Exciton condensation in an extended Falicov-Kimball model in the presence of orbital magnetic fields

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Abstract – We investigate the exciton condensation in the presence of an external, perpendicular magnetic field in a two-dimensional extended spinless Falicov-Kimball model involving itinerant (c) and localized (f) electrons in the half-filled limit, using self-consistent, mean-field approximations. On tuning the orbital magnetic field the excitonic averages \( \Delta_{i} = \langle c_{i}^{\dagger} f_{i} \rangle \) are affected in several ways: the external field usually suppresses the excitonic average but we find that it is also possible to enhance excitonic response at some values of the magnetic field. We further examine the effect of Coulomb interaction and the f-electron hopping on the condensation of excitons for some rational values of the applied magnetic fields. The interband Coulomb interaction enhances \( \Delta \) exponentially and the effect is more pronounced for low hybridization. The strength of excitonic average drops when f-electrons have a dispersion. This trend is independent of the relative sign of the c- and f-electron hopping; although the excitonic response is different for different parity of the c- and f-electrons.

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Introduction. – The weak screening of the Coulomb interaction between the conduction band electrons and the valence band holes causes the formation of an excitonic bound state in some materials. A plethora of theoretical and experimental work has been performed for decades to unravel the physics of the preformed exciton in a variety of systems [1–11]. In the weak-coupling regime, the magnitude of the single-particle gap is almost comparable to that of the excitonic order parameter \( |\Delta| \). This indicates that electron-hole pair formation and condensation may occur simultaneously, such as the Cooper pair formation and condensation of such pairs in BCS theory. The problem of exciton formation can be studied in an extended Falicov-Kimball model (EFKM), since in the weak-coupling mean-field theory, the formation of an order parameter and the condensation thereof are concomitant [12]. The original Falicov-Kimball model [13], involving two spinless fermions, namely the delocalized c-electrons, atomic-like f-electrons and an on-site Coulomb interaction \( U \) between them, is perhaps the simplest lattice model to study many-body effects. The model which was introduced in 1969 to describe the valence or semiconductor-insulator transition in some transition metal oxides has been used beyond its original motivation. It is successful in explaining correlation effects [14], viz. metal-insulator transition in a host of systems [15], mixed-valence phenomena [16], the formation of ionic crystals [17,18] and the charge-density waves (CDW) [19]. The CDW in transition metal dichalcogenides has been discussed in the framework of excitonic mechanisms: a fluctuating preformed excitonic liquid condenses at the CDW transition. In the situation where the valence band is narrow, the gap between the valence and conduction band small, or an orbital-selective Mott transition emerges [20], correlated Hamiltonians like Falicov-Kimball model (FKM) are also used. There are, for example, recent reports of a possible transition to an excitonic insulating state in the quasi–one-dimensional Ta₂NiSe₅ due to Bose-Einstein condensation (BEC) of bound Ni 3d-Se 4p holes and Ta 5d electrons [21,22]. The spontaneous c-f hybridization between the valence states in Ni and Ta ions and single-particle spectra confirms the excitonic insulator state and BEC of spin-singlet exciton.
There are theoretical works in $D > 1$ dimension, where a hybridization between $c$- and $f$-band [23,24] results in a bound state between $c$-electrons and $f$-holes which underwent a BEC of $c$-$f$ “excitons”, provided the system is in a mixed valent regime. As $c$ and $f$ states differ by odd parity, a $k$-dependent hybridization could lead to an “electronic ferroelectricity” [25,26]. Batista et al. [27,28] explicitly showed that in dimensions $D > 1$, as a finite $f$-electron bandwidth breaks the local $U(1)$ symmetry, it induces a non-zero polarization even in the absence of $c$-$f$ hybridization. They reached this conclusion by mapping an EFKM ($V = 0$ but $t_f$ finite) onto a Hubbard model with asymmetric hopping ($t_\uparrow \neq t_\downarrow$) and thence to an effective anisotropic ($xxz$) ($s = 1/2$) spin model in the large $U$ limit with a “field” along the $z$-direction. The intermediate-coupling regime was treated with constrained path Monte Carlo (CPMC) technique. Batista et al. [27,28] and Sarasua and co-workers [29] claimed that a non-local hybridization in an EFKM stabilizes excitonic averages with the inclusion of $f$-electron hopping. Farkašovský performed the same analysis in a spinless extended FKM for $t_f < t_c$ under mean-field approximation with CDW instability in $D > 1$ dimension [30]. Customarily, ferroelectricity appears due to structural transitions. However, it is also possible that there is a non-vanishing $c$-$f$ coherence in a system leading to a hybridization term of purely electronic origin. This coherence could give rise to electric polarization due to Bose-Einstein condensation of $c$-$f$ excitons when the two bands differ by an oddparity. In multiferroic materials this can be induced by a magnetic field which has spawned a considerable interest owing to the tunability of the ferroelectric order through magnetic fields. The unconventional ferroelectricity arising out of $c$-$f$ coherence is, on the other hand, purely of electronic origin and offers an additional route to tuning optical properties with magnetic field. A moot question, therefore, is what happens to such an excitonic condensate in the presence of a magnetic field which acts oppositely on the electron and the hole [11]. Lozovik et al. [5] studied both direct and indirect excitons in coupled quantum wells (CQW) under a long-range Coulomb interaction in the presence of magnetic field. The authors consider a continuum model with screened Coulomb potential and draw a phase diagram for the excitons in magnetic field (normal to the plane) and momentum. They delineate the regions where the Coulomb interaction and magnetic field dominate the exciton formation, respectively.

