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Phase Separation in the Ground State of the Model 2D Spin-Pseudospin System

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We addressed a simplified static 2D spin-pseudospin model which takes into account both conventional Heisenberg spin exchange coupling and the on-site and inter-site charge correlations. Classical Monte-Carlo calculations for large square lattices show that homogeneous ground state solutions found in a mean-field approximation are unstable with respect to phase separation with the charge and spin subsystems behaving like immiscible quantum liquids. For instance, with lowering the temperature one can observe two sequential phase transitions: first, antiferromagnetic ordering in the spin subsystem diluted by randomly distributed charges, then, the charge condensation in the charge droplets. Thermodynamic properties and phase diagram of the 2D spin-pseudospin system are studied by Monte-Carlo simulation with a special attention given to the role played by the on-site correlation.

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1. Introduction

Models with competing or intertwining order parameters are popular in the condensed matter theory in connection with such real systems as, for example, multiferroics or high-T_c cuprate superconductors. In cuprates, the competition of static magnetic order, bulk superconductivity and charge-density waves has attracted a lot of attention over the years, but its nature remains a challenge [1]. Earlier we suggested a simplified static 2D spin-pseudospin model [2, 3] which takes into account both conventional Heisenberg spin exchange coupling and the on-site and inter-site charge correlations. A detailed qualitative and quantitative analysis of the spin-charge competition within the model with the ground state (GS) and temperature phase diagrams was done in the mean field approximation (MFA). Here, in the paper, we present the results of classical Monte-Carlo (MC) calculations in a "strong" exchange limit. An interactive visualization of the actual states of the system allowed us to observe qualitatively different behavior of doped charges in the charge ordered (CO) and antiferromagnetic (AFM) phases with a clear trend to a bulk spin-charge phase separation in the AFM phase and a random distribution of the doped charges in the CO phase.

2. The model

In our model approach [4] to copper oxides such as $La_{2-x}Sr_xCuO_4$ we assume that the on-site Hilbert space is reduced to only three effective charge states (nominally $Cu^{1+;2+;3+}$) of copper ions in the CuO_2 planes. These charge states are associated with components of the $S = 1$ pseudospin triplet with $M_S = -1, 0, +1$. The on-site states are characterized by different hole occupation: $n_h = 0, 1, 2$ for $Cu^{1+;2+;3+}$, respectively, and different conventional spin: $s = 1/2$ for Cu^{2+} and $s = 0$ for $Cu^{1+;3+}$. The doped hole concentration n are related with the pseudo-magnetization: $nN = \sum_i \langle S_{iz} \rangle$. Conventional spin density for mixed valence superpositions can vary inbetween 0 and 1 in accordance with the weight of the Cu^{2+} in the on-site superposition.

Hereafter, in the paper we shall consider only simplified spin-pseudospin Hamiltonian which takes into account the on-site and inter-site correlations, and conventional Heisenberg spin exchange coupling:

$$\mathcal{H} = \Delta \sum_{i=1}^{N} S_{iz}^2 - \mu \sum_{i=1}^{N} S_{iz} + V \sum_{\langle ij \rangle} S_{iz} S_{jz} + J \sum_{\langle ij \rangle} P_{0i} \boldsymbol{s}_i \boldsymbol{s}_j P_{0j}, \quad (1)$$

where the sums run over sites of a two-dimensional square lattice, $\langle ij \rangle$ means the nearest neighbors, S_{iz} and $\boldsymbol{s}_i$ are the on-site pseudospin and conventional spin operators, respectively. The first on-site term with Δ relates with the on-site density-density interactions, the second term with chemical potential μ is needed to account for the charge density constraint, $n = \text{const}$, the third term with $V > 0$ describes the effects of the inter-site density-density interactions. The last term is the antiferromagnetic ($J > 0$) Cu^{2+}–Cu^{2+} Heisenberg spin exchange coupling, where the projection operator $P_{0i} = 1 - S_{iz}^2$ takes into account the on-site occupation dependence. The most important limitation of model is the static character of the charge and spin subsystems, since the Hamiltonian does not contain any transfer terms.

The MFA analysis [3] gives five GS solutions or phases of spin-pseudospin system. The energies and structural characteristics in terms of the on-site charge and spin densities for these phases are given in Table I.

In a "weak" exchange limit, at $J/4 < V$, all the GS phases (COI, COII, COIII, FIM) correspond to the var-

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TABLE I

The mean energy ε, on-site charge density $\langle S_z\rangle_j$ and on-site spin density $\langle P_0\rangle_j$ of the MFA GS phases for 2D spin-pseudospin system. The index $j = 0, 1$ distinguishes two checkerboard sublattices. $k = (1 - |n|)$, $h = (1 - 2|n|)$.

