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Monte Carlo investigation for an Ising model with competitive magnetic interactions in the dominant ferromagnetic-interaction regime

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Abstract. *We apply classical Monte Carlo simulation to examine the thermodynamic properties of perovskites described by the Ising model with competitive magnetic interactions. By correspondingly adjusting the ferromagnetic-interaction and antiferromagnetic-interaction probabilities, p and $(1-p)$, in the regime $p \geq 0.5$, the temperature dependence of magnetization, total energy, spin susceptibility, and specific heat consistently show a ferromagnetic to paramagnetic (FM-PM) phase transition at a critical temperature T_c. Besides, the inverse susceptibility is confirmed to follow Curie-Weiss's law above another critical temperature T_{CW}. By increasing the FM interaction probability, we have observed both FM-PM critical temperature T_c and Curie-Weiss temperature T_{CW} shifted to the higher values but the difference between them seems to slightly reduce.*

Keywords: Monte Carlo simulation; phase separation; magnetic interactions; Ising model.

Classification numbers: 64.75.Qr; 75.10.Hk; 87.55.K; 88.10.gc; 13.40.-f.

1. Introduction

The perovskite metal oxides have revealed rich phase diagrams based on various magnetic and electrical characteristics [1]. By chemical doping, the coexistence of multi-valent ions for example, Co^{3+} and Co^{4+} in perovskite cobaltites induces inhomogeneous magnetic interactions. The competition between antiferromagnetic (AF) interaction between similar valence ions through super-exchange mechanisms and ferromagnetic (FM) interaction between different valence ones through double-exchange mechanism brings many different magnetic phases like spin-glass, magnetic clusters, [1–3] and Griffith-like phases [4] with interesting structural, magnetic and electrical properties such as colossal magneto-resistance, Jahn-Teller distortion, metal-insulator transition, unconventional superconductivity, etc. [5].

The Ising model with random magnetic interactions, one of the simplest theoretical models, has successfully described phase transitions for perovskites having competitive magnetic interactions. Monte Carlo simulations for an Ising model including both AF nearest-neighbor (n.n.) and next-nearest-neighbor (n.n.n.) exchange interactions have shown a phase diagram closely related to experimental results for those perovskites [6]. The formation of FM- and AF-ordered clusters results from competitive magnetic interactions due to spin alignments to minimize total energy at temperatures lower than a critical temperature.

We can approach the magnetic system with inhomogeneous ordered clusters (mixed FM- and AF-ordered clusters) by using another version of the Ising model with random FM and AF interactions as successfully applied in Refs. [7–9]. Following these works, we can directly control the probability of FM and AF interaction bonds and the magnitude of these exchange interactions as input parameters of the models. These works displayed a phase diagram revealing a re-entrant magnetism phenomenon and the step behavior of magnetization and magneto-resistance in the application of external fields experimentally observed in various polycrystalline specimens [10, 11].

It is previously mentioned that the phase diagram of perovskites with competing magnetic interactions includes regions with mixed AF and FM clusters at temperatures lower than the critical temperature T_c where the whole systems sustain a ferromagnetic order with a finite average magnetization. However, many recent works show that a preformed-cluster state may exist above the T_c related to the well-known Griffith phase [12, 13]. The magnetization, specific heat, and spin susceptibility of this system behave differently in these regions when the systems are subjected to an external magnetic field. For this reason, we want to deeply examine and analyze the thermodynamic behaviors of these mixed-magnetic clusters in light of the disorder Ising model.

2. Monte Carlo simulation and disordered Ising model

A typical Ising model includes both FM and AF interactions with a Hamiltonian described in Ref. [8], which is

$$H = -\sum_{<i,j>} J_{ij} S_i^z S_j^z - h\sum_k S_k^z, \tag{1}$$

where S_i^z is the z-component of spin at the i^{th} site, h is an external magnetic field. The first term composes of both anti-ferromagnetic and ferromagnetic interactions expressed by an exchange interaction J_{ij} between nearest-neighbor spins, which follows a probability rule:

$$P(J_{ij}) = p\delta(J_{ij} - J_{FM}) + (1-p)\delta(J_{ij} - J_{AF}). \tag{2}$$

J_{FM} and J_{AF} presented in Fig. 1 are, respectively, ferromagnetic and anti-ferromagnetic exchange interactions adjusted by a fluctuation parameter Δ. Their explicit forms are written by $J_{FM} = J(1+\Delta)$ and $J_{AF} = J(1-\Delta)$ with a constant J. Since we are interested in materials with AF and FM-ordered coexistence, Δ is taken greater than 1. In this work, we set $J = 1$ as an energy unit

