Critical temperature of a mixed ferro-ferrimagnetic ternary alloy

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Abstract. We study the critical properties of a mixed ferro-ferrimagnetic ternary alloy of the type \(AB_pC_{1-p}\) on a cubic lattice consisting of three different Ising spins \(S_A = \frac{3}{2}, S_B = 2,\) and \(S_C = \frac{5}{2}\). The \(A\) ions are linked with either the \(B\) or \(C\) ions which are randomly distributed in the lattice with the concentration \(p\) or \(1-p\), respectively. The exchange interactions between nearest neighbours only, \(J_{AB} > 0\) and \(J_{AC} < 0\), are imposed. To locate the critical temperature of the ternary alloy we employ the mean-field approximation and Monte Carlo simulations. The effects of interaction ratio \(R = |J_{AC}|/J_{AB}\) and concentration \(p\) on the critical behaviour of the system are investigated and the results from both methods are compared. We find that the critical temperature of the mixed ferro-ferrimagnet for a special value of \(R\) does not depend on the concentration \(p\). The relation between the studied model and the structure of the Prussian blue analog such as \((\text{Fe}^{II}_p\text{Mn}^{II}_{1-p})_{1.5}[\text{Cr}^{III}(\text{CN})_6] \cdot n\text{H}_2\text{O}\) is also discussed.

1. Introduction
In recent years, there has been a noticeable interest in the investigation of magnetic properties of ternary mixed-spin Ising systems. These models are interesting not only of purely theoretical point of view but especially because they can describe the magnetic behaviour of a certain class of molecular-based magnetic materials, the so-called multimetal Prussian blue analogs, that exhibit many remarkable properties (see, e.g., [1] and references therein). For instance, the experimental studies of the compounds such as \((\text{X}^{II}_p\text{Mn}^{II}_{1-p})_{1.5}[\text{Cr}^{III}(\text{CN})_6] \cdot n\text{H}_2\text{O}\) \((\text{X}^{II} = \text{Ni}^{II}, \text{Fe}^{II})\) [2 – 4], which include both ferromagnetic \((J_{X\text{Cr}} > 0)\) and antiferromagnetic \((J_{\text{MnCr}} < 0)\) superexchange interactions through the cyanide bridging ligands, reveal an occurrence of a compensation temperature and an photoinduced magnetic pole inversion, respectively. It is worth to note that the magnetic properties (critical point, temperature dependence of magnetization, etc.) of the considered Prussian blue analogs can be tuned during a synthesis process by changing the mixing ratio \(p\) of the different incorporated metal ions.

Up to now, the ternary alloy models which contain three various kinds of magnetic ions with different Ising spins have been investigated by the use of a mean-field (MFT) [5, 6] or an effective-field theory (EFT) [7, 8], Monte Carlo (MC) simulations [9 – 11] and exact recursion relations on the Bethe lattice [12]. One of the most interesting features of the ternary Ising systems, predicted by the approximative methods [5 – 7], is a possibility of independence of a critical temperature on some parameters of the model. However, this outstanding behaviour of the transition temperature was verified by the considerably more accurate methods like the
MC simulations only for ternary alloys on a two-dimensional square lattice [9] and a simplified three-dimensional decorated lattice [10].

Therefore, in the present paper we adopt the MC simulations to investigate the critical temperature of a three-dimensional $AB_pC_{1-p}$ ternary alloy on a cubic lattice, consisting of three different Ising spins $S_A = \frac{5}{2}$, $S_B = 2$, and $S_C = \frac{3}{2}$, which corresponds to the Prussian blue analog of the type $(Fe_pMn_{1-p})_{1.5}[Cr^{III}(CN)_6] \cdot nH_2O$ [3]. In particular, we focus on the study of the effect of the concentration $p$ and interaction ratio $R$ on the critical temperature of the system.

2. Model and Monte Carlo simulations

We consider a mixed ferro-ferrimagnetic $AB_pC_{1-p}$ ternary alloy on a $L \times L \times L$ lattice that consists of two interpenetrating face-centered cubic sublattices, each one comprising $L^3/2$ sites. The $A$ ions of one sublattice are alternately connected with the $B$ or $C$ ions randomly located on the other sublattice with the concentration $p$ or $1-p$, respectively. Thus, the $A$ ion is coupled with the six nearest-neighbouring ions of the type $X$ ($X = B$ or $C$) and vice versa. Now, the Hamiltonian of the ternary alloy can be written as

$$H = - \sum_{(i,j)}^L S_A^i [J_{AB} S_B^j] + J_{AC} S_C^j (1 - \xi_j^j),$$

where $S_A^i = \pm \frac{5}{2}$, $S_B^j = \pm 2$, $\pm 1$ for $A$ ions, and $S_C^j = \pm \frac{3}{2}$, $\pm \frac{5}{2}$, $\pm 1$ for $C$ ions. $\xi_j$ is a random variable which takes the value of unity (zero) if the site $j$ is occupied by the $B$ ($C$) ion. To be consistent with a structure of the above-mentioned Prussian blue analog we also assume that the couplings between nearest neighbours include both the ferromagnetic ($J_{AB} > 0$) and the antiferromagnetic ($J_{AC} < 0$) interactions.

