Phase transitions in the three-dimensional Falicov–Kimball model

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ABSTRACT

The classical Monte-Carlo method is used to study the finite-temperature properties of the three-dimensional (D = 3) Falicov–Kimball model in the symmetric case. It is shown that the critical temperature of the phase transition from the low-temperature ordered phase to the high-temperature disordered phase in D = 3 is considerably enhanced in comparison to the two-dimensional case (D = 2). A significant shift to higher values of the Coulomb interaction U (with respect to D = 2) is also found for the critical point Dc, at which the nature of the phase transition changes from the first to second order. In addition, the temperature dependence of the itinerant electron density of states (DOS) is analysed. For very low-temperatures we have observed a formation of a fine structure inside the principal gap that transforms to a pseudo-gap at higher temperatures and becomes temperature independent for sufficiently large temperatures. In this temperature region we have calculated DOS for different Coulomb interactions and found the Mott-Hubbard transition.

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1. Introduction

Since its introduction in 1969, the Falicov–Kimball model (FKM) has become an important standard model for a description of correlated fermions on the lattice [1]. It has been used in the literature to study a great variety of many-body effects in rare-earth compounds, in which valence transitions, charge-density waves and electronic ferroelectricity are the most common examples [2,3]. The Hamiltonian of the model can be written as a sum of three terms

\[ H = \sum_{ij} t_{ij} d_i^+ d_j + U \sum_i f_i^+ f_i d_i^+ d_i + E_f \sum_i f_i^+ f_i, \]

where \( f_i^+, f_i \) are the creation and annihilation operators for an electron in the localized state at lattice site \( i \) with binding energy \( E_f \) and \( d_i^+, d_i \) are the creation and annihilation operators of the \( d \)-electrons in the \( d \)-band Wannier state at site \( i \). The first term of (1) is the kinetic energy corresponding to the 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 (if not mentioned otherwise, all energies are measured in units of \( t \)). The second term represents the on-site Coulomb interaction between the localized \( f \) electrons. The third term stands for the localized \( f \) electrons whose sharp energy level is \( E_f \).

Recent calculations showed that the FKM can yield the correct physics for a description of ground-state [4] as well as the thermodynamic [5] properties of rare-earth and transition-metal compounds (at least qualitatively). However, most of the results obtained for the FKM have been calculated for the limiting cases of \( D = 1, D = 2 \) and \( D = \infty \) (where \( D \) is the dimension of the system) and it is not clear if these results hold also in the realistic case \( D = 3 \). The first attempt to describe systematically the ground-state properties of the FKM in three dimensions has been made recently [6]. In the present paper we focus our attention to the thermodynamics of the three-dimensional FKM.

2. Method

Since in the FKM the \( f \)-electron occupation number \( f_i^+ f_i \) 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. The Hamiltonian (1) can be rewritten as

\[ H = \sum_{ij} h_{ij}(w) d_i^+ d_j + E_f N_f, \]

where \( h_{ij}(w) = t_{ij} + U w_i \delta_{ij} \) and \( N_f \) is the number of \( f \) electrons. Thus the Hamiltonian (2), for a given \( f \)-electron configuration \( w \) defined on the three-dimensional lattice, is the second-quantized version of the single-particle Hamiltonian \( h(w) = t + Uw \). Therefore, the classical Metropolis algorithm [7,8], instead of the quantum Monte-Carlo (MC) algorithm, can be used directly for the investigation of thermodynamics of the FKM. Using the so called electronic free energy [8]

\[ F(w) = (E_f - \mu) N_f - \frac{1}{\beta} \sum_i \ln(1 + e^{-\beta(\epsilon_i - \mu)}), \]

where \( \epsilon_i \) are the orbital energies of the localized \( f \) electrons and \( \mu \) is the Fermi level.
(where $\beta = \frac{1}{k_B T}$, $\tau = k_B T / \mu$, $\mu$ is the chemical potential and $\epsilon$ are the eigenvalues of the matrix $h$) the grand-canonical partition function can by written as $\mathcal{Z} = \sum_\mathbf{w} e^{-\beta F(w)}$, where the summation runs over all possible $f$-electron configurations. In analogy with the classical spin systems, the electronic free energy is used as the statistical weight in the Metropolis algorithm. The thermal averages can be expressed as

$$\langle X \rangle = \sum_\mathbf{w} X^w \chi_w, \quad \chi_w = \frac{e^{-\beta F(w)}}{\mathcal{Z}}.$$  \tag{4}

