

# Ground-states of the three-dimensional Falicov-Kimball model

P. Farkašovský<sup>a</sup>, H. Čenčariková, and N. Tomašovičová

Institute of Experimental Physics, Slovak Academy of Sciences Watsonova 47, 043 53 Košice, Slovakia

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**Abstract.** A systematic study of ground-state properties of the three-dimensional Falicov-Kimball model is performed by a well-controlled numerical method. The results obtained are used to categorize the ground-state configurations according to common features for weak, intermediate and strong interactions. It is shown that only a few configuration types form the basic structure of the phase diagram. In particular, the largest regions of stability correspond to phase segregated configurations, striped configurations and configurations in which electrons are distributed in diagonal planes with incomplete chessboard structure. Near half-filling, mixtures of two phases with complete and incomplete chessboard structure are determined. The relevance of these results for a description of real materials is discussed.

**PACS.** 75.10.Lp Band and itinerant models – 71.27.+a Strongly correlated electron systems; heavy fermions – 71.28.+d Narrow-band systems; intermediate-valence solids – 71.30.+h Metal-insulator transitions and other electronic transitions

## 1 Introduction

The Falicov-Kimball model (FKM) has become, since its introduction [1] in 1969, one of the most popular examples of a system of interacting electrons with short-range interactions. It has been used in the literature to study a great variety of many-body effects in rare-earth compounds, of which metal-insulator transitions, mixed-valence phenomena, and charge-density waves are the most common examples [2]. The model is based on the coexistence of two different types of electronic states in a given material: localized, highly correlated ionic-like states and extended, uncorrelated, Bloch-like states. It is generally accepted that the above mentioned cooperative phenomena result from a change in the occupation numbers of these electronic states, which remain themselves basically unchanged in their character. Taking into account only the intra-atomic Coulomb interaction between the two types of states, the Hamiltonian of the spinless FKM can be written as the sum of three terms:

$$H = \sum_{ij} t_{ij} d_i^{\dagger} d_j + U \sum_i f_i^{\dagger} f_i d_i^{\dagger} d_i + E_f \sum_i f_i^{\dagger} f_i, \quad (1)$$

where  $f_i^{\dagger}$ ,  $f_i$  are the creation and annihilation operators for an electron in the localized state at lattice site  $i$  with binding energy  $E_f$  and  $d_i^{\dagger}$ ,  $d_i$  are the creation and annihilation operators of the itinerant spinless electrons in the  $d$ -band Wannier state at site  $i$ .

The first term of (1) is the kinetic energy corresponding to quantum-mechanical hopping of the itinerant  $d$  electrons between sites  $i$  and  $j$ . These intersite hopping transitions are described by the matrix elements  $t_{ij}$ , which are  $-t$  if  $i$  and  $j$  are the nearest neighbours and zero otherwise (in the following all parameters are measured in units of  $t$ ). The second term represents the on-site Coulomb interaction between the  $d$ -band electrons with density  $n_d = N_d/L = \frac{1}{L} \sum_i d_i^{\dagger} d_i$  and the localized  $f$  electrons with density  $n_f = N_f/L = \frac{1}{L} \sum_i f_i^{\dagger} f_i$ , where  $L$  is the number of lattice sites. The third term stands for the localized  $f$  electrons whose sharp energy level is  $E_f$ .

Since in this spinless version of the FKM without hybridization the  $f$ -electron occupation number  $f_i^{\dagger} f_i$  of each site  $i$  commutes with the Hamiltonian (1), the  $f$ -electron occupation number is a good quantum number, taking only two values:  $w_i = 1$  or  $0$ , according to whether or not the site  $i$  is occupied by the localized  $f$  electron.

Now the Hamiltonian (1) can be written as

$$H = \sum_{ij} h_{ij} d_i^{\dagger} d_j + E_f \sum_i w_i, \quad (2)$$

where  $h_{ij}(w) = t_{ij} + U w_i \delta_{ij}$ .

Thus for a given  $f$ -electron configuration  $w = \{w_1, w_2 \dots w_L\}$  defined on the three-dimensional lattice with periodic boundary conditions, the Hamiltonian (2) is the second-quantized version of the single-particle Hamiltonian  $h(w) = T + UW$ , so the investigation of the model (2) is reduced to the investigation of the spectrum of  $h$  for different configurations of  $f$  electrons.

