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RELATIONSHIPS BETWEEN ELECTRON BAND FILLING AND TYPE OF CHARGE AND MAGNETIC ORDER

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Charge and spin orderings are studied on the square lattice within the generalized Falicov–Kimball model with Hund coupling between localized and itinerant electrons. Using the restricted phase diagram method (RPDM) we detected in diluted magnets a few simple rules of formation of the unique family of ground state periodic phases that are stable both for large and intermediate values of coupling parameters. It appears, that for this group of phases only non-axial stripes composed of ferromagnetic, or sporadically also antiferromagnetic chains, oriented either along one of the two main diagonal directions or along another direction that is non-parallel to the main crystal axis are formed. The antiferromagnetic order is robust in these phases but occasionally ferromagnetic phases are also found.

Key words: diluted magnets, Falicov–Kimball model, itinerant magnet, charge order.

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I. INTRODUCTION

Charge and magnetic order can be studied using the Falicov–Kimball model (FKM) with Hund coupling [1]. The model describes a system of magnetic and non-magnetic ions localized on a lattice and itinerant electrons that hop between sites of the lattice. The ions interact with the electrons by means of two types of on-site interaction. One, that is proportional to charges of the particles (the Coulomb repulsive force) and another, that is proportional to their spins (the Hund force). It is assumed that the Hund coupling has the simplest Ising type anisotropy.

The model Hamiltonian is

$$H = t \sum_{\langle k,l \rangle} \sum_{\sigma=\uparrow,\downarrow} d^{\dagger}_{k,\sigma} d_{l,\sigma} + U \sum_{k} n^{e}_{k} n^{i}_{k} - J \sum_{k} s^{z}_{k} S^{z}_{k}, \quad (1)$$

where $\langle k,l \rangle$ denotes the nearest neighbor lattice sites k and l, σ is a spin indice, $d_{k,\sigma}$ ($d^{\dagger}_{k,\sigma}$) is an annihilation (creation) operator, n^{e}_{k} (n^{i}_{k}) is an occupation number of electrons (ions) and s^{z}_{k} (S^{z}_{k}) represents a z-component of spin of electron (ion). The on-site interaction between ions and 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 is set equal to one, so we measure all energies in units of t.

The model (1) is similar to the famous *double exchange* model, sometimes referred to as *the ferromagnetic Kondo* or *s–d* model [2–5]. A simplified version of (1) is also considered with the name of *the Ising spin–fermion* model [6].

Some properties of the model (1) were examined in Refs. [7, 8]. Using the restricted phase diagrams method (RPDM) a few simple rules of formation of various sorts of ground state phases were detected [8]. In particular, relationships between density of electrons ρ_e and type of charge and magnetic arrangement of the ions with the density ρ_i were determined in the mixed valence regimes, i.e. for $\rho_e + \rho_i = 1$ and $\rho_e + \rho_i = 2$. In these two cases only axial stripes (vertical or horizontal) were found for intermediate values of the coupling constants. They are composed of ferromagnetic chains (when $\rho_e + \rho_i = 1$) or antiferromagnetic chains (when $\rho_e + \rho_i = 2$) interchanged with non-magnetic ones. For band fillings close to the half filling stripe phases oriented along one of the main diagonal direction are formed.

However, the previous studies do not clarify in a satisfactory a way the formation of phases that appear along the diagonal line $\rho_e + 2\rho_i = 2$ (see Fig. 1) on the canonical ground state phase diagram [8]. The topic is important, as for large values of U periodic phases in diluted magnets are stable only along the diagonal, whereas out of the diagonal mixture phases have lower energy. This is in analogy with the situation found for the spinless FKM where for large values of U periodic phases are found only along the diagonal line, that is then $\rho_e + \rho_i = 1$ [9]. What is more, it appears that charge distributions found in phases located along the diagonal $\rho_e + 2\rho_i = 2$ in diluted magnets and along the diagonal $\rho_e + \rho_i = 1$ in the spinless FKM are the same, at least for J small enough. It comes from the fact, that in the limit $J = 0$ the subsystem of spin up and spin down electrons are equivalent to each other and could be treated separately as spinless electrons. Until now, however, no rules of spin arrangement in diluted magnets have been detected.