The proper expression for the specific heat (at constant volume $V$ and total number of particles $N$) can be obtained from the following equation:

$$C_{V,N} = \left( \frac{\partial (E - \mu N)}{\partial T} \right)_{V,\mu} - T \left( \frac{\partial^2 (E - \mu N)}{\partial \mu^2} \right)_{T},$$  \tag{5}

where $E$ is the internal energy. Another important quantity discussed in this paper is the structure factor defined as:

$$S_q^w(\mathbf{Q}) = \frac{1}{2L} \sum_{j,k} e^{i\mathbf{Q} \cdot (\mathbf{R}_j - \mathbf{R}_k)} \langle w_j w_k \rangle,$$  \tag{6}

where $\mathbf{R}_j$ stands for the position vector of $j$ site. The simulations started mostly at high temperatures with a random configuration. Data were generated typically with $10^4$ MC steps per site after discarding at least $3 \times 10^4$ initial MC steps per site.

### 3. Results and discussion

Since the grand-canonical ensemble is used, the total number of particles is a function of chemical potential $\mu$. Generally, this means a substantial complication, because for the required number of electrons, the chemical potential has to be found for each temperature. To avoid this complication, we consider only the symmetric point ($\mu = U/2$, $E_f = 0$) of the model. In the symmetric case $H - \mu N$ remains unchanged under the particle-hole transformation and the average number of all electrons is $N = L$ (where $L$ is the lattice size). It is known [9], that for this case the ground-state $f$-electron configuration has the chessboard structure for all values of $U > 0$. Moreover, this configuration is stable even for the finite temperatures, with the $U$ dependent critical temperature of the phase transition, as was shown for $D = 2$ and $D = \infty$ [8,10]. We have observed a similar behavior for $D = 3$ by analyzing the temperature dependencies of the specific heat $C_{V,N}(\tau)$. The typical examples obtained for $U = 2$ and $U = 8$ in three dimensions are shown in Fig. 1.

One can see that $C_{V,N}$ as a function of $\tau$ shows a sharp low-temperature peak which scales with the lattice size. Hereby the structure factor $S_q(\tau, \pi)$ changes rapidly from 1 to 0 (see insets in Fig. 1), near the temperature where the maximum of $C_{V,N}$ appears. These facts indicate that the sharp maxima are related to the breaking of the chessboard structure and their locations can be used to estimate the critical temperature of the phase transition from the low-temperature ordered phase to the high-temperature disordered phase. In addition, the critical temperatures for several values of $U$ have been calculated also by using the standard Binder intersection method [11] and a nice correspondence of the results has been observed.

Fig. 2 shows the transition temperatures as functions of $U$. To be able to compare the critical temperatures obtained in different dimensions it is necessary to use the units of $t' = 2t \sqrt{D}$ instead of $t$ [12]. In the limit of small Coulomb interactions the critical temperatures (in all shown dimensions) are comparable. It is evident that outside this limit the critical temperature in $D = 3$ is considerably enhanced in comparison to $D = 2$, and in opposite, it is considerably smaller than in $D = \infty$. In addition, we have found that the maximum of the critical temperature $\tau_c$ shifts to the higher values of $U$ with the increasing dimension.

Let us now discuss the type of phase transitions at $\tau = \tau_c$. It is well-known that the FKM for $U \gg 1$ maps onto an antiferromagnetic Ising model ($J = t^2 / U$), therefore for a large $U$, the phase transition is of the second order. On the other hand, it was shown [8] that in two dimensions the FKM for $U \lesssim \tau$ undergoes a first-order phase transition. Since the thermodynamic behavior of a finite system is smooth when it undergoes a phase transition for both the first order and the second order transition, it is often hard to determine a type of transition. To identify the first order phase transition we have used the method proposed by Challa, Landau and Binder [13], which was generalized for the large class of classical lattice models in the work of Borgs and Kotecky [14]. In the mentioned works, it was shown that in the case of the
first order phase transition one can describe the distribution of states by the sum of two Gaussians with typically rather different parameters. Therefore, if the system is close to the first order phase transition, the probability distribution of the electronic free energy $P(F)$ has to consist of two Gaussians centered at different energies pertaining to the ordered and disordered phase. In Fig. 3 the distribution of electronic free energy $P(F)$ for $U = 2$ and various temperatures near $\tau_c$ is shown. All curves have a double-peak structure, therefore there is clearly the first order phase transition at $\tau_c$. The two-peaks character of $P(F)$ near the temperature of the phase transition persists to surprisingly high values of $U$. The distribution of electronic free energy near the $\tau_c$ for $U = 4$ (Fig. 4) suggests that the first order phase transition can take place up to $U = 4$ in $D = 3$. Although the results obtained for lattice sizes up to $L = 1000$ support this suggestion, one should be careful in making definitive conclusions, since we are not able to exclude completely the influence of the finite size effects. For $U > 4$ we have observed only a simple one-peak structure of $P(F)$ that confirms the second order phase transitions in the strong coupling region (see the inset in Fig. 4).

