Monte Carlo study of mixed-spin $S = (1/2, 1)$ Ising ferrimagnets

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Abstract

We investigate Ising ferrimagnets on square and simple cubic lattices with exchange couplings between spins of values $S = 1/2$ and 1 on neighbouring sites and an additional single-site anisotropy term on the $S = 1$ sites. Mainly on the basis of a careful and comprehensive Monte Carlo study, we conclude that there is no tricritical point in the two-dimensional case, in contradiction to mean-field predictions and recent series results. However, evidence for a tricritical point is found in the three-dimensional case. In addition, a line of compensation points is found for the simple cubic, but not for the square lattice.

1. Introduction

Mixed-spin Ising models have been studied for some time as simple models of ferrimagnets, and there has been renewed interest recently in connection with ‘compensation points’. These are temperatures, below the critical temperature, at which the sublattice magnetizations cancel exactly, giving zero total moment. As the temperature is tuned through such a point the total magnetization changes sign, which may be used in technological applications. In this context, Ising models may be exactly solvable in special cases [1–5] or they may be studied by a variety of powerful approaches, including Monte Carlo [6–11] or other [12–17] methods. In the present work we revisit one of the simplest such models, a mixed-spin Ising model with spins $S = 1/2$ and 1 occupying the sites of a bipartite square or simple cubic lattice with the Hamiltonian

$$\mathcal{H} = -J \sum_{\langle i,j \rangle} \sigma_i S_j + D \sum_{j \in S} S_j^2$$

with couplings $J$ between spins $\sigma_i = \pm 1$ on the sites of sublattice ‘A’, and neighbouring spins $S_j = 1, 0, -1$ on sites forming the sublattice ‘B’. $D$ denotes the strength of a single-ion term acting only on the $S = 1$ spins of sublattice B. Following previous convention, we choose $\sigma_i = \pm 1$ rather than $\pm 1/2$, which has to be taken into account when calculating sublattice magnetizations and when defining the compensation point. The convention simply amounts to a rescaling of the exchange coupling. Note that the nearest-neighbour coupling $J$ may be either antiferromagnetic, $J < 0$, as assumed often for ferrimagnets, or ferromagnetic, $J > 0$. Both cases are completely equivalent by a simple spin reversal on either sublattice. We shall use in this article ferromagnetic couplings. As a consequence, in our case the magnetizations of both sublattices are identical at the compensation point, while in the antiferromagnetic case, at the same compensation point, the sublattice magnetizations have equal magnitude but different sign leading to the above mentioned vanishing of the total magnetization.

The model on the square lattice has been studied by several authors. Kaneyoshi and Chen [14], via a mean-field treatment, found a line of compensation points in a narrow region $4 > D/J \geq 2 \ln 6 (=3.583 \ldots)$ and a tricritical point at $D_t/J = 3.72$, i.e. a first-order transition for $D > D_t$. Buendia and Novotny [9], using transfer matrix methods, supplemented by Monte Carlo simulations, found no evidence of either a compensation point or a tricritical point, although a compensation point was observed in an extended model with additional ferromagnetic interactions between $\sigma$ spins. More recently, Oitmaa and Enting [18] studied the same model using a combination of high- and low-temperature series. No compensation point was found, but evidence for a first-order transition, and hence a tricritical point was observed from an apparent crossing of the high- and low-temperature branches of the free energy with different slopes, for $D/J \geq 3.2$. Thus the phase diagram of this simple model remained uncertain, motivating partly the present extensive Monte Carlo study, improving previous simulations substantially. In fact, our study provides clear evidence that the model in two dimensions has no compensation point or tricritical point. Moreover, the model is found to exhibit very interesting thermal behaviour, both for the specific heat and the magnetization, especially in the low-temperature region near $D = 4$, which has not been discussed.
in detail before. This behaviour is the likely explanation for the apparent ‘first-order’ behaviour observed in [18].

For the simple cubic lattice, to our knowledge, no detailed analyses have been done so far. Of course, mean-field theory may be easily applied, leading again to a tricritical point and a line of compensation points.

The outline of the paper is as follows. In section 2 we present and discuss our results for the square lattice. In section 3 we consider the simple cubic lattice. Here, in contrast to the two-dimensional case, we find a clear occurrence of a line of compensation points. Furthermore, we obtain clear evidence of transitions of first order, and thence of a tricritical line of compensation points.