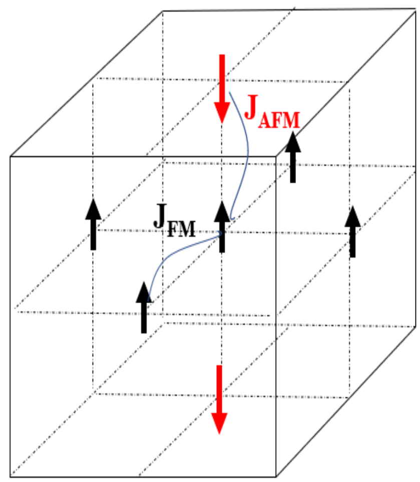


Fig. 1. Cartoon picture illustration for a three-dimensional spin lattice with both ferromagnetic (J_{FM}) and antiferromagnetic (J_{AFM}) exchange interactions between nearest-neighbor spins.

where the external field h and temperature T are considered. We also set $k_B = 1$ for numerical simplicity.

In order to solve this model, we applied a classical Monte Carlo simulation with the Metropolis algorithm [14,15] for a cubic spin-lattice having total spin numbers $V = L \times L \times L$ with its length L. The number of Monte Carlo steps, which is taken as 3×10^6, is good enough to keep our system stable. Some examined thermodynamic quantities include the total energy per spin, which is

$$E = \frac{< H >}{V}, \tag{3}$$

the magnetization per spin,

$$M = \frac{< \sum_{i=1}^{V} S_i^z >}{V}, \tag{4}$$

the magnetic susceptibility per spin,

$$\chi = \frac{< M^2 > - < M >^2}{k_B T}, \tag{5}$$

and the specific heat,

$$C_V = \frac{< E^2 > - < E >^2}{k_B T^2}. \tag{6}$$

Since the finite size effect is non-negligible in phase transition problems, we also check the consistency of simulation calculations by changing the lattice size V shown in Fig. 2. The magnetic phase transition normally occurs at a critical Curie temperature below which the magnetization treated as an order parameter in the Landau theory of phase transition [16] is finite. However, a finite magnetization can be spontaneously formed in a small-size 3D lattice as observed in Fig. 2(a)