Besides the order of phase transitions also other properties of the FKM show a different behavior in the weak and strong coupling limit. For example, we have observed that the double occupancy per site $\delta$ as a function of temperature is clearly different for weak and strong interactions. Fig. 5 illustrates the typical temperature dependencies of the potential energy ($E_P = U \delta$) for $U = 2$ and $U = 8$. In both regimes $E_P$ is approximately constant for $\tau \to 0$, while for $\tau \sim \tau_c$ $E_P$ rapidly increases for $U < U_c$ and decreases for $U > U_c$, where $U_c \sim 4$.

To reveal the influence of dimension $D$ on the thermodynamic characteristics of the FKM we have also examined the $d$-electron density of states (DOS). In Fig. 6 the single-particle DOS for the
three dimensional FKM is shown for $U = 2$ and $U = 8$ and various temperatures $\tau$. We have found that for $\tau \to 0$ there is a gap of width $U$ (at $\mu = U/2$) that is in agreement with the rigorous results for the ground state [16]. As the temperature is raised a fine structure appears in the DOS in the region of $0 < \omega < U$. We have observed that with increasing $\tau$ this fine structure transforms to a pseudo-gap in $D = 3$. This is illustrated in Fig. 7 where the details of the temperature evolution of DOS in the region of $0 \leq \omega \leq U$ for $U = 2$ and $U = 4$ are presented. In the limit of the weak Coulomb interaction, the sufficiently high temperature closes the pseudo-gap (Fig. 7(a)). For an intermediate $U$ the depth of pseudo-gap decreases with increasing $\tau$ but a pseudo-gap persists even for $\tau \to \infty$ (Fig. 7(b)). Finally, for the strong coupling limit, the fine structure reduces the width of the gap but it still remains open even for $\tau \to \infty$ (Fig. 6(f)).

We have found that for temperatures only slightly higher than $\tau_c$ the DOS does not change with an increasing temperature. This allowed us to study the disordered phase (homogenous phase) in the $\tau \to \infty$ limit that is accessible to numerical simulations on much larger clusters. In Fig. 8 the DOS of the homogenous phase is shown for different $U$ and different dimensions. One can see that for small values of $U$ the three-dimensional DOS is almost the free electron DOS. As $U$ increases, the pseudo-gap with a fine structure appears at the Fermi energy, and finally, in the limit of the strong Coulomb interactions the gap is opened. This illustrates the finite temperature Mott-Hubbard transition as a function of $U$ in the real dimension. The same scenario has been observed also for $D \leq 2$ and $D = \infty$. Moreover, Fig. 8 shows that with increasing dimensions, the fine structure becomes smoother, and finally, transforms in the smooth maxima for $D = \infty$.

Summarizing our results the following conclusions can be made for the homogeneous phase. In the weak coupling limit the character of DOS in $D = \infty$ is even closer to the realistic $D = 3$ case than the $D = 2$ case. The opposite is true in the strong coupling limit, where the DOS in infinite dimensions lacks the peaks and local minima near $\omega = \mu \pm U/2$. This fact can play an important role in the correct description of electronic correlations of real materials in $D = 3$ since these details are connected with the interaction between $f$ and $d$ electrons.

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![Fig. 7. The detail of the temperature evolution of the fine structure of DOS in the region of $0 \leq \omega \leq U$ for $U = 2$ and $\tau = {0.04, 0.06, 0.08, 0.1, 0.11, 0.118}$ (from the bottom up in the figure) and for $U = 4$ and $\tau = {0.135, 0.20, 0.35}$ (from the bottom up in the figure) and $\tau \to \infty$ (dotted line).](image1)

![Fig. 8. DOS in the homogenous phase computed for different $U$ and different dimensions. The open boundary conditions and units of $t' = 2t\sqrt{D}$ are used. In the infinite dimension the DOS (computed by DMFT [17]) for the hypercubic lattice (solid line) and the Bethe lattice (dashed line) are plotted.](image2)
References