Ground states of the Falicov-Kimball model with hybridization

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We obtain exact ground states for the spinless Falicov-Kimball model with hybridization in one dimension ($d=1$). Ground-state energies and their corresponding wave functions are given for a restricted region of the parameter space in the limit of strong interactions for the half-filling condition. We find that the ground states possess a diagonal long range order and constitute a condensation of excitons in a zero momentum state. On the other hand, the correlation function $A=\langle d_i^\dagger f_i \rangle$ is purely imaginary, indicating that the ground state does not exhibit spontaneous ferroelectricity.

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I. INTRODUCTION

There are several systems, like rare earth and transition metal compounds, whose physical properties are determined by the presence of almost localized electrons interacting with conduction electrons. As external parameters are modified, some of these materials can exhibit valence, as well as metal-insulator, transitions of first or second order. The theoretical descriptions of such systems are usually based on the periodic Anderson model or the Falicov-Kimball (FK) model. Despite their apparent simplicity, these models are very complicated and few rigorous results are known. The FK model without hybridization has been solved exactly by Brandt and Mielsch[1] in the limit of $d\rightarrow\infty$. However, no general solution for the FK model with hybridization has been given.

The question of whether or not the FK model can have discontinuous transitions in the occupation number is very important to determine whether it is an adequate model for describing the cited compounds. Different answers can be found in the literature since the results are very sensitive to the approximations used.[2] The FK model has also been studied in connection with the formation of excitonic states. Since a d-f exciton carries a dipole moment, such excitonic states have been considered as ferroelectric states[3,4] with a polarization equal to the excitonic correlation $A=\langle d_i^\dagger f_i \rangle$. A related metal-insulator transition driven by an excitonic instability was proposed in the past by various authors.[5] It is worth mentioning that these older works on the FK model were based on mean-field decouplings of the interaction, in which case the formalism becomes very similar to that of the BCS theory of superconductivity. Portengen *et al.*[3] studied the FK model within this approach. They obtained a nonzero value of the excitonic expectation value A in the limit of zero hybridization, which was interpreted as the emergence of a spontaneous ferroelectric polarization. However, later studies performed by Czycholl in the limit $d\rightarrow\infty$,[6] Farkasovsky[7,8] in finite systems for $d=1$, and Sarasua and Continentino[9] in $d=1$ infinite systems did not confirm this result and yielded a vanishing value of A in the limit of zero hybridization. On the other hand, Zlatic *et al*[10] calculated the spontaneous polarization susceptibility in the FK model and found that it diverges in the limit of zero temperature, indicating a possible non-zero polarization in the ground state. Recently, Batista[11] found that a FK model with nonlocalized f electrons can have a nonvanishing parameter A and a ferroelectric ground-state. In these works[10,11] the limit $V\rightarrow 0$ was taken, but, in real systems the overlap between the f and d orbitals is not negligible and a more realistic model should include the presence of hybridization.[12]

As a consequence of the symmetry of the orbitals f and d, the hybridization must satisfy the relation[3,4]

$$V_{-k}=-V_k=V_k^* . \tag{1}$$

This condition excludes the presence of on-site hybridization since this is a constant in momentum space. On-site hybridization can only occur by the influence of an external agent, such as an electromagnetic field. In this case, a nonzero value of A must be interpreted as non spontaneous, but induced by the external field. On the other hand, if the mixing interaction satisfies Eq. (1), which implies that it is imaginary, the value of the excitonic correlation can be considered as truly spontaneous. Thus, in order to be most conclusive about the problem of ferroelectricity, we must study a model with an hybridization of this type.

Recently, Giesekus and co-workers[13,14] presented a method to obtain exact solutions for the Anderson and Hubbard models, for special values of the parameters, in the limit case $U\rightarrow\infty$. The present authors used this technique to study the extended Anderson model including the Falicov-Kimball interaction.[16,17] In this work we study an extended Falicov-Kimball model with hybridization in the limit of strong interactions at $d=1$ using a similar approach. We obtain the exact ground-state energy and a corresponding wave function for the half-filling condition in the limit of infinite interaction. While in previous work[9] we considered the cases of quarter filling and three-quarter filling, here we concentrate on the half-filling case. It is well known that band filling affects the behavior of strongly correlated electrons very much. In fact, the half-filling condition assumed here is the one appropriate to study the excitonic insulator.[5] Also, in Ref. 9 hybridization was taken as a real parameter, such that, we could only consider the case in which there is an intrasite hybridization in the system. In the present study we assume that hybridization occurs only between different sites and is purely imaginary. It has a structure in k space given by Eq. (1) and consequently, it is compatible with the symmetries of the f and d orbitals. A more elaborate model could consider a multiband system, in which case, when projecting the multiband system into a two-band system this could give rise to an hybridization not satisfying Eq. (1).[15] However, we shall restrict the present study to the case of bare bands. We obtain an analytical expression for the correlation parameter $\langle d^\dagger f\rangle$ in the ground state and analyze the possibility of a spontaneous excitonic correlation. It is worth emphasizing that it is always of great interest to obtain exact results for such a relevant physical model as they can be used as guides for different approximate schemes.