Thus, our purpose here is to examine both charge and magnetic order in ground state phases of diluted magnets under the condition $\rho_e + 2\rho_i = 2$. We expect our results could have relevance in some doped nickelate and manganite oxide systems (see for example Ref. [10] and

the references given there), in which a replacement of any magnetic transition ion with a non-magnetic ion implies an increase of a number of itinerant electrons by two.

II. SCHEME OF CALCULATIONS

We performed calculations in 2D (the square lattice) using the RPDM [8] within the configurational space restricted to periodic phases with unit cells containing up to 12 lattice sites. To assure stability of phases appearing on the diagrams, we first constructed *the grand canonical phase diagrams* in the plane of chemical potentials μ_e and μ_i of the electrons and ions, respectively. Then we transformed the diagrams into *the canonical phase diagrams* in the plane of densities ρ_e and ρ_i.

In order to calculate the Gibbs thermodynamic potential, we determined the electronic band structure for the electrons for each candidate periodic phase. We employed a sufficiently tiny grid in the Brillouin zone (up to $N_c = 80 \times 80$ for each bandstructure). This required us to diagonalize up to 12×12 matrices at each discrete momentum point in the Brillouin zone and results in at most 12 different energy bands.

We performed all the calculations separately for spin up and down itinerant electrons. The eigenvalues of the band structure are summed up to determine the ground-state energy for each density of the electrons. Then, the Gibbs thermodynamical potential for a given configuration $\{w_i\}$ of ions is calculated for all possible values of the chemical potentials μ_e and μ_i, through the formula

$$G_{\{w_i\}} = \frac{1}{N} \sum_{\varepsilon_\uparrow, \varepsilon_\downarrow < \mu_e} (\varepsilon_\uparrow(\{w_i\}) + \varepsilon_\downarrow(\{w_i\})) - \mu_e \rho_e - \mu_i \rho_i \quad (2)$$

where the symbol $\varepsilon_\uparrow(\{w_i\})$ ($\varepsilon_\downarrow(\{w_i\})$) denotes energy eigenvalues of a band structure attributed to spin up (down) electrons for a given configuration $\{w_i\}$.

III. RESULTS AND DISCUSSION

Charge order, i.e. distribution of magnetic ions with no regard to their spins, depends mainly on U and for small enough J it is consistent with the charge order in the spinless case [9]. The correspondence occurs if the total density of electrons ρ_e (sum of densities of spin up and spin down electrons) is two times bigger than the density of spinless electrons.

In this paper we focus on physically relevant ranges of the interaction parameters $U \geq 4$ and $J \leq 1$. Then, it appears that the charge order of most of periodic phases located along the diagonal line $\rho_e + 2\rho_i = 2$ can be defined by one dimensional unit cells and translation vectors that are not perpendicular to each other.

The main finding is that almost all magnetically ordered phases have also one-dimensional unit cells and majority of them are just doubled unit cells related to their charge order. The antiferromagnetic order is predominat, but ferrimagnets are also present, as for example the phase d′.

