FALICOV–KIMBALL MODELS OF COLLECTIVE PHENOMENA IN SOLIDS (A CONCISE GUIDE)*

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(Received June 21, 2001)

32 years have passed since Falicov and Kimball proposed a model of correlated electrons attempting to explaine such cooperative phenomena as metal–insulator transitions in some solids. We present a telegraph-style review of the development of the theory and applications of models based on the Falicov and Kimball idea which has taken place during the last 15 years of increased interest in these models. The list of collective phenomena that have been discussed in the framework of Falicov–Kimball models includes crystallization, segregation, phase separation, formation of molecules, flux phases, mixed-valence states, Peierls instability and metal-insulator transitions. We point out new trends and some open problems.

PACS numbers: 71.10.–w

* Presented at the XII School of Modern Physics on Phase Transitions and Critical Phenomena, Lądek Zdrój, Poland, June 21–24, 2001.

1. The "Ising model" of interacting quantum quasiparticles and a glimpse of its history

1.1. The origin

One of the most intriguing features of solids, like for instance transition-metal or rare-earth compounds, are metal–insulator transitions. They are observed when external factors like temperature, pressure or composition are varied [1]. Several mechanisms have been proposed to explain metal–insulator transitions, all based on the assumption that it is sufficient to consider only the electron subsystem of a solid. Clearly, such effects are out of reach of band theories of solids. That is why all the considered mechanisms agree in one respect, namely a theory of a metal–insulator transition has to be based on a model of an interacting electron gas (or more precisely, interacting quasiparticle gas). Probably the first idea, nowadays known as the Mott–Hubbard mechanism, was that the electron–electron interaction is capable of changing the nature of electronic states from localized to itinerant, and the basic model studied was the Hubbard model and related ones [2]. This approach turned out to be rather unsuccessful because of enormous technical difficulties. Another mechanism was proposed by Falicov and Kimball [3]. An analysis of experimental data for transition-metal and rare-earth compounds led them to put forward a new idea. Namely, they suggested that the metal–insulator transitions in these compounds occur due to a change in the occupation numbers of two sorts of electron states, extended Bloch-like states and localized states centered at the sites of the metallic ions in the crystal, while these states remain basically unchanged. This mechanism of metal–insulator transitions is nowadays referred to as the charge-transfer mechanism [4].

1.2. The "backbone Hamiltonian"

The simplest Hamiltonian that grasps the idea of Falicov–Kimball, and is referred to as the Hamiltonian of the one-band spinless Falicov–Kimball Model (FKM), is $H_\Lambda = T_\Lambda + V_\Lambda$, where T_Λ stands for the hopping energy of spinless itinerant fermions that hop on a finite portion of a Bravais lattice (or just a finite graph) Λ with $|\Lambda|$ sites, and V_Λ is the interaction energy between the itinerant fermions and localized (immobile) particles (with any statistics). In terms of creation and annihilation operators a_x^*, a_x of a fermion at a site x of Λ, the T_Λ term reads

$$T_\Lambda = -\sum_{x,y\in\Lambda} t_{xy} a_x^* a_y, \tag{1}$$

with the hopping amplitudes t_{xy} being matrix elements of a $|\Lambda| \times |\Lambda|$ Hermitian matrix t_Λ. The lattice Λ is assumed to be of the alternant type, *i.e.*,

it consists of two sublattices Λ^0, Λ^e such that the nearest neighbours of a site on one sublattice belong to the other one. Moreover, the hopping amplitudes t_{xy} are nonzero if and only if sites x, y belong to different sublattices. Typically, Λ is a piece of a d-dimensional hypercubic lattice and then Λ^0, Λ^e are the odd and even sublattices, respectively, and t_{xy} assumes a real value $t > 0$ if x and y are nearest neighbour sites and vanishes otherwise. The interaction energy V_Λ can be expressed in terms of occupation number operators of itinerant fermions, $n_x = a_x^* a_x$, and occupation number operators of localized particles, $w(x)$:

$$V_\Lambda = -2U \sum_{x \in \Lambda} w(x) n_x. \tag{2}$$

Apparently, the occupation number operators of localized particles, $w(x)$, commute with H_Λ, thus they can be looked upon as a classical field taking values from the set of natural numbers; in the case of fermions $w(x)$ can be simply 0 or 1. Since the itinerant particles are fermions, for a fixed set of occupation numbers $w(x)$, in the literature of the model referred to as the configuration of localized particles, the Hamiltonian H_Λ is a second-quantized form of a one-particle Hamiltonian $h_\Lambda = t_\Lambda + 2UW_\Lambda$, where t_Λ is a Hermitian matrix with xy matrix element equal to t_{xy} and W_Λ is a diagonal $|\Lambda| \times |\Lambda|$ matrix with xx matrix elements equal to $w(x)$.