II. HAMILTONIAN MODEL

We consider an extended version of the spinless Falicov-Kimball model in which an intersite hybridization is included. The Hamiltonian of the model is

$$H=t_f\sum_{\langle ij\rangle} f_i^\dagger f_j+t_d\sum_{\langle ij\rangle} d_i^\dagger d_j+G\sum_i n_i^f n_i^d+E_f\sum_i n_i^f + \sum_i [V(f_i^\dagger d_{i+1}+d_i^\dagger f_{i+1})+\mathrm{H.c.}], \tag{2}$$

where $f_i^\dagger$ (f_i) and $d_i^\dagger$ (d_i) are creation (annihilation) operators for f and d bands respectively, E_f is the site energy of f electrons, G is the Coulomb interaction between f and d electrons, and the last term represents the hybridization. Here the hoppings t_f, and t_d are real numbers, with $t_d>0$ for simplicity, and the hybridization V is purely imaginary. This type of hybridization follows from symmetry considerations as stated before.[3,4] The corresponding dispersion relations for each band and the hybridization in k space are given by $\epsilon_k^d=-2t_d\cos(k)$, $\epsilon_k^f=E_f-2t_f\cos(k)$, and $V_k=2V\sin(k)$. We define the operator

$$a_i^\dagger=\alpha d_i^\dagger+\beta d_{i+1}^\dagger+\gamma f_i^\dagger+\delta f_{i+1}^\dagger . \tag{3}$$

Hamiltonian (2) can be expressed in terms of the operators a_i and the occupation numbers n_f and n_d in the form

$$H=\sum_i a_i a_i^\dagger+2t_d\sum_i n_i^d+(2t_d|\eta|^2+E_f)\sum_i n_i^f+G\sum_i n_i^f n_i^d -2t_d(1+|\eta|^2)L \tag{4}$$

if the following condition is fulfilled:

$$t_f t_d=V^2, \tag{5}$$

with $|\alpha|^2=t_d$, $|\alpha|^2-|\beta|^2=0$, $\gamma=(t_f/V)\alpha$, $\delta=-(t_f/V)\beta$, $\eta=t_f/V$, and L the number of lattice sites.[14,16,17] In order to obtain an expression in terms of the occupation number $n=n_f+n_d$ the following condition is imposed:

$$E_f=2(1-|\eta|^2)t_d . \tag{6}$$

Thus, Hamiltonian (2) can be rewritten in the form

$$H=P+2t_d\sum_i (n_i^d+n_i^f)+P_G-2t_d(1+|\eta|^2)L, \tag{7}$$

where $P=\Sigma_i a_i a_i^\dagger$, and $P_G=G\Sigma_i n_i^f n_i^d$. In this form, Hamiltonian (2) is a sum of a constant, the positive semi-definite operator P and the interaction term which is also positive semi-definite. Thus, a lower bound to the ground-state energy ϵ_H can be obtained,

$$\epsilon_l=2\langle n\rangle t_d L-2t_d(1+|\eta|^2)L, \tag{8}$$

where L is the lattice size. Notice that for conditions (5) and (6) and in the absence of interactions ($G=0$), the energy excitations are split in two bands separated by a direct gap $\Delta=4|t_f|$. Then, in the noninteracting case, the system has a band insulating behavior.

III. SOLUTION

We now proceed to construct an exact ground-state solution for the strong coupling limit $G=\infty$. This condition prohibits the presence of a d electron in a site occupied by a f-electron. To construct the states, we can use a projector P_P which removes all the states containing local f-d electron pairs. Alternatively, this constraint can be imposed to the creation operators by

$$f_i^\dagger d_i^\dagger=0, \tag{9}$$

such that the projector is implicitly implemented. Consider the wave function given by