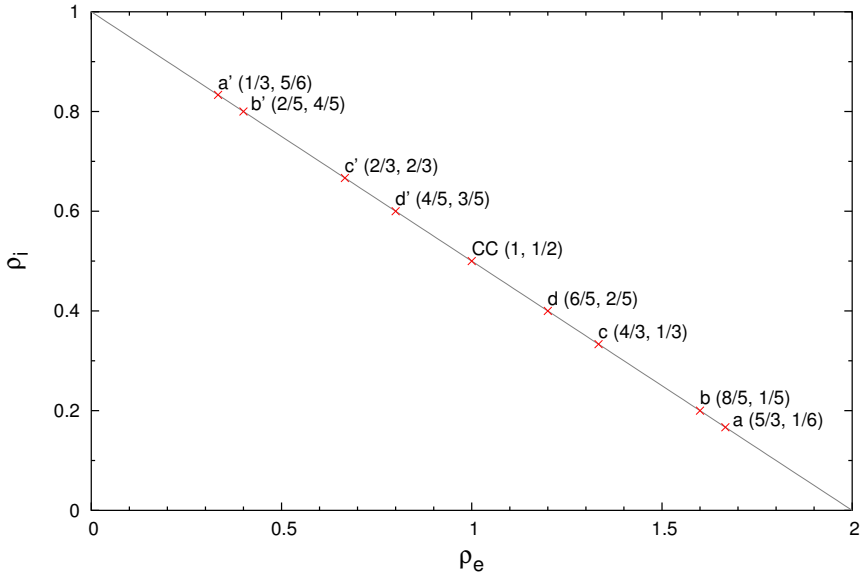

Fig. 1. The restricted ground state phase diagram of the FKM with Hund coupling with marked positions (with the crosses) of arbitrarily chosen characteristic periodic phases CC, a, b, c, d, a′, b′, c′, d′ discussed in this paper and displayed in the following figures. All the phases are located along the line $\rho_e + 2\rho_i = 2$. The numbers given in the parentheses denote the densities attributed to a given phase, ρ_e and ρ_i, respectively.

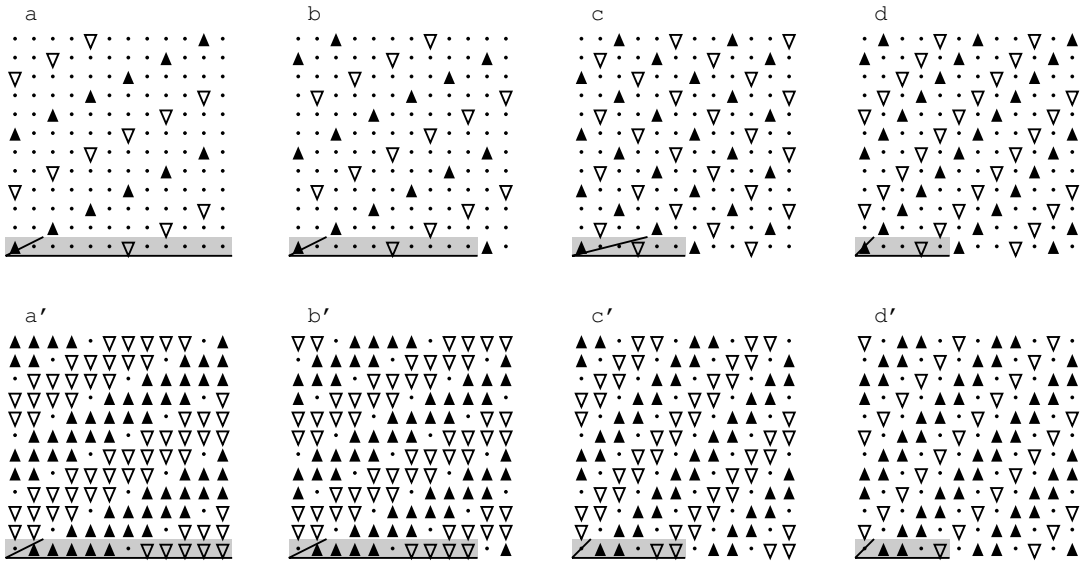

Fig. 2. Periodic phases discussed in this paper and located along the line $\rho_e + 2\rho_i = 2$ on the phase diagram displayed in Fig. 1. The symbol ▲ (▽) denotes spin up(down) *magnetic ion* and the dot denotes *nonmagnetic ion*. The shaded rectangular in the left bottom part of the picture marks a unit cell of the corresponding phase and the straight line segments mark the translation vectors.

In Fig. 2 we display a representative set of four configurations a, b, c, d with $\rho_i \leq 0.5$ and conjugated to them configurations a′, b′, c′, d′ with $\rho_i \geq 0.5$ that are present on the diagram in Fig. 1. Pairs of phases denoted by the same letter with no upper index or supplied with the prime upper index, as for example a and a′ are placed symmetrically with respect to the point $(\rho_e = 1, \rho_i = 0.5)$, where the checkeboard configuration CC displayed in Fig. 3 is stable.

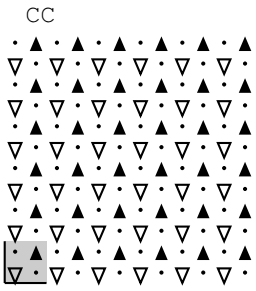

Fig. 3. The checkerboard ground state configuration *CC* stable for $\rho_e = 1$ and $\rho_i = 0.5$. See the caption to Fig. 2 for more explanations.

