Compensation temperature of 3d mixed ferro-ferrimagnetic ternary alloy

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In this study, we have considered the three dimensional mixed ferro-ferrimagnetic ternary alloy model of the type $\text{AB}_p\text{C}_{1-p}$ where the A and X (X=B or C) ions are alternately connected and have different Ising spins $S^A=3/2$, $S^B=1$, and $S^C=5/2$, respectively. We have investigated the dependence of the critical and compensation temperatures of the model on concentration and interaction parameters by using MC simulation method. We have shown that the behavior of the critical temperature and the existence of compensation points strongly depend on interaction and concentration parameters. In particular, we have found that the critical temperature of the model is independent on concentration of different types of spins at a special interaction value and the model has one or two compensation temperature points in a certain range of values of the concentration of the different spins.

I. INTRODUCTION

Molecular-based magnetic materials have recently attracted considerable interest and study of the magnetic properties [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17]. A special class of these materials, the so-called Prussian blue analogs, such as $(X^p\text{Mn}^{II}_{1-p})_{1.5}[\text{Cr}^{III}(\text{CN})_6]_{n}\text{H}_2\text{O}$ ($X^p=\text{Ni}^{II}, \text{Fe}^{II}$) [4, 5] and $(X^p\text{Mn}^{II}_{1-p}\text{Fe}^{II}_{1.5})[\text{Cr}^{III}(\text{CN})_6]_{n}\text{H}_2\text{O}$ [6] which exhibit many unusual properties, for instance, occurrence of one [4] or even two [6] compensation points, magnetic pole inversion [3, 7], the photoinduced magnetization effect [8, 9], inverted magnetic hysteresis [10]. These ternary alloys have ferromagnetic-ferrimagnetic properties since they include mixed both ferromagnetic ($J>0$) and antiferromagnetic ($J<0$) superexchange interactions between the nearest-neighbor metal ions. Theoretical investigations of these systems are difficult because of their structural complexity. However, to obtain magnetic properties of the molecular-based magnetic materials, up to now, these systems have been studied by using effective-field theory [11], mean field theory [12, 13, 14] and Monte Carlo simulation (MC) methods [15, 16, 17].

In this study we consider three dimensional ferro-ferrimagnetic $\text{AB}_p\text{C}_{1-p}$ ternary alloy, consisting of three different Ising spins $A=3/2$, $B=1$, and $C=5/2$, which correspon to the Prussian blue analog of the type $(X^p\text{Mn}^{II}_{1-p})_{1.5}[\text{Cr}^{III}(\text{CN})_6]_{n}\text{H}_2\text{O}$ [4]. In this system, the coupling Cr-Ni is ferromagnetic and Mn-Cr is antiferromagnetic. Our aim, in this study, is to clarify the effects of the concentration and the interaction parameters on the magnetic behavior of the three dimensional ternary alloy model by using MC simulation method.

II. THE MODEL AND ITS SIMULATION

Three dimensional ferro-ferrimagnetic $\text{AB}_p\text{C}_{1-p}$ Ising model consists of two interpenetrating cubic sublattices as seen in Fig. 1. It can be assumed that the A ions are located on the first cubic sublattice and the B and C ions are randomly distributed on the second cubic sublattice with the concentration $p$ and $1-p$, respectively. Also, to construct a Hamiltonian for this system, the ions A can be represented by spin $S^A$, and on the other hand, ions B and C can be represented by Ising spins $S^B$ and $S^C$, respectively. If the interactions between nearest neighbors can be chosen such as A ions ferromagnetically interact with B, on the other hand, antiferromagnetically interact with C ions, thus, spins of the Prussian blue analog of the type $(X^p\text{Mn}^{II}_{1-p})_{1.5}[\text{Cr}^{III}(\text{CN})_6]_{n}\text{H}_2\text{O}$ can be represented by this model where $S^A$, $S^B$, and $S^C$ correspond to Cr, Ni and Mn, respectively. In this study we also consider next-nearest neighbor interactions between spins $S^A$ .