$$|\psi\rangle=\prod_i a_i^\dagger|0\rangle, \tag{10}$$

where $|0\rangle$ is the empty state, and the creation operators must obey restriction (9). The variational principle asserts that $\langle\psi|H|\psi\rangle/\langle\psi|\psi\rangle$ is an upper bound to the ground-state energy.[18] It can easily be checked that the property $(a_i^\dagger)^2=0$ holds and then, $P|\psi\rangle=0$. Due to the restrictions imposed to the $f^\dagger$ and $d^\dagger$ operators, the operator P_G vanishes. Thus, $|\psi\rangle$ is an eigenfunction of H with energy $\epsilon_s=\epsilon_l$ given by Eq. (8). Furthermore since the lower and upper bounds coincide and $|\psi\rangle$ contains one electron per site ($\langle n\rangle=1$), the ground-state energy is

$$\epsilon_H=2t_d L-2t_d(1+|\eta|^2)L. \tag{11}$$

It is convenient to express $|\psi\rangle$ in the more simplified form

$$|\psi\rangle=\left[C_1\prod_i (d_i^\dagger-\eta f_i^\dagger)+C_2\prod_i (d_i^\dagger+\eta f_i^\dagger)\right]|0\rangle, \tag{12}$$

where C_1 and C_2 are arbitrary constants. In order to show how this simplification occurs, we define new operators

$$D_i^\dagger=\alpha(d_i^\dagger+\eta f_i^\dagger),\quad F_i^\dagger=\beta(d_i^\dagger-\eta f_i^\dagger),$$

so that the operators $a_i^\dagger$ can be expressed as $a_i^\dagger=D_i^\dagger+F_j^\dagger$. Notice that constraint (9) implies that $D_i^\dagger F_i^\dagger=0$. Now we consider the products of operators $a_i^\dagger a_j^\dagger$ at two neighboring sites appearing in the expression of $|\psi\rangle$, Eq. (10). This is given by

$$a_i^\dagger a_j^\dagger=D_i^\dagger D_j^\dagger+D_i^\dagger F_l^\dagger+F_j^\dagger D_j^\dagger+F_j^\dagger F_l^\dagger ,$$

where $j=i+1$ and $l=i+2$. In the above product the third term clearly vanishes. Since the second does not create any particle at the site j and the infinite interaction prohibits the presence of two electrons in any of the other sites, it gives rise to states with a total particle number less than L. Since we are restricted to the case in which the number of particles is strictly L, this term must be excluded. Then, only the terms $D_i^\dagger D_j^\dagger$ and $F_j^\dagger F_l^\dagger$ remain and the wave function $|\psi\rangle$ adopts the form given by Eq. (12). It is easy to check directly from Eq. (12), using relations (3) and (9), that $a_i^\dagger|\psi\rangle=0$, which in turn implies that $P|\psi\rangle=0$. We also see from Eq. (12) that $|\psi\rangle$ contains one electron in each site and represents an insulating state, since $\langle\psi|d_i^\dagger d_j|\psi\rangle=\langle\psi|f_i^\dagger f_j|\psi\rangle=\langle\psi|d_i^\dagger f_j|\psi\rangle=0$. Notice that the intersite hybridization used in Eq. (2) satisfies the symmetry constraint of Eq. (1) and is the same as that adopted in Ref. 3. Thus it is particularly interesting to compare our results with those obtained in this reference. If the value of A is calculated using the expression for $|\psi\rangle$, Eq. (12), we get

$$A=\frac{\langle\psi|d_i^\dagger f_i|\psi\rangle}{\langle\psi|\psi\rangle}=\frac{(|C_1|^2-|C_2|^2)(1+|\eta|^2)^{L-1}\eta}{(|C_1|^2+|C_2|^2)[(1+|\eta|^2)^L+(1-|\eta|^2)^L]}-\frac{(C_1^*C_2+C_2^*C_1)(1-|\eta|^2)^{L-1}\eta}{(|C_1|^2+|C_2|^2)[(1+|\eta|^2)^L+(1-|\eta|^2)^L]}. \tag{13}$$

In the thermodynamic limit $L\to\infty$, this gives

$$A=\frac{(|C_1|^2-|C_2|^2)\eta}{(|C_1|^2+|C_2|^2)(1+|\eta|^2)}, \tag{14}$$

such that our solution admits states with broken symmetry. In fact, if we first take $C_2=0$ and next $C_1=0$ in Eq. (12), we obtain, respectively, the two ground states