2. The model on the square lattice

Let us first consider the ferrimagnet, equation (1), in the case of a square lattice. We have performed mainly standard Monte Carlo simulations, using the Metropolis algorithm with single-spin flips, providing, indeed, the required accuracy, so that there was no need to apply other techniques like cluster-updates or the Wang–Landau approach [19]. We studied lattices with \( L \times L \) sites, employing full periodic boundary conditions. \( L \) ranged from 4 to 80, to study finite-size effects. Typically, runs of \( 10^7 \) Monte Carlo steps per spin have been done, with averages and error bars obtained from evaluating both from the energy fluctuations and from differentiating the probability of the system.

We recorded the energy per site, \( E \), the specific heat, \( C \), both from the energy fluctuations and from differentiating \( E \) with respect to the temperature, and the absolute values of the sublattice magnetizations of the two sublattices

\[
|m_A| = \left| \langle \sum_A \sigma_i \rangle \right| / \left( 2L^2/2 \right) \tag{2}
\]

and

\[
|m_B| = \left| \langle \sum_B S_j \rangle \right| / \left( L^2/2 \right) \tag{3}
\]

as well as the absolute value of the total magnetization,

\[
|m| = \left| \langle \sum_A \sigma_i + \sum_B S_j \rangle \right| / L^2 \tag{4}
\]

where the brackets \( <> \) denote the thermal average. Note the factor of \( 1/2 \) in the definition of \( |m_A| \), taking into account the correct length of the \( S = 1/2 \) spins, so that \( |m_A(T = 0)| = 1/2 \), while \( |m_B(T = 0)| = 1 \) for the ferromagnetic ground state. In addition, the corresponding susceptibilities, \( \chi_A \), \( \chi_B \), and \( \chi \), have been computed from the fluctuations of the magnetizations. We also analysed histograms for the total magnetization, \( p(m) \), i.e. the probability to encounter a configuration with the magnetization \( m \), as well as the fourth-order cumulant of the order parameter, the Binder cumulant [20], defined by

\[
U = 1 - \langle m^4 \rangle / 3 \langle m^2 \rangle^2 \tag{5}
\]

with \( \langle m^2 \rangle \) and \( \langle m^4 \rangle \) being the second and fourth moment of the total magnetization. Finally, we monitored typical equilibrium Monte Carlo configurations, illustrating the microscopic behaviour of the system.

To test the accuracy of the simulations, we computed numerically exact results for various quantities by enumerating all possible configurations for small lattices with \( L = 4 \).

In agreement with previous work, the model is observed to display a ferromagnetic ground state and low-temperature phase for \( D/J < 4 \). The energy to flip a B spin from its ferromagnetic orientation, ‘+’ or ‘−’, surrounded by four A spins of the same orientation, to the state 0 is obviously \( \Delta E = 4J - D \) which vanishes at \( D = 4J \). Hence the ground state at \( D/J = 4 \) will comprise configurations with ‘0’ states on B sites and arbitrarily oriented spins on the neighbouring A sites, as well as ferromagnetic plaquettes (of either sign) on B sites and neighbouring A sites. Due to the resulting high degeneracy, one may call \( D/J = 4, T = 0 \) the ‘degeneracy point’. For \( D > 4J \) at zero temperature, all B spins will be in the state 0, with the A spins being randomly oriented. This leads to a lower, but still macroscopic degeneracy. At \( D/J > 4 \), there is no ordered phase even at zero temperature.

Most of our Monte Carlo work deals with the interesting range \( 3 < D/J < 4 \), which had been discussed controversially before, augmented by some simulations at lower values of \( D/J \). The resulting phase diagram is depicted in figure 1, based on monitoring the size dependence of the position of the (critical) maxima in the specific heat and susceptibility, and the intersection points of the Binder cumulant, see below. Our findings are in accordance with a continuous transition in the Ising universality class for all values of \( D/J \) we studied, \( D/J \leq 3.98 \). There is no compensation point.

In the following, we shall discuss main properties of the physical quantities mentioned above.

