Charge and magnetic order at finite temperatures

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We perform a simplified analysis of finite temperature properties of the generalized Falicov-Kimball model with Hund coupling. The model is studied on the square lattice for fixed values of \( f \)-electron and \( d \)-electron densities \( \rho_f = 1/2, \rho_d = 1 \). Using an approxiamte scheme we estimated free energies of the following three phases: the charge and spin ordered (SCO), the charge ordered (CO) and the non-ordered (NO). Comparing the free energies we detected phase transitions and found how the transition temperatures depend on the on-site interaction parameters. It appears, that the transition temperature between SCO and CO is much lower than between CO and NO, but their maximum values are attained at values of coupling constants very close to each other.

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I. INTRODUCTION

Charge and magnetic order are observed in many correlated electron systems as, for example, in \( R_{2-x}Sr_xNiO_4 \), where \( R = La, Nd [1] \). One of the models capable to describe such superstructures is the Falicov-Kimball model, extended by the spin-dependent local interaction, that reflects the first Hund’s rule [2].

Until now the model has been studied only at zero temperature. Using the method of restricted phase diagrams or small cluster numerical diagonalization it was shown how the charge distribution and spin arrangement change with parameters of the local interaction and densities of localized and itinerant electrons [2, 3]. In particular, for \( \rho_f = 1/2 \) and \( \rho_d = 1 \) it was found that in 2D the ground state forms the checkerboard charge pattern consistent with the simplest antiferromagnetic order [2].

Here we investigate behaviour of the system at finite temperatures. Our purpose is to determine the way of transformation of the ordered phase into disordered one when temperature increases. The transformation appears to split into two phase transitions: at \( T_{MO} \) between the low temperature phase, where both charge and spin are ordered (SCO) and the intermediate phase, with only charge but not spin ordered (CO), and then at \( T_{CO} \) between CO and the high temperature phase, that is disordered with respect to both the spin and charge (NO). A visualization of arrangements of the localized electrons in the three phases is shown in Fig. 1.

\[ H = t \sum_{\langle i,j \rangle} \sum_{\sigma = \uparrow, \downarrow} d_{i,\sigma}^+ d_{j,\sigma} + U \sum_i \sum_{\sigma,\eta = \uparrow, \downarrow} n_{i,\sigma}^d n_{i,\eta}^f - J \sum_i (n_{i,\uparrow} - n_{i,\downarrow}) (n_{i,\uparrow}^f - n_{i,\downarrow}^f), \tag{1} \]

where \( \langle i,j \rangle \) means the nearest neighbor lattice sites \( i \) and \( j \), \( \sigma \) and \( \eta \) are spin indices, \( d_{i,\sigma} (d_{i,\sigma}^+) \) is an annihilation (creation) operator, and \( n_{i,\sigma}^d (n_{i,\sigma}^f) \) is an occupation number of itinerant(localized) electrons. The on-site interaction

\[ \sum_{i,\sigma} \epsilon_d n_{i,\sigma}^d + \sum_{i,\sigma} \epsilon_f n_{i,\sigma}^f + U \sum_i \sum_{\sigma,\eta = \uparrow, \downarrow} n_{i,\sigma}^d n_{i,\eta}^f, \]

\[ -J \sum_i (n_{i,\uparrow} - n_{i,\downarrow}) (n_{i,\uparrow}^f - n_{i,\downarrow}^f), \]
between localized and itinerant electrons is represented by two coupling constants: $U$, which is spin-independent Coulomb-type and $J$, which is spin-dependent and reflects the Hund’s rule force. The hopping amplitude $t$ we set equal to one, so we measure all energies in units of $t$.

Double occupancy of the localized electrons is forbidden, implying the on-site Coulomb repulsion $U_{ff}$ between two $f$-electrons is infinite. Consequently, at a given site the $f$-electron occupancy is assumed to be $n_f = n_{f,\uparrow} + n_{f,\downarrow} \leq 1$ and the $d$-electron occupancy to be $n_d = n_{d,\uparrow} + n_{d,\downarrow} \leq 2$. So there are 3 states per site allowed for the $f$-electrons ($n_f = 0$; $n_{f,\uparrow} = 1$ and $n_{f,\downarrow} = 0$; $n_{f,\uparrow} = 0$ and $n_{f,\downarrow} = 1$) and 4 states per site allowed for the $d$-electrons ($n_d = 0$; $n_{d,\uparrow} = 1$ and $n_{d,\downarrow} = 0$; $n_{d,\uparrow} = 0$ and $n_{d,\downarrow} = 1$; $n_d = 2$).