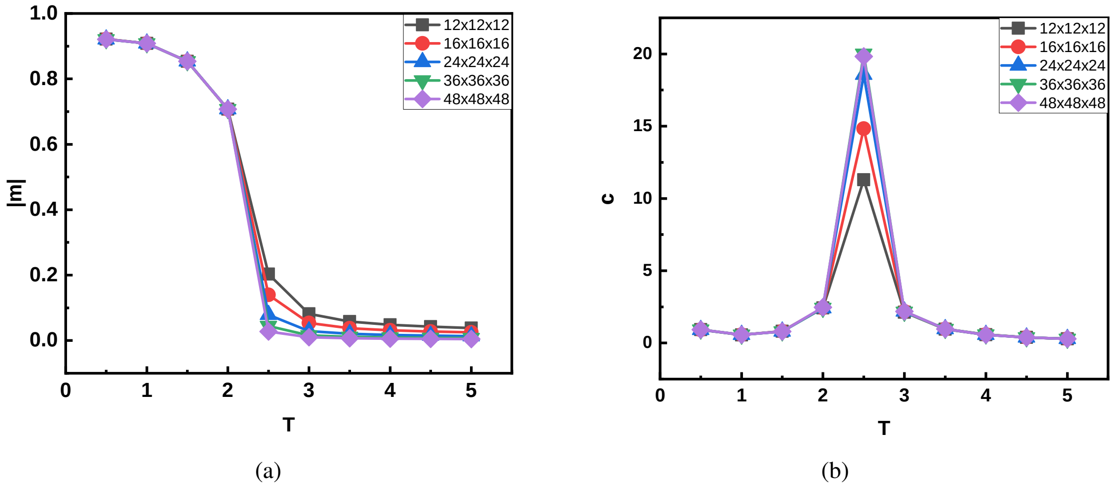


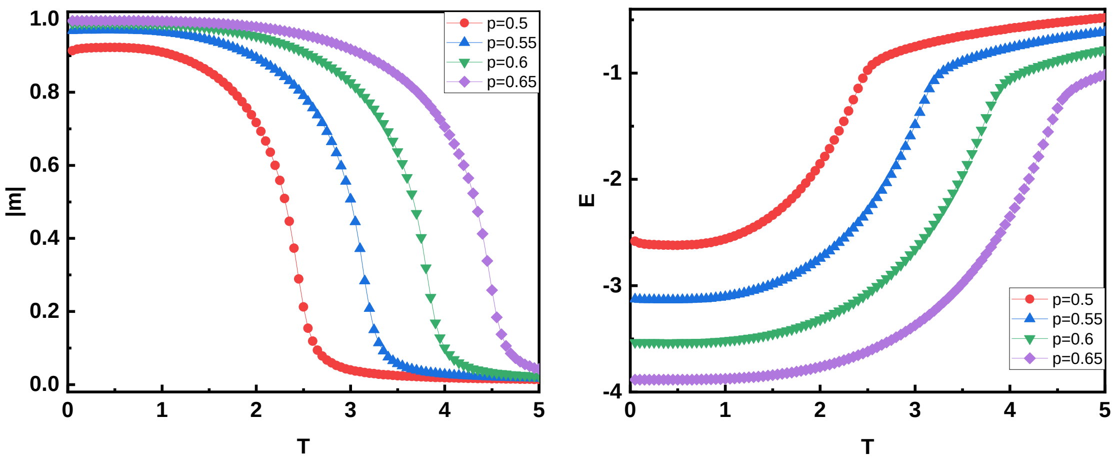
Fig. 2. The absolute value of magnetization (a) and the spin susceptibility (b) versus temperature with different lattice sizes. Here $h = 0$, $p = 0.5$, $\Delta = 1.01$.

with $V = 12 \times 12 \times 12$. Furthermore, we can see that the magnetization and the maximum peak position of spin susceptibility insignificantly modify when the lattice size is up to $V = 24 \times 24 \times 24$. Due to time-saving reasons, our simulation measurements in the following parts are carried out with the lattice size $V = 24 \times 24 \times 24$ which ensures enough reliability to determine physical quantities and the critical temperature.

3. Calculation results and discussions

The most simplified example of the phase diagram of doped perovskites such as $La_{1-x}Sr_xCoO_3$ (LSCO) is possibly referred to Refs. [2, 3] where a spin-glass behavior was realized at low Sr doping while short-range ferromagnetic clusters and phase separation were found out at high Sr concentration. The presence of FM clusters and phase separation originate from the competition between super-exchange and double-exchange interaction. It is worth noting that the effect of the AF and FM interactions are parameterized in our model by two factors which are (i) the magnitude of exchange interactions where the FM magnitude $J_{FM} = J(1+\Delta)$ is presumably greater than the AF magnitude $J_{AF} = J(1-\Delta)$ since $\Delta > 1$ and (ii) the probabilities of FM and AF interaction. The random distribution of FM and AF interactions in the materials can be controlled by the probabilities p and $(1-p)$ in our model, respectively.