1.3. The effective interaction

The most convenient Gibbs ensemble to study such systems as FKM is the grand-canonical ensemble and the grand-canonical partition function of FKM has the form

$$Z_\Lambda = \sum_{\{w(x)\}} \operatorname{Tr} \exp[-\beta(H_\Lambda - \mu_i N_i - \mu_l N_l)]\,, \tag{3}$$

where β is the inverse temperature and μ_i (N_i), μ_l (N_l) are the chemical potentials (total particle numbers) of itinerant and localized particles, respectively, and Tr stands for the trace over the fermion-particle Fock space. There is direct interaction neither between the itinerant particles nor between the localized ones, still the simple on-site interaction between the two kinds of particles involved induces a complicated, many-body and long-range effective interaction within each group of particles. This effective interaction is most easily defined for the localized particles. Denoting it by $F_\Lambda(\beta, \mu_i, \mu_l, W_\Lambda)$, we have the following defining formula (see [11])

$$Z_\Lambda = \sum_{\{w(x)\}} \exp[-\beta F_\Lambda(\beta, \mu_i, \mu_l, W_\Lambda)]\,, \tag{4}$$

which implies in turn the following compact form of $F_\Lambda(\beta, \mu_i, \mu_l, W_\Lambda)$

$$F_\Lambda(\beta, \mu_i, \mu_l, W_\Lambda) = -\beta^{-1} \operatorname{Tr} \ln \cosh \left[\beta \frac{h_\Lambda - \mu_i}{2} \right] - \mu_l \sum_{x \in \Lambda} w(x) - |\Lambda| \frac{\mu_i}{2} - \beta^{-1} |\Lambda| \ln 2 \,. \tag{5}$$

In the above formula Tr stands for the trace over the $|\Lambda|$-dimensional one-particle state-space. There are not many instances in physics literature, where an effective interaction can be written explicitly. Thus, it is a remarkable fact that in the Falicov–Kimball model it can be not only determined but also described by such a simple formula. Without exaggeration, one can say that studies of phase diagrams, that have been carried out up to now, can be considered as studies of the above effective interaction, to large extent in the $\beta \to \infty$ limit.

1.4. The history loop: from solid state theory, via statistical mechanics to solid state theory

The first 15 years (1970–1985) of studies of FKM and related models, can be classified as a solid-state theory era. The interest of researchers was directed mainly towards such phenomena as metal–insulator transitions [3,5–7] and mixed-valence phenomena; see [1] for review of the results obtained in that period and an extensive list of references (however some general, model oriented studies were also carried on [8]). The approximate methods used (typically, various decoupling schemes in the Green function technique) did not result in a clear view of the situation, a great deal of confusion arised. A break through in the studies of FKM models occurred in 1986, when two papers, one by Brandt and Schmidt [9] and another by Kennedy and Lieb [10] appeared. The papers contained mathematical arguments proving the existence of a phase transition in FKM. These two papers renewed interest in the model and started the decade, 1986–1996, of intensive research which can be named the statistical mechanics/mathematical physics era of the research of the model. The achievements of this decade are summarized in an excellent and comprehensive review by Gruber and Macris [11], who contributed a great deal to the theory of FKM. In 1989 another important development took place, namely Brandt and Mielsch [12] noticed that in the appropriate limit of infinitely dimensional lattice the Green functions technique applied to FKM does not result in an infinite chain of coupled equations but leads to a finite number of equations, that is, in this limit the FKM is exactly solvable. There are good reasons to believe that the results obtained in that limit for FKM are close to those which hold in the three-dimensional system and even in the two-dimensional one [13,14]. This achievement was the first

sign of the comeback of the solid-state era in about a decade. In late nineties, after about a quarter of a century, again the main questions studied were suggested by the solid-state physics. The possibility of calculating response functions without uncontrolled approximations (like decoupling schemes of Green functions), in the limit of infinite dimensionality, for the first time opened possibilities of comparing theory with experimental data measured in such systems as for instance NiI_2, as has been shown by Freericks and coworkers [4, 15]. Thus, the story of FKM seems to be a classic example of the spiral of development. We have found out the level of interest in Falicov–Kimball models, as measured by the number of those papers per year which contain the key-word *Falicov–Kimball model*, to give a quantitative representation of the described above history loop, see Fig. 1.