In a very strong magnetic field, the two Zeeman-split bands of electrons are well separated in energy, and, only the lower band is relevant, effectively reducing the model to a spinless one. The field then couples to the orbital degrees via the canonical conjugate momenta. Zeeman coupling being absent, the field couples to the “orbital degrees.” In the presence of a gauge field, the hopping is now associated with a phase: the “Peierls phase.” Such gauge field can be experimentally realized in ultracold particles (fermions and bosons) in optical lattices [31]. An optical lattice provides the discrete periodic spatial structure and uses lasers instead of a magnetic field to induce a phase for particles hopping around a closed path in the lattice resembling an effective magnetic field. The strength of this effective magnetic field can be varied by laser parameters and be made arbitrarily large [32]. Moreover, of late, there are proposals for the realization of FKM in optical lattices with mixtures of light atoms in the correlated, disordered environment of heavy atoms [33,34]. Evidently, the exciton, an electron-hole bound state, will have strong effect from a magnetic field as the electron and the hole couple to the field with opposite signs. The interference between the hole’s and the electron’s Peierls phases affects the dispersion of the exciton. Thus, the dependence on the magnetic field becomes a periodic function of the flux quanta $\alpha$ [1]. There are very few results on the effect of orbital field in these situations. Therefore, it is quite pertinent to appraise an extended FKM (EFKM) in the presence of an artificial gauge field.

This paper is organized as follows: in the following section we describe the extension of the Falicov-Kimball model with different terms and outline the self-consistent mean-field theory to get the excitonic average. In the third section we examine our results without a magnetic field and then study its effect on exciton condensation for different model parameters in the fourth section. We also study the effect of the finite bandwidth of $f$-electrons in the exciton condensation in the presence of a magnetic field. Finally, we close with a short summary and conclusion in the last section.

**Model.** – The original FKM describes a two-band system of localized $f$-electrons and itinerant $c$-electrons with short-ranged $c$-$f$ Coulomb interaction $U$:

$$H_0 = -\sum_{\langle i, j \rangle} t_c (c_i^{\dagger} c_j + \text{h.c.}) + U \sum_i c_i^{\dagger} c_i f_i^{\dagger} f_i \tag{1}$$