The Hamiltonian of the considered system can be written in the form

$$H = -\sum_{\langle nn \rangle} S^A_i [J_{AB} S^B_j \varepsilon_j + J_{AC} S^C_j (1 - \varepsilon_j)] - J_{AA} \sum_{\langle nnn \rangle} S^A_i S^A_k$$

(1)

where $S^A = \pm 3/2, \pm 1/2$ for A, $S^B = \pm 1, 0$ for B and $S^C = \pm 5/2, \pm 3/2, \pm 1/2$ for C, on the other hand, $\varepsilon_j$ is a random variable which takes the value of unity if there is a spin X ($S^B$ or $S^C$) at the site $j$, if it not is zero. In Eq. (1), the first sum is over the nearest-neighbor and the second one is over the next-nearest neighbor spins. In this Hamiltonian the nearest neighbor interactions are chosen as $J_{AB} > 0$ and $J_{AC} < 0$, and the next-nearest neighbor interactions are chosen as $J_{AA} > 0$.

In order to show the effects of the concentration $p$ and the interaction parameters on the compensation and crit-
ical temperature of the three dimensional ternary alloy model, we simulate the Hamiltonian given by Eq. (1). To simulate this model, we employed Metropolis Monte Carlo simulation algorithm \[20\] to the \( L \times L \times L \) three-dimensional lattice with periodic boundary conditions for \( L = 10, 12, 16, 20, 24 \). One of the cubic sublattice is fully decorated with spin \( S^A \), and spins \( S^B \) and \( S^C \) are randomly distributed on the other cubic sublattice with the concentration \( p \) or \( 1 - p \), respectively. All initial spin states in the \( L \times L \times L \) three-dimensional lattice are randomly assigned. Configurations are generated by making single-spin-flip attempts, which were accepted or rejected according to the Metropolis algorithm. To calculate the averages, data, over 20 different spin configuration, is obtained by using 50000 Monte Carlo steps per site after discarding 10000 steps.

The sublattice average magnetizations per site are obtained by

\[
M_A = \frac{2}{L^3} \left( \sum_i S_i^A \right),
\]

\[
M_B = \frac{2}{L^3} \left( \sum_{j=1}^{N_B} S_j^B \right),
\]

\[
M_C = -\frac{2}{L^3} \left( \sum_{j=1}^{N_C} S_j^C \right)
\]

where \( N_B \) denotes the number of B ions \( N_B = pL^3/2 \), whilst \( N_C \) represents the number of C ions \( N_C = (1 - p)L^3/2 \) on the same cubic lattice. Total magnetization per site is given by

\[
M = \frac{1}{2} (M_A + M_B + M_C).
\]

**III. RESULTS AND DISCUSSION**

In this section, we have given the simulation results of the ternary alloy model \( \text{AB}_p\text{C}_{1-p} \) and we have also discussed the dependence of the critical and compensation temperature on the concentration and other interaction parameters in the Hamiltonian. Simulation results have been obtained for the system with lattice size \( L = 10, 12, 16, 20 \) and 24, however, here, we have only presented the results of the model with lattice size \( L = 20 \). We also note that the critical temperature of the system for the different interaction rates and concentrations have been obtained by using of the method of the finite-size scaling \[20\].

In a recent study \[15\] it was reported that two dimensional ternary alloy model does not show a compensation temperature point when there is no next-nearest neighbor interactions term in the Hamiltonian i.e., \( J_{AA} = 0 \). However, our simulations show that the system has a compensation point for all \( R \) (we set \( R = |J_{AC}|/J_{AB} \)) values in interval of \( 0.1 \leq R \leq 2.642 \) at \( p = 0 \) when \( J_{AA} = 0 \). This point will be considered below. Now, in order to compare with the previous results \[15 \, 18\], in Figs. 2 and 3 we discuss the dependence of the critical temperature of the three dimensional ternary alloy model \( \text{AB}_p\text{C}_{1-p} \) on interaction rate \( R \) and concentration \( p \) for next-nearest neighbor interactions i.e., \( J_{AA} = 0 \).