<sup>a</sup> e-mail: farky@saske.sk

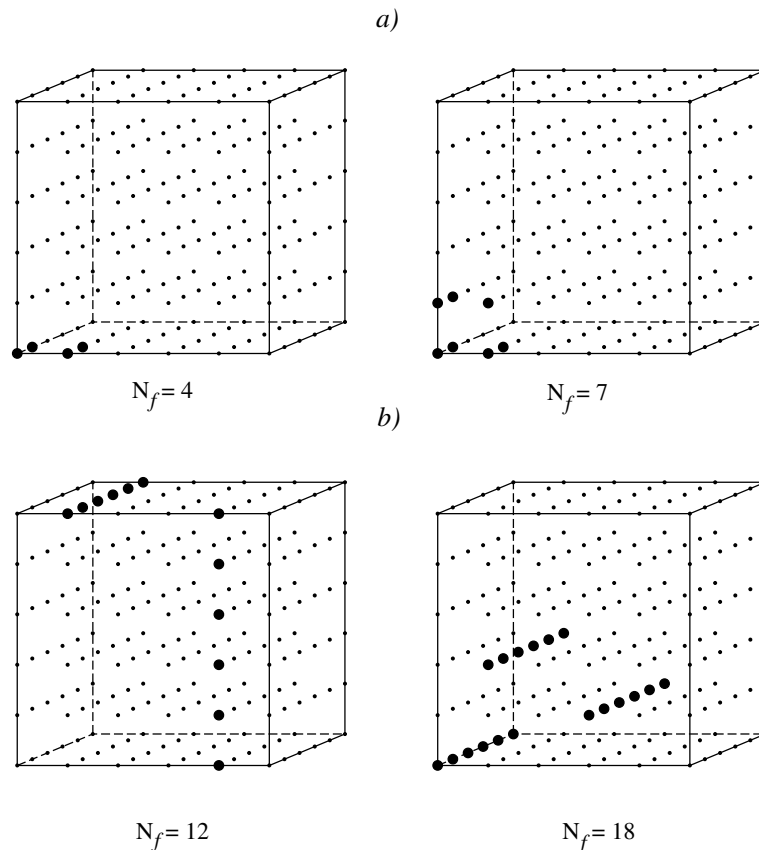
Despite its relative simplicity and an impressive research activity in the past, the properties of this model remained unclear for a long time. The crucial break in this direction has been done recently by exact analytical [3–5] and numerical [6, 7] calculations. These calculations showed that the spinless FKM can describe (at least qualitatively) such important phenomena observed experimentally in some rare-earth and transition metal compounds like the discontinuous valence and metal insulator transitions, phase separation, charge ordering, stripes formation, etc. In addition, it was found [8] that at non-zero temperatures the model is able to provide the qualitative explanation for the anomalous large values of the specific heat coefficient and for the extremely large changes of the electrical conductivity found in some intermediate valence compounds (e.g., in  $\text{SmB}_6$ ). These results indicate that the spinless FKM, in spite of its simplicity, could be a convenient microscopic model for a description of ground-state, thermodynamic and transport properties of real materials. However, real materials are usually three dimensional while the most of above mentioned results have been obtained for the limiting cases of  $D = 1$ ,  $D = 2$  and  $D = \infty$ . Thus one can ask if these results, or at least some of them hold also in three dimensions. This is the question that we would like to answer in this paper. Here we focus our attention on the ground-state properties of model. The special attention is devoted to examine the three dimensional analogs of phase segregation, charge ordering, stripes formation and metal-insulator transitions observed in  $D = 1$  and  $D = 2$ . From this point of view the paper represents the first attempt to describe systematically the ground-state properties of the FKM in three dimensions. To attain this goal we use a well-controlled numerical method that we have elaborated recently [9]. The method is based on the simple modification of the exact diagonalization method on finite clusters and consists of following steps. (i) Chose a trial configuration  $w = \{w_1, w_2 \dots w_L\}$ . (ii) Having  $w$ ,  $U$  and  $E_f$  fixed, find all eigenvalues  $\lambda_k$  of  $h(w) = T + UW$ . (iii) For a given  $N_f = \sum_i w_i$  determine the ground-state energy  $E(w) = \sum_{k=1}^{L-N_f} \lambda_k + E_f N_f$  of a particular  $f$ -electron configuration  $w$  by filling in the lowest  $N_d = L - N_f$  one-electron levels (here we consider only the case  $N_f + N_d = L$ , which is the point of the special interest for valence and metal-insulator transitions caused by promotion of electrons from localized  $f$  orbitals ( $f^n \rightarrow f^{n-1}$ ) to the conduction band states). (iv) Generate a new configuration  $w'$  by moving a randomly chosen electron to a new position which is chosen also as random. (v) Calculate the ground-state energy  $E(w')$ . If  $E(w') < E(w)$  the new configuration is accepted, otherwise  $w'$  is rejected. Then the steps (ii)-(v) are repeated until the convergence (for given  $U$  and  $E_f$ ) is reached. Of course, one can move instead of one electron (in step (iv)) simultaneously two or more electrons, thereby the convergence of method is improved. Indeed, tests that we have performed for a wide range of the model parameters showed that the latter implementation of the method, in which  $1 < p < p_{max}$  electrons ( $p$  should be chosen at random) are moved to new positions overcomes better the

