kaneko, T. Toriyama, T. Konishi, and Y. Ohta, Phys. Rev. B 87, 035121 (2013); T. Kaneko, T. Toriyama, T. Konishi, and Y. Ohta, ibid. 87, 199902(E) (2013).
[8] L. M. Falicov and J. C. Kimball, Phys. Rev. Lett. 22, 997 (1969); P. Farkašovský, Eur. Phys. J. B 20, 209 (2001).
[9] C. D. Batista, Phys. Rev. Lett. 89, 166403 (2002).
[10] T. Kennedy, Rev. Math. Phys. 6, 901 (1994).
[11] J. K. Freericks and V. Zlatić, Rev. Mod. Phys. 75, 1333 (2003).
[12] A. Taraphder, S. Koley, N. S. Vidhyadhiraja, and M. S. Laad, Phys. Rev. Lett. 106, 236405 (2011).
[13] B. Zenker, D. Ihle, F. X. Bronold, and H. Fehske, Phys. Rev. B 85, 121102R (2012).
[14] P. M. R. Brydon, Phys. Rev. B 77, 045109 (2008); B. Zenker, D. Ihle, F. X. Bronold, and H. Fehske, ibid. 81, 115122 (2010).
[15] V. N. Phan, K. W. Becker, and H. Fehske, Phys. Rev. B 81, 205117 (2010).
[16] K. Seki, R. Eder, and Y. Ohta, Phys. Rev. B 84, 245106 (2011); T. Kaneko, K. Seki, and Y. Ohta, ibid. 85, 165135 (2012).
[17] T. Kaneko, S. Ejima, H. Fehske, and Y. Ohta, Phys. Rev. B 88, 035123 (2013).
[18] T. Portengen, T. Östreich, and L. J. Sham, Phys. Rev. Lett. 76, 3384 (1996).
[19] U. K. Yadav, T. Maitrala, I. Singh, and A. Taraphder, Europhys. Lett. 93, 47013 (2011).
[20] C. D. Batista, J. E. Gubernatis, J. Bonča, and H. Q. Lin, Phys. Rev. Lett. 92, 187601 (2004).
[21] P. Farkašovský, Phys. Rev. B 59, 9707 (1999); P. Farkašovský, ibid. 65, 081102 (2002).
[22] C. Schneider and G. Czycholl, Eur. Phys. J. B 64, 43 (2008); P. Farkašovský, Phys. Rev. B 77, 155130 (2008).
[23] S. R. White, Phys. Rev. Lett. 69, 2863 (1992).
[24] Y. Ohta, T. Shimozato, R. Eder, and S. Maekawa, Phys. Rev. Lett. 73, 324 (1994); Y. Ohta, A. Nakauchi, R. Eder, K. Tsutsui, and S. Maekawa, Phys. Rev. B 52, 15617 (1995).
[25] G. Fáth, Z. Domański, and R. Lemański, Phys. Rev. B 52, 13910 (1995).
[26] J. des Cloizeaux and M. Gaudin, J. Math. Phys. 7, 1384 (1966); M. Takahashi, Thermodynamics of One-Dimensional Solvable Models (Cambridge University Press, Cambridge, 1999).
[27] A. N. Kocharian and J. H. Sebold, Phys. Rev. B 53, 12804 (1996).
[28] P. Calabrese and J. Cardy, J. Stat. Mech. P06002 (2004).
[29] S. Nishimoto, Phys. Rev. B 84, 195108 (2011).