where $\langle i, j \rangle$ are nearest-neighbour site indices on a square lattice (lattice constant $a = 1$), $c_i$ ($f_i$) are itinerant (localized) electron annihilation operators at site $i$. The first term is the kinetic energy due to hopping between nearest neighbours, where, $t_c$ is the hopping integral for $c$-electrons (kept fixed at 1 throughout our calculation). The second term represents the on-site Coulomb interaction between $c$- (density $n_c = \frac{1}{2} \sum_i c_i^{\dagger} c_i$) and $f$-electrons (at $E_f$, with density $n_f = \frac{1}{2} \sum_i f_i^{\dagger} f_i$; $N$ being the number of sites). This Hamiltonian commutes with $n_{f,i}$, in which case the local occupancy of an $f$-electron is either 0 or 1. It can be “solved” numerically by Monte-Carlo annealing over the $f$-electron positions [35]. As long as the $f$-electron number is conserved, no coherence between $c$-$f$ can be established. If the hybridization term $H_v = \sum_i V c_i^{\dagger} f_i + \text{h.c.}$ between these two bands is included, the local $U(1)$ symmetry of the $f$-electrons is
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Fig. 1: The mean-field behaviour of excitonic order parameter \( \Delta = \langle c_i^\dagger f_i \rangle \) calculated for different values of \( U \) with \( t_f = 0 \) in the zero-field limit for the symmetric case, i.e., \( E_f = 0 \).

lifted. The same happens for an extended FKM with finite \( f \)-electron bandwidth \((\sum |i\rangle \langle j| - t_f (f_i^\dagger f_j + h.c.)\)). The hybridization term \((V)\) or finite \( f \)-bandwidth removes the local \( U(1) \) gauge symmetry associated with the conservation of \( n_f \), and the Hamiltonian is no longer exactly solvable, albeit in the above sense. We, therefore, take recourse to the usual self-consistent mean-field approximation as, for example, in Portengen et al. [26] to obtain the excitonic average \( \Delta = \langle c_i^\dagger f_i \rangle \). The inhomogeneous mean-field theory discussed below allows for a local excitonic order parameter (EOP) and involves diagonalizing the mean-field Hamiltonian matrix followed by the calculation of the EOP and local occupancies from the eigenvalues. The new EOPs and densities are then fed back into the Hamiltonian and the process continues till convergence is reached in all the averages (up to \( 10^{-8}\% \)).

Results and discussions. – To check our numerical procedure we start with the extended FKM \((H = H_0 + H_v)\) in the symmetric case \((E_f = 0)\) and \( n_c = n_f = 0.5 \) without the transverse field. We study the effect of Coulomb interaction on the stability of excitons. The Coulomb interaction \( U \) gave rise to a nonvanishing \( c-f \) coherence \( \langle c_i^\dagger f_i \rangle \neq 0 \) even in the limit of \( V \to 0 \) [25] in the presence of a putative homogeneous ground state under the mean-field approximation. Czycholl [36] showed later that \( \langle c_i^\dagger f_i \rangle \to 0 \) as \( V \to 0 \) if the right ground state is considered (an inhomogeneous CDW order), i.e., there is no spontaneous symmetry breaking, consistent with Eliitzur’s theorem [37] (but not contradictory at \( T = 0 \) though, where Eliitzur’s theorem does not forbid an order). For a small non-zero \( V \) the inhomogeneous (CDW) phase is stable, and there is a coexisting finite EOP. Similar conclusions were then reached for the triangular lattice as well [38]. Our calculation shows there is no finite excitonic average in the \( V \to 0 \) limit as expected on symmetry grounds. As we increase hybridization between \( c \)- and \( f \)-electrons for a fixed Coulomb correlation, we find an enhancement in the excitonic order parameter. Figure 1 reveals that for a finite \( V \), there is a non-vanishing \( \Delta \) that is strongly enhanced as \( U \) increases. The stronger the effect of \( U \), the smaller the value of \( V \); the order parameter is exponentially enhanced as expected. These results are in complete agreement with previous results in a wide parameter regime [36,38–40].