local minima of the ground state energy. In this paper we perform calculations with  $p_{max} = N_f$ . The main advantage of this implementation is that in any iteration step the system has a chance to lower its energy (even if it is in a local minimum), thereby the problem of local minima is strongly reduced (in principle, the method becomes exact if the number of iteration steps goes to infinity). On the other hand a disadvantage of this selection is that the method converges slower than for  $p_{max} = 2$  and  $p_{max} = 3$ . To speed up the convergence of the method (for  $p_{max} = N_f$ ) and still to hold its advantage we generate instead the random number  $p$  (in step (iv)) the pseudo-random number  $p$  that probability of choosing decreases (according to the power law) with increasing  $p$ . Such a modification improves considerably the convergence of the method. Repeating this procedure for different values of  $E_f$  and  $U$  one can immediately study the dependence of the  $f$ -electron occupation number  $N_f = \sum_i w_i^{min}$  on the  $f$ -level position  $E_f$  (valence transitions) or the phase diagram of the model in the  $n_f - U$  plane. This method was first used in our recent paper [9] to study the ground-state properties of the one and two-dimensional FKM. It was found that for small and intermediate clusters, where the exact numerical solution is possible ( $L \sim 30$ ), the method is able to reproduce exactly the ground states of the spinless FKM, even after relative small number of iterations (typically 10000 per site).

## 2 Results and discussion

To examine ground-state properties of the spinless FKM in three dimensions we have performed an exhaustive numerical study of the model for weak ( $U = 1$ ), intermediate ( $U = 2$ ) and strong ( $U = 8$ ) interactions. For each selected value of  $U$  and  $N_f$  ( $N_f = 0, 1, \dots, L$ ) the ground-state configuration  $w^{min}$  is determined by the above described method (we remember that the total filling is fixed at 1). To reveal the finite-size effects numerical calculations were done on two different clusters of  $4 \times 4 \times 4$  and  $6 \times 6 \times 6$  sites. A direct comparison of numerical results obtained on  $4 \times 4 \times 4$  and  $6 \times 6 \times 6$  clusters showed that the ground-state configurations fall into several different categories which stability regions are practically independent of  $L$ . Let us start a discussion of our results with a description of these configuration types for different values of  $U$  and  $N_f$  (in the remainder of the paper the values of  $N_f$  always correspond to  $6 \times 6 \times 6$  cluster).