$$|\psi^{(+)}\rangle=\prod_i (d_i^\dagger+\eta f_i^\dagger)|0\rangle,$$

$$|\psi^{(-)}\rangle=\prod_i (d_i^\dagger-\eta f_i^\dagger)|0\rangle, \tag{15}$$

which have nonzero values of A given by

$$A=\pm\frac{\eta}{(1+|\eta|^2)}$$

for $|\psi(\pm)\rangle$. Notice that these values of A are purely imaginary since η is purely imaginary. A remarkable fact is that $|\psi^{(+)}\rangle$ and $|\psi^{(-)}\rangle$ are states that break the symmetry of Hamiltonian (2). Since the hybridization satisfies Eq. (1), the Hamiltonian [Eq. (2)] remains invariant under a change in the sign of V together with an inversion in the sense of the x axis. A change of the sign of V, which implies a change of the sign of η, produces the transformations $|\psi^{(+)}\rangle\to|\psi^{(-)}\rangle$, $|\psi^{(-)}\rangle\to|\psi^{(+)}\rangle$. This shows that these states do not have the symmetry of the Hamiltonian.

Thus, the FK model with hybridization, has ground states with broken symmetry in which the values of A are nonvanishing. However, following Ref. 3, the ferroelectric polarization is given by *the real part of A*. Since the values of A obtained from $|\psi^{(\pm)}\rangle$ are purely imaginary, these states are not ferroelectric. Although these results are in contradiction with those obtained in Ref. 3, it is remarkable that the structure of the above ground states is very close to that obtained in mean-field approximation.[3–5] On the other hand, the fact that the results of Refs. 6–8 yield $A=0$, in the case without hybridization, raises doubts about the reliability of the mean-field studies of excitonic insulators where this state is characterized by a nonzero value of A. Then, it is interesting to compare the properties of the ground states $|\psi^{(\pm)}\rangle$ with those obtained in the mean-field studies.

IV. EXCITONIC STATES

We note that the interaction G promotes the formation of f-d electron-hole pairs. Since the states with local f-d electrons pairs have been projected out, the average number of f-d-electron-hole pairs is larger in $|\psi\rangle$ than in the noninteracting state. The presence of the excitons can be put in a more manifest form, noting that $|\psi\rangle$ can be expressed as

$$|\psi\rangle=c_1\prod_i (u-vd_i^\dagger f_i)|\varnothing\rangle+c_2\prod_i (u+vd_i^\dagger f_i)]|\varnothing\rangle, \tag{16}$$

where $|\varnothing\rangle=\Pi_i f_i^\dagger|0\rangle$ is the state with a totally filled f band, without electrons d, and with $u/v=\eta$. This expression shows that an f hole at site i and an electron in the same site are either both present or both absent. Notice that Eq. (16) for $|\psi\rangle$ is very similar to the mean-field ground state obtained in Ref. 5, which is given by

$$|\psi_{MF}\rangle=\prod_k (u_k-v_k d_k^\dagger f_k)|\varnothing\rangle. \tag{17}$$

Despite the close analogy with the BCS wave function, $|\psi_{MF}\rangle$ does not possess superconducting properties.[5] While in the BCS ground state there is off-diagonal long range order,[19] which implies the Meissner effect, the excitonic mean field ground state [Eq. (17)] possesses diagonal long range order (DLRO). This DLRO is characterized by a nonvanishing value of $\langle\psi_{MF}|d_i^\dagger f_j^\dagger d_j f_i|\psi_{MF}\rangle$ in the limit case $|r_i-r_j|\to\infty$, where r_i, and r_j denote the positions of the sites i and j.[5] We shall now investigate if $|\psi\rangle$ has this property. Using the expression for $|\psi\rangle$, Eq. (16), we obtain that

$$\lim_{|r_i-r_j|\to\infty}\frac{\langle\psi|d_i^\dagger f_j^\dagger d_j f_i|\psi\rangle}{\langle\psi|\psi\rangle}=\frac{|\eta|^2}{(1+|\eta|^2)^2}. \tag{18}$$

Then in the limit $L\to\infty$, the exact solution has the same type of long range order that the mean-field solution $|\psi_{MF}\rangle$ has. This property is independent of the values of c_1 and c_2, which means that it is not related to the broken symmetry discussed before. Another feature that is usually attributed to excitonic insulators is that they can be interpreted as a condensation of excitons. Notice that Eq. (18) can be expressed as

$$\lim_{|r_i - r_j|\to\infty} \frac{\langle \psi | b_i^\dagger b_j | \psi \rangle}{\langle \psi | \psi \rangle} = \frac{|\eta|^2}{(1+|\eta|^2)^2}, \tag{19}$$