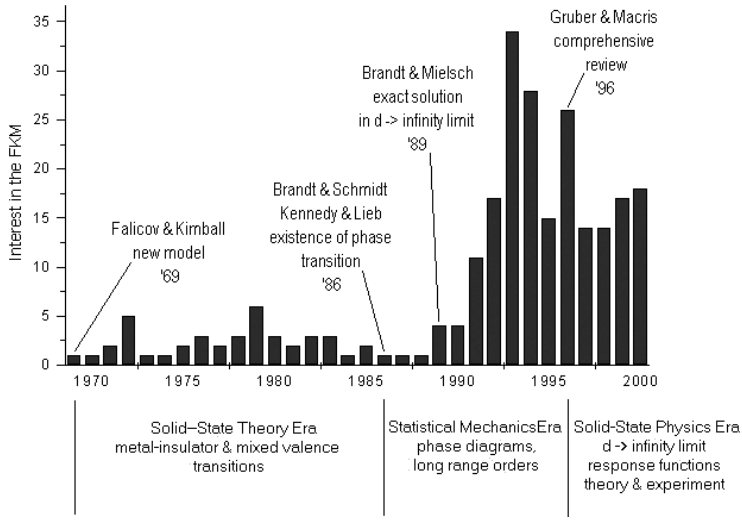

Fig. 1.

2. Falicov–Kimball models — extensions of the "backbone Hamiltonian"

2.1. Changes of quantum statistics

Kennedy and Lieb [10] discussed already the effect of replacing the itinerant fermions by itinerant bosons. They have described the ground state of itinerant bosons and have shown that the phase transition, they found in the fermionic case, disappears. Some more information about the bosonic ground state can be found in [16]. If, however, instead of bosons one takes hard-core bosons (their creation and annihilation operators satisfy the ca-

nonical anticommutation relations if both operators involved in the anticommutator are labeled by the same site and they commute if they are labeled by different sites), then, as shown by Gruber *et al.* [17], the properties of the modified model are, in many respects, analogous to the original one. Replacing fermions by hard-core bosons is a highly nontrivial step, since even for a fixed configuration of localized particles H_Λ fails to be a second quantized form of a one-particle Hamiltonian.

2.2. Modifications of lattices and/or hopping amplitudes of itinerant particles

Gruber *et al.* [17] studied the FKM on the triangular lattice. In the same paper complex hopping amplitudes, whose phase can be related with an external magnetic field acting on moving charged particles (Lorentz force), were taken into account. Recently, one can observe an increased interest in models with so called correlated hopping, *i.e.* with the hopping amplitudes t_{xy} depending on the localized-particle occupation numbers at sites x and y [18, 19].

2.3. Additional degrees of freedom

One can consider for instance many bands of itinerant fermions [20, 21] and supply both kinds of particles with spin [22], which can be different for the itinerant and localized particles [23].

2.4. Modifications of localized-particle occupation numbers

The variables $\{w(x)\}_{x\in\Lambda}$ taking values from the set $\{1, 0, -1\}$ or taking any real value have been studied by Lebowitz and Macris [24] while Gruber and Macris considered them to represent a vector field [11].

2.5. Additional interactions of localized particles

Brandt *et al.* [22] studied an extension of FKM, where both kinds of particles had spin 1/2, so there were two kinds of the localized-particle occupation numbers: $w_+(x)$ and $w_-(x)$. They introduced an on-site interaction of the form $\sum_{x\in\Lambda} w_+(x)w_-(x)$. In [24], where continuous variables $w(x)$ have been considered, an interaction of the form $\sum_{x\in\Lambda} w^2(x)$ have been taken into account.