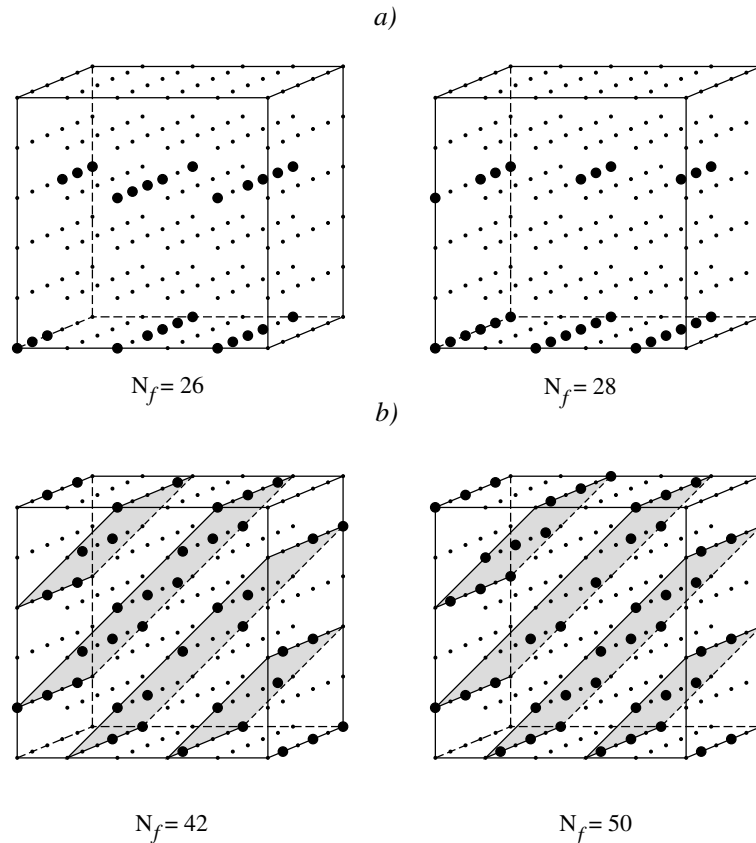
The largest number of configuration types is observed in the weak-coupling limit. Going with  $N_f$  from zero to half-filling ( $N_f = L/2$ ) we have observed the following configuration types for  $U = 1$ . At low  $f$ -electron concentrations the ground-states are the phase segregated configurations ( $f$ -electrons clump together while remaining part of lattice is free of  $f$ -electrons) listed in Figure 1a for two selected values of  $N_f$ . Since the ground-states corresponding to the segregated configurations are metallic [10] we arrive at an important conclusion, and namely, that the metallic domain that exists in the one and two dimensional FKM persists also in three dimensions. In the



**Fig. 1.** Typical examples of phase segregated (a) and striped (b) configurations obtained for  $U = 1$  and  $L = 6 \times 6 \times 6$ . Large dots: occupied sites; small dots: vacant sites.

one dimensional case the region of stability of this metallic domain was restricted to low  $f$ -electron concentrations  $n_f < 1/4$  and small Coulomb interactions  $U \leq 1$  [9,10]. The numerical calculations performed in two dimensions revealed [9] that with increasing dimension the stability region of this metallic domain shifts to higher values of  $U$  ( $U \sim 3$ ). From this point of view it is interesting to examine if this trend holds also for three dimensions. To verify this conjecture we have determined the ground-state configurations for increasing  $U$  at low  $f$ -electron concentrations on  $4 \times 4 \times 4$ ,  $6 \times 6 \times 6$  and  $8 \times 8 \times 8$  clusters. We have found that the metallic region in  $D = 3$  extends up to  $U \sim 5$ , what confirms the trend conjectured from two dimensional calculations (in addition, in accordance with two-dimensional results we have found that the critical value of  $U_c$  decreases with increasing  $n_f$ ). It should be noted that this result is crucial for description of insulator-metal transitions in real materials (like rare-earth and transition metal compounds). In these materials the values of the interaction constant  $U$  are much larger than the values of hopping integrals  $t_{ij}$  [1], and thus for the correct description of valence and metal-insulator transitions in these compounds one has to take the limit  $U > t$  and not  $U < t$ . On the other hand it should be mentioned that in the Falicov-Kimball picture it is possible to get the metal-insulator transition much easier, for example by including spins. Indeed, numerical calculations