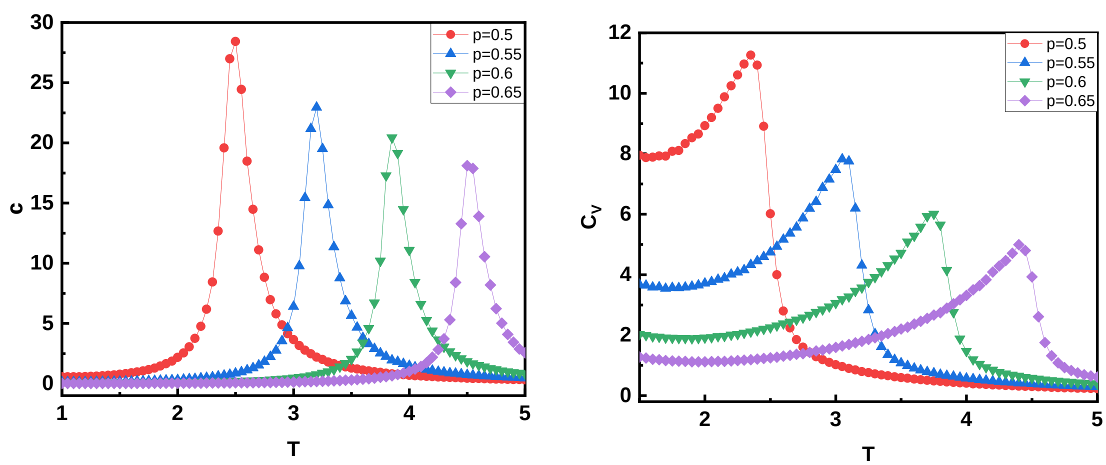
At first, we investigate the variation of magnetization at different probabilities p in a wide range of temperatures illustrated in Fig. 3. Increasing the probability p greater than 0.5 implies the dominant occurrence of the FM interaction. The consequence of AF and FM order competition is the occurrence of FM short-range clusters embedded in AF clusters which is very sensitive to temperature change. The increase of FM interaction probability p is expected to enhance the magnetization at low temperatures, which is consistent with its behavior obtained in Fig. 3(a). The different finite values of magnetization at very low temperatures indicate the inhomogeneous spin texture in the systems having phase separation as observed in various experimental works



(a) (b)

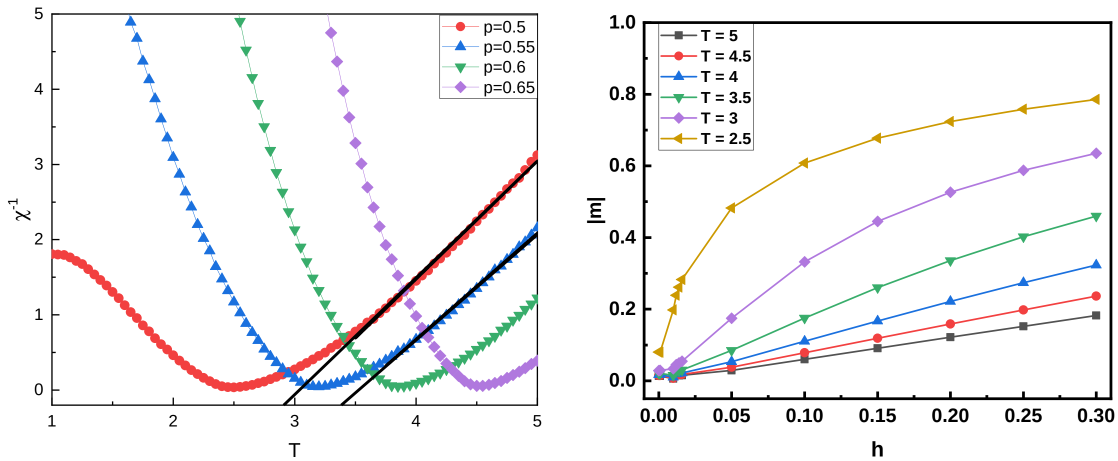
Fig. 3. The temperature dependence of the absolute value of magnetization with different probabilities p. Here $h = 0.01$, $\Delta = 1.01$.

[3, 13, 17]. Besides, the critical transition temperature T_c from the dominant FM to PM phase is also shifted to the higher temperature while increasing the FM probability as observed in various experiments. We also present in Fig. 3(b) the temperature dependence of average energy with the same probability values taken in Fig. 3(a). The lowered energy is agreeable to the increase of the FM exchange interaction bonds due to the inverse relationship between magnetization and total energy defined by Eq. (1-4).



(a) (b)

Fig. 4. The temperature dependence of spin susceptibility (a) and specific heat (b) with different probabilities p. Here $h = 0.01$, $\Delta = 1.01$.