Effect of perpendicular magnetic field. – When a magnetic field is applied, the field couples to the spinless, mobile fermions via canonically conjugate momenta only. We choose the Landau gauge \( \vec{A}(r) = B(0, m a, 0) \), for a uniform magnetic field of magnitude \( B \) perpendicular to the plane of the lattice, and therefore the hopping integral does not change in the \( x \)-direction. Along the \( y \)-direction a “Peierls phase” \( \tau_{ij} = -t \exp(\pm i e / h \int A(r') d\vec{r}') = -t \exp(\pm 2\pi i m \alpha) \) appears. \( \phi = B a^2 \) is the flux per plaquette of a square lattice which is the Aharonov-Bohm (AB) phase around a closed path along the plaquette. We consider only the rational flux, i.e., \( \phi = \frac{p}{q} \phi_0 = \alpha \phi_0 \) with \( p, q \) co-prime integers and the Dirac flux quantum is \( \phi_0 \).

Lattice periodicity is lost along the \( x \)-direction due to the magnetic field. As is customary, the lattice is discretized into \( q \) sublattices in the \( x \)-direction —each site is then indexed by an integer “\( m \).” The Hamiltonian is invariant only for lattice translations in the magnetic translation group [41]. This leads to a unit cell \( q \) times larger than the original one to enfold an integer flux \( p \phi_0 \). Therefore, to accommodate a magnetic flux \( \Phi = \frac{2\pi}{q} \), the magnetic supercell will now be a strip of length \( L \) [35,42,43] (maximum \( L = 36 \) used). We work in the half-filled limit \( n_c + n_f = 1 \), at zero temperature. The spectrum, in the non-interacting limit, shows a self-similar structure, reminiscent of the Hofstadter problem [44,45], in which widths and gaps appear and disappear for different values of the magnetic fluxes chosen.

The inclusion of flux \( \alpha \) per plaquette affects the exciton formation quite strongly. As we see from fig. 2, the value of excitonic average is generally suppressed with the application of the field, though this is highly non-monotonic.

It is clear from fig. 2(a), a prominent dip at \( \alpha = 0.25 \) and a peak at \( \alpha = 1/3 \) can be found for \( V = 0.06 \). There is a large dip at \( \alpha = 0.5 \), signifying the value of applied field
Fig. 3: (Colour online) (a) The variation of excitonic average with hybridization for different $\alpha$ at $U = 0.20$. The black line shows the “enhancement” of exciton average for $\alpha = 1/3$ in the small $V$ limit. (b) The variation of CDW for several magnetic flux $\alpha$ per plaquette. The inset shows the enhancement in $\Delta$ (solid lines) with $V$ and melting of the CDW order parameter (dashed lines) with increasing $V$ at different values of the applied magnetic field.

at which $\Delta$ is a minimum and the trend is the same for all the values of $V$ studied. These undulations become lesser with larger $U$ (fig. 2, inset (a)) as well as larger $V$ (fig. 2, inset (b)). As the Hamiltonian is symmetric with respect to $\phi$ and $1 \pm \phi$, the $\alpha$-dependence of EOP is symmetric with respect to $\alpha = 0.5$ and, expectedly, displays a typical Hofstadter characteristics in the non-interacting limit.

The variation of $\Delta$ with $V$ (fig. 3(a)) more or less follows the same pattern as in the absence of the magnetic field. For a fixed $U$, the excitonic average reduces with increasing magnetic field. Interestingly, it is also possible to get an enhancement in the excitonic average in a region where hybridization and correlation are quite weak (fig. 2(a)) as shown for $\alpha = 1/3$. The ground state of the FKM without hybridization shows long-range CDW order [19,46,47] on a bipartite lattice at half-filling ($n_e = n_f = 0.5$): the $f$-electrons order in a checkerboard pattern. It is known, though, that above a certain value of $V < V_c$, this charge density pattern melts. The inset of fig. 3(b) implies that when the CDW order drops the excitonic average takes over. We found that the critical value $V_c$ at which the CDW melts is the same for all the applied magnetic fields, while the CDW order below $V_c$ strongly depends on the field. In fig. 3(b), where $V$ is kept less than $V_c$, one can see that although the CDW gets extra stability at $\alpha = 1/3$, for other magnetic fluxes considered the magnitude of CDW order decreases. Note that this value $\alpha = 1/3$ also represents the flux where EOP has a pronounced peak (fig. 2) as corroborated in fig. 3(a) (for $V < V_c$). This again shows that CDW and EOP orders are correlated, as for $\alpha = 1/3$, the EOP has a pronounced peak and the strength of CDW has a higher value (black line in fig. 3(b)).