As for a charge order only, the symmetric position of phases belonging to the same pair implies also the hole-particle symmetry between them in that sense that sites occupied by magnetic and nonmagnetic ions are interchanged in the two phases. However, the symmetry is broken when magnetic degrees of freedom are included. Indeed, an ambiguity emerges when nonmagnetic ions are replaced by magnetic ones, as either spin up or spin down ions can appear. On the other hand, a replacement of magnetic ions into nonmagnetic is unambiguous.

Perhaps the most prominent effect of the broken symmetry is that some phases have uncompensated magnetic moment (they are ferrimagnetic) as for example the phase d′, even though its counterpart, the phase d, is antiferromagnetic. The reason is, that there is an odd number of magnetic ions in the unit cell of d′, while even number of ones in the unit cell of d.

It is quite interesting that almost all phases we found along the line $\rho_e + 2\rho_i = 2$ have the following common features: a) all magnetic ions located at the nearest neighbouring sites inside an unit cell form clumps with the same spin orientation; b) the consecutive clumps separated by nonmagnetic ones have opposite spins. If the clumps are symmetric, as in the case of the confiurations a, b, c, a′, b′ ,c′, d (see Fig. 2), then the system is antiferromagnetic and its magnetic unit cell is just doubled charge unit cell. On the other hand, if the clumps are asymmetric, as in the case of the configuration d′, the system has uncompensated magnetic moment (it is ferrimagnetic).

In general, one can notice a strong tendency to antiferromagnetic ordering, because energy spectra of spin up and spin down electrons are then the same and every eigenstate is double occupied by two electrons with opposite spins. In contrast, spin polarized states with ferromagnetic order make a relative displacement of the energy spectra. Consequently, the lowest energy eigenstates are then merely single occupied by spin-polarized electrons and the total energy increases when the total number of electrons is preserved.

All phases discussed in this paper can be viewed as stripe phases composed of magnetic chains separated by nonmagnetic ones that are parralel to them. At half filling ($\rho_e = 1$) and close to it the chains have the slope α against the horizontal line equal to $\pi/4$ (see the configurations CC, c, d, c′ d′), whereas far from half filling α reduces (see the configurations a, b, a′ b′). The magnetic chains are ferromagnetic for almost all phases and

only for the most symmetric configuration CC and the configuration c they are antiferromagnetic. In fact, if one look at the configuration c along another direction that characterize its structure, one can see that it also can be viewed as beeing composed of ferromagnetic chains. It appears that configurations with $\rho_d \leq 1$ consist of ferromagnetic chains along one direction and at the same time they consist of simple antiferromagnetic chains along the other direction. The only configuration that is antiferromagnetic along the two diagonal directions is CC.

In summary, we analyzed a family of ground state phases of diluted magnets located on the phase diagram along the line $\rho_e + 2\rho_i = 2$, that are only stable periodic phases for large values of the Coulomb coupling U. Most of the phases are arranged antiferromagnetically, even though the assumed on-site Hund coupling is ferromagnetic. The phases form non-axial stripes composed of ferromagnetic and, occasionally, also antiferromagnetic chains non-parallel to the main lattice axes. Magnetic unit cells are usually one-dimensional and composed of two adjacent charge unit cells.

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ЗВ'ЯЗОК МІЖ ЗАПОВНЕННЯМ ЕЛЕКТРОННОЇ ЗОНИ ТА ТИПОМ ЗАРЯДОВОГО Й МАГНІТНОГО ВПОРЯДКУВАННЯ

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Досліджено зарядові та спінові впорядкування на квадратній ґратці в межах узагальненої моделі Фалікова–Кімбала з хундівською взаємодією між локалізованими та рухомими електронами. Використовуючи метод обмежених фазових діаграм, ми виявили для розведеного магнетика кілька простих правил формування єдиної сім'ї періодичних фаз основного стану, що є стабільними як для великих, так і для проміжних значень параметра зв'язку. Виявилося, що для цієї групи фаз формуються лише неаксіальні смуги, що складаються з феромагнітних або деколи також з антиферомагнітних ланцюжків, орієнтованих уздовж одного з двох головних діагональних напрямків або вздовж іншого напрямку, який є непаралельним до головної кристалічної осі. Антиферомагнітне впорядкування переважає у цих фазах, але деколи також трапляються феромагнітні фази.