performed for the spin-one-half FKM showed [11] that the metallic domain is stable in this model for a wide range of model parameters, including large values of  $U$  and  $n_f$ . Above the region of phase segregation we have observed the region of stripes formation ( $N_f = 10, \dots, 20$ ). In this region the  $f$ -electrons form the one-dimensional charge lines (stripes) that can be perpendicular or parallel (see Fig. 1b). This result shows that the crucial mechanism leading to the stripes formation in strongly correlated systems should be the competition between the kinetic and short-range Coulomb interaction. Going with  $N_f$  to higher values of  $N_f$  the stripes vanish and again appear at  $N_f = 26$ , however in a fully different distribution (see Fig. 2a). While at smaller values of  $N_f$  the stripes have been distributed inhomogeneously (only over one half of lattice) the stripes in the region  $N_f = 26, \dots, 31$  are distributed regularly. Above this region a new type of configurations (see Fig. 2b) starts to develop. We call them the diagonal charge planes with incomplete chessboard structure, since the  $f$ -electrons prefer to occupy the diagonal planes with slope 1 and within these planes they form the chessboard structure. Of course, there is a considerable freedom in categorization of ground state configurations according to some common features and the case of diagonal planes used by us is only one of possible ways. The region of diagonal charge planes is relatively broad and extends up to  $N_f \sim 50$ . Then follows the region in which



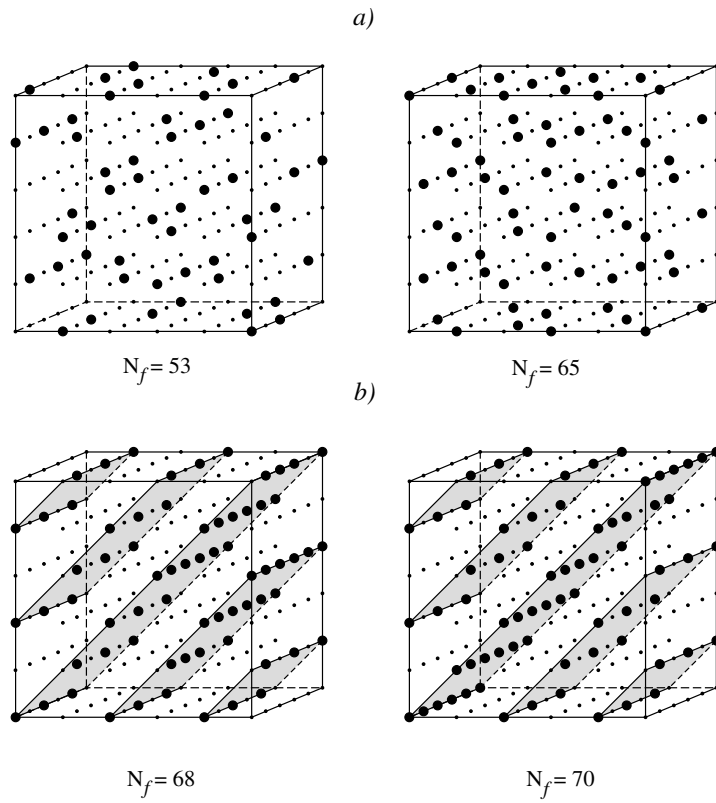
**Fig. 2.** Typical examples of striped configurations with regular distribution (a) and diagonal charge planes with an incomplete chessboard structure (b) obtained for  $U = 1$  and  $L = 6 \times 6 \times 6$ .

the chessboard structure starts to develop. As illustrated in Figure 3a the  $f$ -electrons begin to occupy preferably the sites of sublattice A, leaving the sublattice B free of  $f$ -electrons. In addition, the configurations that can be considered as mixtures of previous configuration types are also observed in this region (see Fig. 3b). However, with increasing  $N_f$  the configurations of chessboard type become dominant. Analysing these configurations we have found that the transition to the purely chessboard configuration realizes through several steps. The first step, the formation of the chessboard structure has been illustrated in Figure 3a. The second step is shown in Figure 4a. It is seen that the chessboard structure is fully developed in some regions (planes) that are separated by planes with incomplete developed chessboard structure. Such a type of distribution is replaced for larger values of  $N_f$  by a new type of distributions (step three), where both regions with complete and incomplete chessboard structure have the three-dimensional character (see Fig. 4b).