(a) (b)

Fig. 5. (a) The temperature dependence of inverse spin susceptibility (a) with different probabilities p at $h = 0.01$ and $\Delta = 1.01$; (b) The variation of magnetization in the magnetic field at different temperatures at $p = 0.5$, $\Delta = 1.01$.

Another important question is how to define the critical temperature T_c in the ferromagnetic-paramagnetic (FM-PM) phase transition. Therefore, we examine the behavior of the spin susceptibility and the specific heat under a certain external magnetic field presented in Fig. 4. In general, the FM-PM phase transition temperature T_c corresponds to the position of the maximum peak either in the susceptibility $\chi(T)$ or in the specific heat curve clearly observed in Fig. 4. At a specified probability p, these peaks occur at the same critical temperature which is also extracted in the experimental observation in the same way.

Above the critical temperature T_c, the spin susceptibility χ in the paramagnetic phase may follow Curie-Weiss' law which is

$$\chi = \frac{1}{T - T_{CW}}, \tag{7}$$

with the Curie-Weiss temperature T_{CW}. According to our understanding, the FM-PM critical temperature T_c and the Curie-Weiss temperature T_{CW} are not always identical, particularly in the competing interaction systems. Experimental observations show that the Curie-Weiss temperature T_{CW} is generally larger than the FM-PM critical temperature T_c, thus the question of what really occurs above T_c and under T_{CW} has not been clearly answered yet. For this reason, we use a linear-temperature fitting of the inverse susceptibility to estimate the temperature T_{CW} as observed in Fig. 5(a). This fitting shows a good agreement with Curie-Weiss' law and reveals the shift of T_{CW} to the larger value with the increase of the FM interaction probability, which is consistent with the other thermodynamic quantity behavior. These two temperatures are also listed in Table 1.

As we can see, the gap between the critical FM-PM phase transition temperature T_c and the Curie-Weiss temperature T_{CW} slightly reduces when increasing FM interaction probability p. Since this difference is kindly marginal, it is hard to determine if the reason is because of either a numerical artifact or the increasing phase homogeneity of the system with increasing FM interaction.

To emphasize the field effect, we present in Fig. 5(b) the variation of magnetization behavior with the magnetic field at different temperatures. At sufficiently high temperatures $(T = 5)$ where the system is in the paramagnetic phase, a low external field vaguely induces the magnetization involving only single spin-flip processes. As a consequence, the field-dependent magnetization inherits a linear behavior. At lower temperatures, for example, $T = 2.5$ close to the critical temperature T_c, since the external field is comparable to thermal fluctuations, other magnetic processes including cluster formation, or cluster rotation can occur. Therefore, the linear dependence no longer maintains in the system as observed in both our simulations and experiments [2, 13].

4. Conclusions

We have presented the Monte Carlo simulation approach to examine the magnetic properties of the three-dimensional inhomogeneous systems having competitive magnetic interactions. The competition of the FM and AF exchange interactions in the system is successfully constructed by controlling their probabilities p and $1-p$ in the classical Ising model, respectively. In the regime of the dominant FM interactions, the Monte Carlo simulation results have shown the signals of phase separation expressed by the different finite magnetization and total energy at very low temperatures. Despite the presence of the mixed FM and AF clusters, the spin susceptibility in the paramagnetic phase behaves as the Curie-Weiss' law where the extracted Curie-Weiss temperature T_{CW} displays a gap with the FM-PM critical temperature Tc. The question about whether short-range FM- and AF-clusters coexist above Tc and below a crossover temperature and whether the crossover temperature directly relates to the Curie-Weiss temperature T_{CW} motivates our future research.

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Authors contributions

Oanh K.T. Nguyen: Resources, Methodology Investigation, Validation, Visualization. **Phong H. Nguyen**: Methodology, Investigation, Validation, Data Curation. **Niem T. Nguyen**: Resources, Investigation. **Huy D. Nguyen:** Visualization, Validation, Editing. **Cong T. Bach**: Validation, Conceptualization, Supervision. **Giang H. Bach**: Visualization, Formal analysis, Writing - Review - Editing, Funding acquisition.

Conflict of interest

The authors have no conflict of interest to declare.

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