In Fig. 2, the critical temperature of the three dimensional ternary alloy model has been plotted as a function of \( R \) for various values of \( p \) when \( J_{AA} = 0 \). It can be seen from Fig. 2 that the critical temperature of the system has a linear dependence on the interaction ratio \( R \) and there is a critical behavior at a special \( R \) value. When \( R_c = R = 0.513 \), the critical temperature of the
system has a fixed value of $T_c = 5.47$ for all $p$ values. At $R_c$, the critical temperature of the system does not change with concentration $p$. This means that neither the spin-1 ions nor spin-5/2 ions substitution to system change the critical temperature of the system at $R_c$. This critical behavior has been reported in theoretical and experimental studies \cite{13,14,15}. The value of the $R_c$ for ternary alloy $\text{AB}_p\text{C}_{1-p}$ whose spins consist of $S^A = 3/2$, $S^B = 1$ and $S^C = 5/2$ has been obtained as $R_c = 0.4781$ in the study based on mean field approximation \cite{15} and as $R_c = 0.49$ in the Monte Carlo simulation of two dimensional system \cite{15}. Furthermore, the experimental measurements indicate that there are Prussian blue analogs at the $R = 0.45$ have a $T_c$ which is almost independent of $p$ \cite{15}. Fig. 2 also reveals that concentration $p$ plays an important role for the ternary alloy model $\text{AB}_p\text{C}_{1-p}$ since it determine the kinds of the spins and interactions in the system. For example, when $p = 1$ and $p = 0$, the system $\text{AB}_p\text{C}_{1-p}$ fully reduces to the ferromagnetic mixed spin-3/2 and spin-1 and ferrimagnetic mixed spin-3/2 and spin-5/2 Ising system, respectively. As seen in Fig. 2, although $T_c$ of the system is independent of $p$ at $R_c$, however, the total magnetization of the system may considerably change owing to relatively small variation of the concentration $p$. Indeed, for different values of $p$, the dependence of critical temperature of the system on the interaction ratio $R$ is very different above and below of $R_c$. This behavior can be explained by the change of the concentration $p$ in the system. On the other hand, it can be detected from Fig. 2 that when $R < R_c$, the critical temperature of the mixed spin-3/2 and spin-5/2 system is smaller than mixed spin-3/2 and spin-1 system. On the contrary, when $R > R_c$, the critical temperature of the mixed spin-3/2 and spin-5/2 system has the highest value. On the $T_c$ lines, the critical temperature of the mixed spin-3/2 and spin-1 Ising system is equal to that of the mixed spin-3/2 and spin-5/2 Ising one.

In Fig. 3, the dependence of the critical temperature of the three dimensional $\text{AB}_p\text{C}_{1-p}$ system on the concentration $p$ has been shown for several values of $R$ when $J_{AA} = 0$. The lines represent part of the second-order phase transition separating the ferrimagnetic and paramagnetic. Fig. 3 provides the argument that the concentration $p$ determines the magnetic features of the system mentioned above. Indeed, Fig. 3 clearly shows that the critical temperature of the system is changed by the concentration $p$ for fixed values of $R$. As seen from this figure that, when $R < R_c$, the critical temperature of the system linearly increases with increasing of $p$, whereas, when $R > R_c$, the critical temperature of the system linearly decreases with increasing of $p$ for fixed values of $R$. However, when the values of $R$ close up $R_c$, the critical temperature of the system more slowly, but linearly, change with increasing $p$, and at the critical $R_c$ value, the critical temperature of the system denoted by triangle-line in Fig. 3 is independent of the concentration $p$. On the other hand, Fig. 3 also shows that the interaction rate $R$ plays an important role on the critical temperature of the three dimensional $\text{AB}_p\text{C}_{1-p}$ system. Finally we state that the critical temperature of the model are consistent with previous result \cite{15}.

In this study we recognize that the three dimensional ternary alloy model $\text{AB}_p\text{C}_{1-p}$ has one compensation behavior for $J_{AA} = 0$, however, for $J_{AA} \neq 0$ it has one
or multi compensation points, when other conditions are satisfied. However, the appearance of the compensation temperature is strongly affected by the interaction and concentration parameters. Indeed we see in the present study that the model has not a compensation point for all values of \( p \) and \( R \). The dependence of the compensation temperature behavior on concentration and other interaction parameters has been discussed below. For discussion, although the system has been simulated in the intervals of \( 0 \leq p < 1.0 \) and \( 0.1 \leq R \leq 2.642 \), the value of \( J_{AA} \) used in the present study is chosen based on previous theoretical study \[15\]. The results of simulation for \( R = 1.0 \) and \( R = 2.642 \) are respectively represented in Figs. 4 and 5 for chosen parameters.