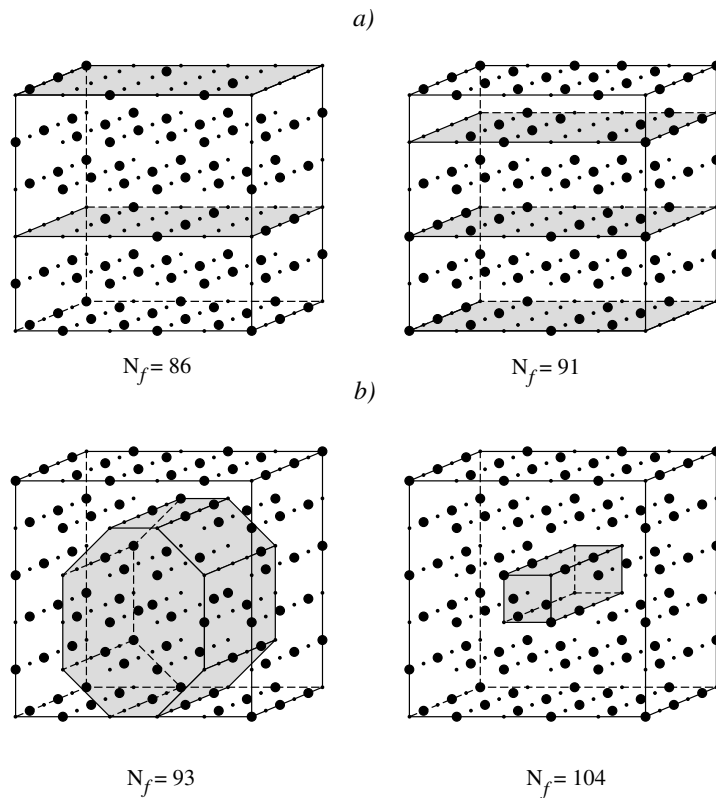
The same picture we have observed also for intermediate values of Coulomb interactions ( $U = 2$ ). The larger values of  $U$  only slightly modify the stability regions of some phases, but no new configuration types appear. In particular, the domain of phase segregation, as well as the domain of stripes formation are reduced while the domain of diagonal planes with chessboard structure increases. This trend is observed also for larger values of

$U$ . In the strong coupling limit ( $U = 8$ ) the phase segregated and striped phases absent and the region of stability the diagonal planes extends to relatively small values of  $N_f \sim 20$ . Below this value a homogeneous distribution of  $f$ -electrons is observed. Thus we can conclude that all fundamental results found in one and two-dimensional solutions of the FKM (the phase segregation, the stripes formation, the phase separation, etc.) holds also in three dimensions, thereby the FKM becomes interesting for a description of ground-state properties (e.g., valence and metal-insulator transitions induced by doping and pressure) of real (three dimensional) systems [12]. The work in this direction is currently in progress.

In summary, the ground-state properties of the three-dimensional FKM were examined by a well-controlled numerical method. The results obtained were used to categorize the ground-state configurations according to common features for weak ( $U = 1$ ), intermediate ( $U = 2$ ) and strong interactions ( $U = 8$ ). It was shown that only a few configuration types form the basic structure of the phase diagram in the  $n_f - U$  plane. In particular, the largest regions of stability correspond to phase segregated configurations, striped configurations and configurations in which electrons are distributed in diagonal planes with incomplete chessboard structure. Near half-filling, mixtures of two phases with complete and incomplete chessboard structure were determined.



**Fig. 3.** The ground state configurations for intermediate  $f$ -electron concentrations. (a) The formation of the chessboard structure. (b) The examples of ground-state configurations that can be considered as mixtures of configuration types with smaller  $n_f$  ( $U = 1$ ,  $L = 6 \times 6 \times 6$ ).



**Fig. 4.** Examples of an incomplete chessboard structure obtained for  $U = 1$  and  $L = 6 \times 6 \times 6$ . (a) The chessboard structure is fully developed in some regions (planes) that are separated by planes with incomplete developed chessboard structure. (b) Both regions with complete and incomplete chessboard structure have the three-dimensional character.

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