All single-ion interactions included in $\mathbf{H}$ preserve states of the localized electrons, i.e. the itinerant electrons traveling through the lattice change neither occupation numbers nor spins of the localized ones. In other words, $[\mathbf{H}, f_{\mathbf{i}\mathbf{n}} f_{\mathbf{i}\mathbf{n}}] = 0$ for all $i$ and $\eta$, so the local occupation number is unchanged.

The localized electrons play the role of an external, charge and spin dependent potential for the itinerant electrons. This external potential is ”adjusted” by annealing, so the total energy of the system attains its minimum. In other words, there is a feedback between the subsystems of localized and itinerant electrons, and this is the feedback that is responsible for the long-range ordered arrangements of the localized ones, and consequently for the formation of various charge and/or spin distributions in low temperatures.

II. METHOD OF CALCULATION

In this paper we consider only the case of $\rho_f = 1/2$ and $\rho_d = 1$. Then, for the system composed of $N$ sites there are $N!/(N/2)!^2$ possible charge distributions and, for each of them, there exist $2^{N/2}$ spin configurations. So the entropy (per site) resulting from various possible distributions of $f$-electrons is a sum of the charge part equal to $\frac{1}{2} \log \frac{N!}{(N/2)!^2}$, and the magnetic part equal to $\frac{1}{2} \log 2$. In the limit of $N \to \infty$ the charge part of entropy tends to $\log 2$.

For any fixed configuration of localized electrons $C$ one can calculate all thermodynamic characteristics of the $d$-electron subsystem from standard formulas (see for example [3]). In particular, the Gibbs potential per site $g_C$ is given by the expression

$$g_C(\mu) = -\frac{1}{N\beta} \sum_n \log \left(1 + e^{-\beta E_n(C) - \mu}\right)$$

and the free energy $f_C$ by

$$f_C(\rho_d) = g_C(\mu(\rho_d)) + \mu(\rho_d)\rho_d$$

with the following condition fulfilled (in our case $\rho_d = 1$):

$$\rho_d = \frac{1}{N} \sum_n \frac{1}{1 + e^{\beta E_n(C) - \mu}}$$

Then, the partition function per site $z$ is obtained from the summation over all configurations $C$ of $f$-electrons

$$z = \frac{1}{N} \sum_C e^{-\beta f_c N}$$

and the free energy $f$ is equal to

$$f = -\frac{1}{N\beta} \log \left(\sum_C e^{-\beta f_c N}\right).$$

Since we are not able to determine $f$ exactly in the whole temperature range, we replace it by three different approximate functions, relevant for three different phases: CSO, CO and NO. In order to do this we first divide the sum in (6) as follows

$$\sum_C e^{-\beta f_c N} = e^{-\beta f_{c,\text{SCO}}} + \sum_{C \in \mathbf{C}_{ch}} e^{-\beta f_c} + \sum_{C \in \mathbf{C}^*} e^{-\beta f_c},$$

where $f_{c,\text{SCO}}$ is the free energy of the SCO phase, $\mathbf{C}_{ch}^*$ denotes the set of checkerboard configurations $\mathbf{C}_{ch}$ (with respect to charge) of the $f$-electrons with all possible spin arrangements but SCO and $\mathbf{C}^*$ denotes all C configurations but $\mathbf{C}_{ch}$. 
With an increase of temperature the subsequent terms in (7) contribute to \( f \): for the lowest temperatures only the first term, then, for intermediate temperatures two first terms and, finally, for higher temperatures all three terms. So for temperatures very close to zero the main contribution to \( f \) comes from the free energy \( f_{C_{CSO}} \), attributed to the ground state configuration CSO of the \( f \)-electrons

\[
f 
\approx - \frac{1}{\beta} \log(e^{-\beta f_{C_{CSO}}}) = f_{C_{CSO}}.
\]  