We employ a standard importance sampling method to simulate the Hamiltonian given by equation (1). Periodic boundary conditions on the cubic lattice of the system with the linear size $L = 20$ are imposed. All initial spin states are randomly assigned. Hereafter, configurations are generated by random passing through the lattice and making single-spin flip attempts. The flips are accepted or rejected according to the heat-bath algorithm [13]. Finally, averages are calculated by using $5 \times 10^4$ MC steps per site after discarding the first $10^4$ MC steps per site.

In order to study the critical temperature of the system we calculate the specific heat

$$\frac{C}{k_B} = \beta^2 \left( \langle H^2 \rangle - \langle H \rangle^2 \right),$$

where $\beta = 1/k_BT$. Then, a value of the transition temperature is estimated by locating the maximum of a specific heat curve.

3. Results and discussion

In this section we investigate the critical temperature of the three-dimensional mixed ferro-ferrimagnetic $AB_pC_{1-p}$ ternary alloy that is fairly affected by both the model parameters, the exchange interaction ratio $R = |J_{AC}|/J_{AB}$ and the concentration $p$.

In figure 1, we plot the transition temperature $T_c$ as a function of the interaction ratio $R$ for several values of the concentration $p$. The horizontal dependence for $p = 1.0$ corresponds to the critical temperature of the mixed spin-$\frac{3}{2}$ and spin-$2$ Ising system that is not affected by the parameter $R$. On the other hand, for $p = 0$ the ternary alloy reduces to the mixed-spin system with $S_A = \frac{5}{2}$ and $S_C = \frac{3}{2}$ whose transition temperature is the most sensitive to a variation of the nearest-neighbour couplings. As is clearly seen from the figure, for $0 < p < 1.0$ one can find a critical value of the interaction ratio $R_c \approx 0.83$ (denoted by the dotted line) such that the transition temperature of the ternary alloys with $0 < R < R_c$ is smaller than that of
the mixed spin-\(\frac{5}{2}\) and spin-2 Ising system and gradually decreases with a lowering of the value of the mixing ratio \(p\). However, in the case of \(R > R_c\) the critical temperature of the ternary systems increases with a decrease of the concentration \(p\). It is worth to remark that qualitatively the same critical behaviour of the mixed ferro-ferrimagnetic ternary alloy is also predicted by the simple MFT. Moreover, the critical value of the exchange interaction ratio obtained within this approximation, \(R_c^{\text{MFT}} = (\frac{24}{35})^{1/2} \approx 0.8281\) [6], is surprisingly very close to that of the MC simulations. We note that this unforeseen agreement of both approaches has also been achieved for another ternary Ising system on a three-dimensional decorated lattice [10].

In order to see the effect of the concentration \(p\) on the transition temperature \(T_c\) of the \(AB_pC_{1-p}\) ternary alloy more clearly we show in figure 2 the \(T_c\) vs \(p\) curves for several values of the interaction ratio \(R\). The solid lines and circles correspond to the results obtained by the MFT and MC simulations, respectively. We note that details of our mean-field calculations can be found elsewhere [6]. As seen from the figure, the critical temperature of the ternary system with the interaction ratio \(R = R_c^{\text{MFT}}\) and \(R = R_c\) within the MFT and MC simulations (horizontal dependences), respectively, takes a constant value for any concentration \(p\). Thus, in this special case the transition temperature \(T_c\) does not vary even if the spins \(S_B = 2\) are substituted by the spins \(S_C = \frac{5}{2}\). However, despite of a very good accordance between the MFT and MC simulations in determining the critical value of the interaction ratio, the corresponding

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure1.png}
\caption{The dependences of the critical temperature of the \(AB_pC_{1-p}\) ternary alloy on the interaction ratio \(R\) for selected values of the concentration \(p\). The dashed lines are guides for the eye.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure2.png}
\caption{The dependences of the critical temperature of the \(AB_pC_{1-p}\) ternary alloy on the concentration \(p\) obtained by the MFT (solid lines) and the MC simulations (circles), respectively, when the value of \(R\) is changed.}
\end{figure}
transition temperature obtained by the MFT \( (k_B T_c/J_{AB} \approx 9.49) \) is overestimated in comparison with that of the MC simulations \( (k_B T_c/J_{AB} \approx 7.70) \). Further, it is seen from the figure that, contrary to the critical temperature \( T_c \) of the system with \( R = 0.1 \), the \( T_c \) of the ternary alloy with \( R = 2.642 \) (this value corresponds to the above-mentioned Prussian blue analog [3]) is a decreasing function of the concentration \( p \). This behaviour of the transition temperature is in agreement with that of figure 1.

Finally, it is worth mentioning that the studied \( AB_pC_{1-p} \) ternary alloy is composed of the \( A \) and \( X \) \( (X = B \) or \( C) \) ions having the same numbers of nearest neighbours. However, the structure of this model differs from that corresponding to the ternary Prussian blue analog in which the numbers of nearest neighbours of the \( A \) and \( X \) ions may be unequal and can be controlled by the stoichiometry of the system. We note that the value of \( R_c^{MFT} \) is independent on the nearest-neighbour coordination numbers of the ternary alloy under consideration [6]. Therefore, it will be interesting to verify this prediction by the MC simulations. The solving of this task is now in progress.

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