One compensation point has been found in the intervals of \( 0.0 \leq p < 0.3 \) and \( 0.1 \leq R < 2.642 \) when \( J_{AA} = 7.5 \). However, it is seen that the system has not compensation behavior for the same values of parameters when \( p \geq 0.3 \). For \( R = 1.0 \) and several values of \( p \), the compensation behavior of the system can be seen from Fig. 4. On the other hand, as seen from Fig. 5, the considered system has a multi compensation behavior at \( R = 2.642 \) and \( p = 0.3 \) for \( J_{AA} = 7.5 \) while it has one compensation point for \( 0.2 \leq p < 0.3 \). Furthermore, our simulation data introduce that the system shows compensation behavior at \( p = 0 \) for all values of \( R \), when \( J_{AA} = 0 \). In addition, in the case \( J_{AA} = 0 \), the compensation point has been found for \( R = 0.25 \) at \( p = 0.2 \), \( 0.25 \); for \( R = 0.75 \) at \( p = 0.3 \); for \( R = 1.25 \) at \( p = 0.3 \); for \( R = 2.0 \) at \( p = 0.1, 0.3, 0.4 \); for \( R = 2.642 \) at \( p = 0.1, 0.2, 0.3, 0.4 \). Whereas, it has been reported in previous study that there is no compensation point for \( J_{AA} = 0 \) in two dimensional model \[15\].

The effect of the interaction parameters on the compensation behavior of the three dimensional ternary model is also discussed in Fig. 6. This figure shows dependence of the compensation temperature \( T_{comp} \) of the model on interaction parameters in Hamiltonian only for a fixed value of the concentration parameter \( p \) (\( p = 0.25 \)). In this figure, square-line represents the behavior of the compensation point vs \( J_{AA} \) for fixed values of \( J_{AB} = 5 \), \( J_{AC} = -5 \) and \( p = 0.25 \), circle-line indicates the behavior of the compensation point vs \( J_{AB} \) for fixed values of \( J_{AA} = 7.5 \) and \( J_{AC} = -5 \) and \( p = 0.25 \), and on the other hand, the behavior of the compensation point vs \( J_{AC} \) is plotted for fixed values of \( J_{AA} = 7.5, J_{AB} = 5 \) and \( p = 0.25 \) with triangle-line. As seen from Fig. 6 that for fixed \( p \), \( J_{AB} = 5 \) and \( J_{AC} = -5 \), the compensation temperature decreases slowly as the strength of the \( J_{AA} \) increases. Similarly for fixed \( p \), \( J_{AA} = 7.5 \) and \( J_{AC} = -5 \), the compensation temperature decreases slowly with increasing of \( J_{AB} \). However, for fixed \( p \), \( J_{AA} = 7.5 \) and \( J_{AB} = 5 \), the compensation temperature dramatically increases as \( |J_{AC}| \) increases. These results indicate that the compensation temperature has a strong dependence on the parameter \( J_{AC} \) whereas its dependence on \( J_{AA} \) and \( J_{AB} \) is relatively weak. The characteristic behavior of the dependence of the compensation temperature on the parameters of present model consistent with the results of two dimensional model \[15\].
IV. CONCLUSION

In this study, we have considered the three-dimensional ternary model $AB_pC_{1-p}$ whose spins consist of $S^A = 3/2$, $S^B = 1$ and $S^C = 5/2$. We have investigated the dependence of the critical and compensation temperature behavior of the considered model on concentration and interactions by using MC simulation method. We have observed that the behavior of the critical temperature and the existence of compensation points strongly depend on interaction and concentration parameters. Particularly, we have found that the critical temperature of the model is independent on concentration of different types of spins at a critical $R_c$ value and the model has one or two compensation temperature points in a certain range of values of the concentration of the different spins. We concluded that magnetic properties of the system $AB_pC_{1-p}$ can be controlled by changing the relative concentration of the different species of ions. As a result, we would like to stress that these theoretical results can be very useful for designing molecular magnets in experimental studies since the existence of compensation in the ternary alloy $AB_pC_{1-p}$ that can be setup by adjusting the proportion of compounds.

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