where $b_i^\dagger = d_i^\dagger f_i$ creates an exciton at site i and b_j destroys an exciton at site j. This condition is formally the same as that for Bose condensation of excitons, in turn given by $\lim_{|r_i - r_j|\to\infty} \langle \psi | b_i^\dagger b_j | \psi \rangle = \alpha_c n \exp[i\mathbf{k}\cdot(\mathbf{r_i} - \mathbf{r_j})]$,[19] where $\mathbf{k}$ is the wave vector characterizing the state at which the macroscopic occupation occurs and α_c is the condensate fraction. Since the right hand side of Eq. (19) is constant and $n=1$, the condensation takes place in the zero-momentum state with $\alpha_c = |\eta|^2/(1+|\eta|^2)^2$. This condensate fraction has a maximum value of $\alpha_c = 0.25$ for $|\eta| = 1$, which due to the restrictions imposed by Eqs. (6) and (5) corresponds to the case $E_f = 0$ and $t_f = -t_d$, i.e., the symmetric case with the two bands with the same bandwidth.

Another subject that has been extensively investigated in the FK model is the emergence of phase separation (PS). PS in the FK model has been rigorously demonstrated for dimension $d=1$.[20] We point out that in the ground state $|\psi\rangle$ there is no phase separation nor charge ordering for any value of η. Taking into account the differences in the models, this can be caused by the presence of a finite hopping t_f or by the influence of hybridization. The suppression of PS for very large values of V is expected since in this limit the proper distinction between f and d electrons becomes meaningless. Although we cannot be conclusive due to the restrictions in the parameters, conditions (5) and (6) certainly provide a region of parameter space in which phase separation does not take place.

V. CONCLUSIONS

We have obtained the exact ground-state energy and the corresponding wave function for the extended Falicov-Kimball model in $d=1$, for the half-filling condition $\langle n \rangle = 1$ and in the limit of strong correlations. We have included a hopping term for the f electrons and an intersite imaginary hybridization. We find that the ground state is a linear combination of two degenerate symmetry broken states $|\psi^{(\pm)}\rangle$, each of them also being a ground state. The states $|\psi^{(\pm)}\rangle$ are characterized by a nonvanishing value of the A expectation value. However, the value of A is purely imaginary, so that the ground states do not exhibit spontaneous ferroelectricity.

The ground states we obtain are consistent with the excitonic states predicted in mean-field studies. They possess the same structure of the mean-field ground state, i.e., they are insulating and have the same type of long range order that can be interpreted as a Bose condensation of excitons. An analytical expression for the condensate fraction was obtained. These ground states can be viewed as a realization of the excitonic insulator in the Falicov-Kimball model.

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[1] U. Brandt and C. Mielsch, Z. Phys. B: Condens. Matter **75**, 365 (1989).
[2] E. Baeck and G. Czycholl, Solid State Commun. **43**, 89 (1982).
[3] T. Portengen, T. Ostreich, and L.J. Sham, Phys. Rev. Lett. **76**, 3384 (1996).
[4] J. Duan, D.P. Arovas, and L.J. Sham, Phys. Rev. Lett. **79**, 2097 (1997).
[5] D. Jérome, T.M. Rice, and W. Kohn, Phys. Rev. **158**, 462 (1967); B.I. Halperin and T.M. Rice, Rev. Mod. Phys. **40**, 755 (1968), and references therein.
[6] G. Czycholl, Phys. Rev. B **59**, 2642 (1999).
[7] P. Farkasovsky, Phys. Rev. B **60**, 10776 (1999).
[8] P. Farkasovsky, Phys. Rev. B **65**, 081102 (2002).
[9] L.G. Sarasua and M.A. Continentino, Phys. Rev. B **65**, 233107 (2002).
[10] V. Zlatic, J.K. Freericks, R. Lemanski, and G. Czycholl, Philos. Mag. B **81**, 1443 (2001).
[11] C.D. Batista, Phys. Rev. Lett. **89**, 166403 (2002).
[12] A. Giesekus and L.M. Falicov, Phys. Rev. B **44**, 10449 (1991).
[13] U. Brand and A. Giesekus, Phys. Rev. Lett. **68**, 2648 (1992).
[14] R. Strack, Phys. Rev. Lett. **70**, 833 (1993).
[15] M. Kollar, R. Strack, and D. Vollhard, Phys. Rev. B **53**, 9225 (1996).
[16] I. Orlik and Z. Gulacsi, Philos. Mag. B **81**, 1587 (2001); J. Phys. A **34**, L359 (2001).
[17] P. Gurin and Z. Gulacsi, Phys. Rev. B **64**, 045118 (2001).
[18] A. Messiah, *Quantum Mechanics* (North-Holland, Amsterdam, 1986).
[19] C.N. Yang, Rev. Mod. Phys. **34**, 694 (1962).
[20] P. Lemberger, J. Phys. A **25**, 715 (1992).