For higher temperatures we calculate mean free energies by averaging them over appropriate sets of \( f \)-electron configurations. In order to find the mean free energies we take into account a representative class of configurations \( C \) of the \( f \)-electrons only, composed of all periodic phases with periods not exceeded four lattice sites. Then our trial set contains 13 non-equivalent configurations. Three of them belong to the \( C_{ch} \) class. For all these configurations we calculated exact free energies per site in the limit of large \( N \) by solving the eigenvalue problem and finding the eigenvalues \( E_n(C) \). This required us to diagonalize up to \( 4 \times 4 \) matrices and result in analytical formulae for at most 4 different energy bands, separately for spin-up and spin-down \( d \)-electrons (for more details see Refs. [2, 5, 6]).

For intermediate temperatures we take an average \( f_C \) over the family \( C_{ch} \) only. Their free energies \( f_C \) are different, but the difference is quite small, even for \( T = 0 \), and tends to zero with temperature (see Fig. 2).

The whole contribution to \( f \) coming from this family could be expressed by a mean value \( \langle f_C \rangle_{C \in C_{ch}} \) in the following way.

\[
f 
\approx f_{CO} = - \frac{1}{\beta} \log(2^{N/2}e^{-\beta N(f_C)_{C \in C_{ch}}}) = - \frac{1}{2\beta} \log2 + \langle f_C \rangle_{C \in C_{ch}}
\]  

We get the appropriate mean free energy \( \langle f_C \rangle_{C \in C_{ch}} \) using the arithmetical average of the relevant free energies \( f_C \). The averaging procedure we used is equivalent to taking the geometrical mean value of relevant ingredients of the partition function. Indeed, we get \( 9 \) if we replace each element of the sum \( \sum_{C \in C_{ch}} e^{-\beta N f_C} \) by \( M_{C_{ch}} \cdot \left( \prod_{C \in C_{ch}} e^{-\beta N f_C} \right)^{1/M_{C_{ch}}} \), where \( M_{C_{ch}} = 2^{N/2} \) is the number of configurations in \( C_{ch} \).
The same procedure we use when calculating an approximative mean free energy \( f_{NO} \) of the non-ordered phase. However, then we take an arithmetic average of free energies of all configurations from the chosen set, and take into account the fact, that in the large \( N \) limit there are \( 2^{N/2} \frac{N!}{(\frac{N}{2})!^2} \approx 2^{N^2/2} \) such configurations. So we have

\[
f \approx f_{NO} = -\frac{1}{2\beta} \log 2 - \frac{1}{\beta} \log 2 + \langle f_{C} \rangle_{altC}.
\]

We tested the averaging procedure for the spinless Falicov-Kimball model and found, that the transition temperatures we got are nearly the same as those obtained by the Monte Carlo simulations in the whole range of the interaction parameter \( U \).

III. RESULTS AND CONCLUSIONS

The results of our calculations are displayed in Fig. 3. In the upper panel a) it is shown the magnetic order-disorder transition temperature \( T_{MO} \) between SCO and CO phases and in the lower panel b) the charge order-disorder transition temperature \( T_{CO} \) between the CO and NO phases. Both \( T_{MO} \) and \( T_{CO} \) were calculated for \( J = 0.2U \) as functions of the rescaled on-site interaction parameter \( \frac{U}{U+1} \).

Since we used rather crude approximative scheme of calculations of the free energies, the obtained values of the transition temperatures could not be very precise. However, we expect that the quantitative picture is correct. In particular, two main conclusions seem to be relevant. First, the maxima of the both transition temperatures are
attained at very close to each other values of the parameter $\frac{U}{U+1} \approx 0.8 \div 0.85$. It corresponds to $U \approx 4 \div 5.67$ (and $J \approx 0.8 \div 1.13$, respectively).

Another important message resultant from the calculations is the fact, that $T_{MO}$ appears to be almost two orders of magnitude lower than $T_{CO}$, even though the Hund coupling constant $J$ is assumed to be only 5 times smaller than $U$. The possible cause of this rather unexpected outcome is the lack of direct interaction between the spin-up and spin-down itinerant electrons (the Hubbard term) in the model. We anticipate that the inclusion of this type of interaction would enhance the magnetic couplings in the system, resulting in an increase of the magnetic transition temperature.

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