3. Phase diagrams for the "backbone Hamiltonian"

The vast majority of papers devoted to FKM and its extensions is, up to now, concerned with equilibrium properties. Even the criteria for metal–insulator transitions refer usually to equilibrium [10]; only recently, in the $d \to \infty$ limit, a criterium based on the conductivity, defined in the framework of the linear response theory, was used [4]. From the point of view of statistical mechanics, FKM is a two-component system governed by a unique interaction parameter, say U/t. The ultimate goal would be to determine the grand canonical (or canonical) phase diagram in the four-dimensional space with a coordinate system whose axes are labeled by the two chemical potentials, μ_i, μ_l (or, in the canonical ensemble, by the corresponding particle densities of the itinerant and localized particles), the unique interaction parameter U/t and the inverse temperature β. The regions occupied by pure phases would contain information about the localized-particle configurations and the state of the itinerant particles. This is, of course, hardly achievable. The obtained results refer only to some sections of the complete phase diagram. Typically, one finds zero-temperature and a fixed U/t-value sections (ground-state phase diagrams), with a limited region in the plane of the chemical potentials (particle densities), for instance, a vicinity of the hole–particle symmetry point or the region where the neutrality condition is satisfied (*i.e.* for $U < 0$ the densities of the itinerant and localized particles are equal). Moreover, in most papers only the configurations or long-range orders in the subsystem of localized particles are determined while the state of the itinerant particles remains unknown.

Concerning the dependence of phase diagrams on the unique interaction parameter U/t, rigorous results that hold for all values of this parameter were obtained only at the hole–particle symmetry point and in its vicinity [10, 25]. Away from the symmetry point the existing phase diagrams have been obtained by means of numerical methods, while analytical, rigorous results are limited to the weak coupling regime or the strong coupling one. The weak coupling regime is notoriously difficult (see open problems), thus the vast majority of non numerical results refers to the strong coupling regime. Concerning numerical work, many interesting results have been obtained by means of restricted phase diagrams, first obtained in [26], and then in [27–29]. In the strong coupling regime, the $1/U$-expansion of the effective interaction at the zero-temperature limit, and the corresponding phase diagram, has been first constructed by Gruber *et al.* [30] and, then made rigorous and extended by Kennedy and coworkers [31,32]. Remarkable results in the strong coupling limit, but restricted to one-dimensional FKM, have been obtained by Lemberger [33]. Recently, powerful methods of constructing effective interactions have been developed, see for instance [34,35], which extend considerably the class of FKM-like models that can be studied by means of rigorous methods.

4. Collective phenomena that have been discussed in the framework of Falicov–Kimball models

In this section we simply give a list of most important, in our view, applications of the FKM and its extensions for describing collective phenomena in solids. Even brief descriptions of the items of this list would exceed the limits for volume of this concise guide. Therefore, we supply the reader with relevant references (but we do not pretend to give a complete list) and advice also consulting the Gruber and Macris review [11].

- Crystallization [10, 27–33]
- Segregation [27, 33, 36–39]
- Phase separation [28, 29, 40–42]
- Formation of molecules [27–29]
- Effects of magnetic flux and flux phases [17, 43]
- Effects of additional degrees of freedom [4, 22, 44]
- Effects of correlated hopping [18, 19, 45, 46]
- Peierls instability [24]
- Mixed-valence transitions [47–49]
- Metal–insulator transitions [4, 50, 51]

5. New trends

The dynamic mean field theory appears to be particularly successful when applied to the FKM, since within its formalism both thermodynamic and dynamic properties of the system, including correlation functions of localized and itinerant particles, could be calculated exactly in the limit of large dimensions [12,23,52] (but rigorous results concerning itinerant-particle correlation functions are still scarce, see [24, 53, 54]). As a result, many finite temperature characteristics of the model, including *e.g.* susceptibilities against external forces or transport properties could be determined [55, 56]. The theory opens new opportunities for modeling strongly correlated electron systems, where calculated quantities could be compared and verified by experimental measurements. In our opinion, studies in this direction will be dominating in coming years.

6. Open problems

No doubt, physical problems of interest can be formulated as questions about the nature of metal–insulator or mixed-valence transitions *etc.* But to answer these and related questions we have to extend our knowledge of FKM phase diagrams. Here are some technical problems that require further studies:

- Perturbation theory for small coupling U in dimensions $d > 1$; the $d = 1$ case has been done in [42].
- Properties of non-neutral (not half-filled) system; there are some numerical results [27–29] and the results for strong coupling that refer to segregation [33, 36–39].
- The influence of additional degrees of freedom, like spin, on the phase diagrams is almost not known; see [4, 22, 44] for the results in this direction, which signal important new effects.

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