**Effect of $f$-electron hopping.** So far, the $f$-electrons are kept localized without hybridization. We now allow delocalization of the $f$-electrons with a dispersive term $\sum_{\langle ij \rangle} -t_f(f_i^\dagger f_j + \text{h.c.})$ in the Hamiltonian $H_0 + H_\alpha$.

This breaks the local $U(1)$ symmetry of the $f$-electrons even when $H_e$ is absent and hence, clearly, gives rise to a non-zero $\Delta$ without the presence of an explicit hybridization term ($V$) in the Hamiltonian in $D \geq 1$ dimension [27]. This also precludes the formation of a CDW order. We, therefore, examine the effect of $f$-electron hopping on the excitonic averages, and in particular, investigate the effects of different parities of $f$-electron hopping term. The parity of the $\mathbf{f}$-orbital determines the sign of $t_f$. The magnitude as well as sign of $f$-electron hopping integral $t_f$ play important roles in the formation of excitons. The value of excitonic order parameter is found to decrease with increasing $t_f$ due to the enhanced kinetic energy of the $f$-electrons destabilizing the local excitonic order parameter. This happens for both the signs of $t_f$.

A common issue in the study of FKM is the valence transitions, $n_f$ vs. $E_f$ behaviour. In the presence of the local $U(1)$ symmetry, the well-known staircase-like behaviour is observed, giving way to a quantum mixed valence in the presence of $H_e$. The step-like behaviour of the $n_f - E_f$ curve is present even if $t_f$ is finite, as long as $V = 0$. The remarkable feature here is that the sign of $t_f$ determines the behaviour of the steps for $V \neq 0$. In fig. 4, and its insets, different scenarios are presented: $t_f$ positive and negative, with $V = 0$ or finite. The step-like transition in $n_f$ is replaced by a more smooth variation (inset (a) in fig. 4) for $t_f < 0$, $V = 0.05$ and $U = 0.4$. In fig. 4, inset (b), $t_f$ is positive and for both $V = 0$ and $V$ finite, the steps persist ($U$ is kept 0.4 here). This is likely to be the result of the change of curvature of the $f$-band as the parity changes. When $t_f$ is negative, fig. 4, inset (c), there is a drastic difference between $V = 0$ and $V$ finite, the latter gives rise to a smooth variation now unlike the situation with $t_f$ positive. This is a theme repeated throughout as discussed below.

For a fixed value of $U$, as seen from fig. 5, $\Delta$ drops with an increase in $t_f$. Clearly, this is expected as the average $f$-electron occupancy at a site is lower now as compared to the case when it was occupied by a localized $f$-electron.
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Fig. 5: (Colour online) Variation of excitonic order parameter for negative $t_f$ at a fixed $U(=0.1)$ and $\alpha = 0.0$. The insets (a) and (b) show the variation of EOP at (a) $\alpha = 1/4$ and (b) $\alpha = 1/3$, respectively. (restricting it to 1). With negative $t_f$ (i.e., $t_c/t_f < 0$) the EOP increases with $V$ in a regular fashion. The same problem is then studied in the presence of an external magnetic field; which changes the phases of charged particles (see insets (a) and (b) in fig. 5); excitonic condensation is further suppressed with $\alpha$, which now affects the hopping of the $f$-electrons via the Aharonov-Bohm phase as well. If we choose $f$- and $c$-electrons of same parity (sign of $t_f$ and $t_c$ same), we get a very different behaviour in the variation of EOP with $V$. Quite remarkably, as shown in fig. 6 and its insets, plateaus are formed in the excitonic order parameter. This signifies there are valence transitions fig. 6 and its insets, plateaus are formed in the excitonic or-

Fig. 6: (Colour online) Variation of excitonic order parameter for different choices of positive $t_f$ at a fixed $U(=0.1)$ at $\alpha = 1/4$. Inset (a) shows EOP as a function of $V$ for $\alpha = 0.0$ and inset (b) shows $\alpha = 1/3$ for the same $t_f$ values.