Phase	$\varepsilon = \langle\mathcal{H}\rangle/N$	$\langle S_z\rangle_j$	$\langle P_0\rangle_j$
COI	$\Delta - 2Vh$	$n + (-1)^j k$	0
COII	$\lvert n\rvert\Delta - 2Vh$	$n + (-1)^j k$	$k(1 - (-1)^j \operatorname{sgn} n)$
COIII	$k\Delta - 2Vh$	$n + (-1)^j k$	$\lvert n\rvert - (-1)^j n$
FIM	$\lvert n\rvert\Delta - \frac{J}{2}h$	$n + (-1)^j \lvert n\rvert$	$1 - \lvert n\rvert - (-1)^j n$
AFM	$\lvert n\rvert\Delta - \frac{J}{2}k^2 + 2n^2V$	n	$1 - \lvert n\rvert$

ious types of charge order. The COI is a charge-ordered phase without spin centers. In the COII and COIII phases the charge order is diluted by the non-interacting spins. In the FIM phase charge and spin orders coexist. In a "strong" exchange limit, at $J/4 > V$, there are only COI and AFM phases.

3. Numerical results

Here we present some results of classical MC calculations in a"strong" exchange limit ($J/4 > V$) with the heat-bath algorithm on the square lattice 256×256 under periodical boundary conditions. In numerical calculations, we used the Ising type spin-spin interaction with the same account of the on-site occupation dependence as in (1). As an initial state, we choose the random distribution of pseudospins and spins with a fixed total z-component of pseudospins for a given value of n. We implemented high-performance parallel computing on NVIDIA graphics cards and an interactive visualization of the actual states of the system. This allows us to observe the relaxation of the system to the ground state in the process of calculation.

First we address the on-site correlations $\Delta < 0$ when these stabilize the GS COI phase. In Fig. 1 the temperature dependence of the specific heat $C(T)$ and snapshots of the real states of the system at some characteristic points are shown for $\Delta = -1.5$ and $n = 0.1$. The $C(T)$ dependence reveals the maximum near $T/J \approx 0.22$. The plateau at $T/J \approx 0.5$ is related with a freezing of the spin subsystem. A direct observation of the state of the system shows that the spin excitations mostly disappear at $T/J \approx 0.5$ before the ordering in the spin subsystem occurs. Qualitatively this part of the $C(T)$ dependence can be described within a rough approximation of free charge and spin doublets. In this case $V = 0$ and $J = 0$, so taking into account the charge density constraint we come to the expression for the specific heat as follows:

$$C = \frac{\Delta^2 \mathrm{e}^{\Delta/T}\left(1 - n^2\right)^2}{T^2 f\left(f + \mathrm{e}^{\Delta/T}\right)^2}, \qquad (2)$$

where $f = \sqrt{1 - n^2 + n^2 \mathrm{e}^{2\Delta/T}}$. The $C(T)$ dependence (2) is shown in Fig. 1 by the dashed line 1 for $n = 0.1$ and $\Delta = -1.5$.

The $C(T)$ peak at $T/J \approx 0.22$ is related to the charge ordering. The snapshots *a,b* clearly demonstrate this feature. The temperature of the charge ordering nearly corresponds to the Ising value $T^* = 2V/\log(1 + \sqrt{2}) \approx 2.26$ V. With increasing of the charge doping this maximum is rapidly reduced and almost disappears above the $n \approx 0.3$. The distribution of doped charges over the CO matrix remains random with the temperature decrease, so the energy of the low temperature state is exactly equal to the MFA GS energy.

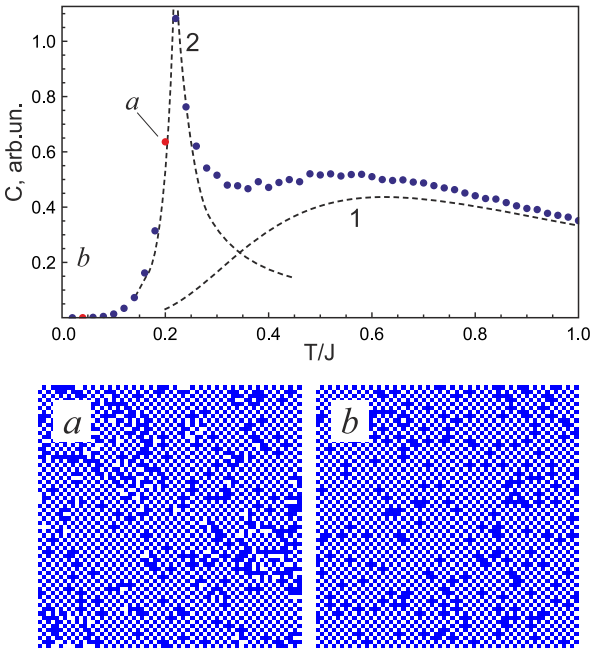

Fig. 1. The MC calculated temperature dependence of specific heat and snapshots of real states of 2D lattice for the CO phase at $n = 0.1$, $\Delta = -1.5$, $V = 0.1$, $J = 1$. The dashed line 1 corresponds to the "free doublets" approximation (2) for $n = 0.1$ and $\Delta = -1.5$. The dashed line 2 corresponds to the Ising type dependence of specific heat for the charge subsystem at given $V = 0.1$. In the snapshots of real states of a lattice in the CO phase blue and white colors correspond to the on-site value of $\langle S_z\rangle = \pm 1$, respectively. Comparison of the snapshots a and b points to a weak temperature dependence of the random character of the doped charge distribution over the CO matrix.

Qualitatively different temperature behavior one observes for the on-site correlations $\Delta > 0$ when these stabilize the GS AFM phase. Temperature dependence of the specific heat for the AFM phase at $n = 0.1$ is shown in Fig. 2. The sharp maximum at $T/J \approx 0.45$ corresponds to the AFM ordering of the spin subsystem. The temperature of the ordering nearly corresponds to the value $T^\star \approx 2.26(1 - n^*)J/4$, where $n^* \approx 0.2$ is a total concentration of the doped and the excited charge centers. This maximum is rapidly flattened with the charge doping. When the temperature is lowered, the specific heat demonstrates second peak at $T/J \approx 0.08$. The snapshots a and b show that this puzzling peculiarity is related with a condensation of doped charges in the charge droplets. Phase separation in the AFM phase exists for a whole range of the doped charge concentrations except

for $n = 0$ and $n = \pm 1$. The calculated energy of the low temperature phase separated state is lower than the MFA GS energy of AFM phase, as the surface energy effects are omitted in MFA.

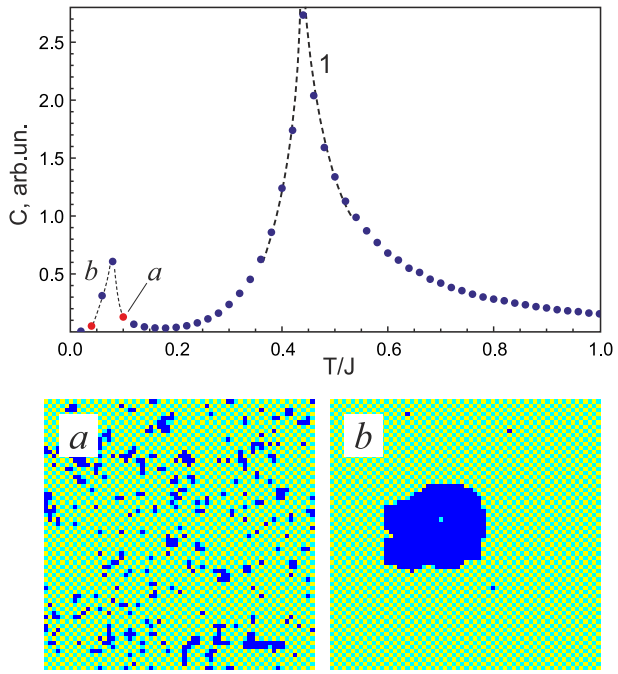

Fig. 2. The temperature dependence of the specific heat and snapshots of real states of 2D lattice for the AFM phase at $n = 0.1$, $\Delta = 0.5$, $V = 0.1$, $J = 1$. The Ising type high-temperature peak corresponds to the AFM ordering in spin subsystem at $T^{\star} \approx 0.45$ (dashed line 1). The low-temperature peak 2 corresponds to the charge droplet condensation, shown in a and b snapshots. Blue color points to doped charge distribution, $\langle S_z \rangle = 1$, yellow and green colors correspond to the on-site spin values: $\langle s_z \rangle = \pm 1/2$, respectively.

We have performed classical MC calculations for the 2D spin-pseudospin system with competing CO and spin AFM orders in a"strong" exchange limit. The behavior of the system strongly depends on the sign and value of the on-site correlation parameter Δ stabilizing CO ($\Delta < 0$) or AFM ($\Delta > 0$) phase, respectively. We show that homogeneous ground-state AFM solutions found in the MFA [1] are unstable with respect to phase separation with the charge and spin subsystems behaving like immiscible quantum liquids. The specific heat temperature dependence reveals two sequential phase transitions: first, antiferromagnetic ordering in the spin subsystem diluted by randomly distributed charges, then, the charge condensation in the charge droplets. The inhomogeneous droplet phase reduces the energy of the system and changes the diagram of the GS. Charge doping does suppress the long-range spin order, but the phase separation of doped charges and short-range spin order exists for a whole range of the charge doping. Specific heat for the system with the GS COI phase shows a feebly marked maximum due to a spin freezing at elevated temperatures with a low-temperature singular peak due to the charge ordering. The doped charges remain distributed randomly over the CO matrix up to $T = 0$ as for the near-neighbor interaction the energies of all possible distributions of extra charges over the CO matrix are equal. For this reason the GS energy of the COI MFA solutions exactly matches the energy of the low-temperature MC state and the entropy of the low-temperature state in the doped CO phase is higher than in the doped AFM phase.

Acknowledgments

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