$\alpha$ is incorporated (compare inset (a) in fig. 6 with the main figure). This reflects the extra stability and the pinning of the corresponding ground state coming from the commensurability of the magnetic field. Note that the “Peierls phase” appears in the $f$-electron hopping term as well and therefore the relative magnitude and parity of $t_c$ and $t_f$ have strong effect on the commensurability coming from the magnetic field. The magnitude of EOP is suppressed as $\alpha$ increases (interestingly, an exception is found at $\alpha = 1/3$ again, see inset (b) in fig. 6, possibly due to commensurability effect). At $\alpha = 1/2$ the value of EOP for different $t_f$ almost vanishes (not shown).

It is fascinating to observe the effect of both $U$ and $t_f$ on the exciton condensation (fig. 7). As we saw earlier ($t_f = 0$ case), the FKM-type $U$ between $c$- and $f$-electrons supports a bound state between the electron-hole of $c$- and $f$-bands. This trend is maintained even in the presence of finite $t_f$, provided the sign of $t_f$ is negative. The opposite phenomena occur for $f$-electrons with same parity, i.e. the signs of $t_c$ and $t_f$ are the same. In this case, the EOP drops as the correlation between $c$- and $f$-electrons increases. Moreover, the step-like behaviour in the $\Delta$-$t_f$ curve occurs only when there is a finite $U$. As $U$ changes from zero, so does the chemical potential for $n_f$ and the steps appear.

Summary and conclusion. – In this paper we have provided numerical results for an exciton in a transverse magnetic field in a 2D correlated lattice model, i.e., the extended Falicov-Kimball model. We observe that there is competition between applied magnetic field $\alpha$ and $f$-electron hopping integrals apart from Coulomb correlation $U$ and $c$-$f$ hybridization $V$. The interband Coulomb interaction exponentially enhances the excitonic average. The magnetic field has a localizing effect on the mobility of the participating particles, affecting the exciton coherence; a drop in the value of exciton order parameter with both commensurate (except $\alpha = 1/3$, at low $V$)
and incommensurate flux is observed. The flux sensitivity of the exciton arises as the charge couples to the vector potential and the coupling has opposite signs for an electron and a hole. The magnetic field suppresses the electron-hole binding as the participating particles and holes are affected differently in a close ring of applied gauge field. Although, in the small $V$ limit, the mobile fermions behave like quasi-free electrons and the Hofstadter physics dominates. In the high magnetic-field regime, the $c$-$h$ Coulomb attraction forces the excitons to low-momentum states reducing the ground-state radius for exciton and increasing the binding energy; hence the increase of $T_c$, as mentioned by Lozovik et al. [5], corroborates with the enhanced order parameter. There is of course a strong commensurability effect in this case, though it favours the formation of an exciton at the special flux $\alpha = 1/3$ per plaquette, thereby opening the possibility of tuning ferro-electricity via magnetic field. With non-zero $t_f$ values, further reduction in the magnitude of excitonic average is observed, irrespective of the sign of $t_f$. There is also an interesting plateau structure of EOP for same-parity $c$- and $f$-electrons. In the present study, we work only with rational magnetic fields; as an irrational field is known to induce, in the non-interacting limit, a Cantor set structure of the spectrum. To achieve fractional quantum Hall effect, one needs to consider small magnetic flux (the magnetic length must be much larger than the lattice periodicity). In that limit, one would expect to see fractional quantum Hall effects at right fillings in an incompressible fluid. However, the effect of $f$-electrons on the possible FQHE states is not clear. In the $V \to 0$ limit, they act as annealed disorder and often lead to charge ordering while any finite $V$ (or $f$-electron hopping) will bring in their dynamics and the effects of magnetic field thereof. This is an extremely interesting problem, not amenable to mean-field treatments and beyond the scope of our present study.

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