The specific heat \( C \), for negative or relatively small positive \( D/J \), is observed to resemble qualitatively that of...
the nearest-neighbour Ising model on a square lattice. There is a unique maximum in $C(T)$, for finite $L$, turning into a logarithmic singularity in the thermodynamic limit. Indeed, in the limit $D/J \to -\infty$, one recovers the simple Ising model. Increasing $D/J$, as displayed in figure 2(a) for $D/J = 3.0$, an additional shoulder or maximum evolves at a lower temperature, $T_n$, being largely independent of lattice size and being non-critical. Its origin becomes clear by further increasing $D/J$, as shown in figure 2(b) for $D/J = 3.8$. In fact, one finds $k_B T_n/J \approx 0.42(4 - D/J)$, reflecting the thermally activated flipping of $B$ spins from the ferromagnetic state ’1’ (or ’−1’) to the state zero, requiring, as stated above, an energy proportional to $4 - D/J$. It is interesting to note that the height of the pronounced non-critical peak, signalling the partial disordering of the B sublattice, depends only very weakly on $D/J$. In the range $3.5 \leq D/J < 4$, one has $C(T_n) \approx 0.22$.

As illustrated in figure 2(b) for $D/J = 3.8$, the critical peak, located at $T_m$, may separate from the upper maximum, at $T_n$, when increasing the strength of the single-ion term. Thus, the specific heat may display a three-peak structure, with two non-critical maxima and a critical peak in between. The origin of the maximum at $T_n$ is due to the fact that at the critical point, the $\sigma$ spins on the $A$ sublattice form rather large clusters of different orientations, leading to the vanishing of the order parameter. That behaviour may be seen by monitoring typical equilibrium configurations. These clusters shrink quickly near $T_n$, due to thermally activated flipping of $\sigma$ spins, determined by the coupling constant $J$. Indeed, $T_n$ is essentially independent of $D$. As seen in figure 2(b), the maximum in $C$ at $T_n$ depends rather weakly on the size of the lattice, $L$, demonstrating its non-critical character.

The height of the critical maximum at $T_m$ is expected, for Ising universality, to increase logarithmically with $L$ for sufficiently large values of $L$. Our results are consistent with this expectation. However, on approach to the degeneracy point, the background contribution to the specific heat becomes more and more relevant. Then larger and larger lattices, with $L$ exceeding a critical value, $L_0$, are needed to approach the anticipated logarithmic behaviour. For example, at $D/J = 3.6$, one gets $L_0 \approx 40$, and 60 at $D/J = 3.95$. In fact, in that range, the Ising-like character of the transition may be inferred more clearly from other quantities, as discussed below.

The partial disordering of the $B$ sublattice, near $T_n$, leads to a rapid decrease of the magnetization $|m_B|$, as illustrated in figure 3. Actually, the anomaly in $|m_B|$ becomes more and more dramatic on approach to the degeneracy point. In contrast, the magnetization of the $A$ sublattice, $|m_A|$, is hardly affected by the disordering of the $B$ sublattice. Indeed, this behaviour may open the possibility of a compensation point, at which the two sublattice magnetizations, $|m_A|$ and $|m_B|$, would coincide. However, as depicted in figure 3, we find no evidence for such a compensation point in two dimensions for all cases we studied, with $D/J$ going up to 3.95.

The susceptibility $\chi$ is found to show, in all cases we studied, only one maximum, close to the critical temperature. The background term is much weaker than for the specific heat, allowing an analysis of critical properties for smaller lattices. In fact, as illustrated in figure 4, the size dependence of the height of the maximum in $\chi$, $\chi_{\text{max}}(L)$, is observed to be nicely compatible with the asymptotic form $\chi_{\text{max}} \propto L^{7/4}$, expected for the Ising universality class, for all cases studied and sufficiently large lattices. Note that the susceptibility

![Figure 2](image-url)

![Figure 3](image-url)
Critical Binder cumulant $U$ is also true for the present model, as shown in figure 5 for corrections often turn out to be rather small. Actually, this shows a very mild anomaly near $T_c$, where the specific heat shows a pronounced maximum, close to the degeneracy point. At that anomaly, $\chi(T)$ exhibits a maximal slope, as may be easily identified using exact enumeration for small lattices. We do not include a special figure to depict the very weak anomaly. The shrinking of the $A$ clusters, as indicated by the broad maximum in $C$, at $T_c$, leads to no obviously unusual features in the susceptibility.

As usual, one may estimate the bulk transition temperature, $T_c$, from the size dependent position of the corresponding peaks in $\chi$ and $C$. We obtain consistent estimates, shown in figure 1, with the location of the maxima varying, for large $L$, proportionally to $1/L$, as expected for Ising-like transitions. Of course, one gets distinct proportionality factors for the two quantities.

The transition temperature may be also conveniently estimated from the Binder cumulant, $U$. Indeed, the estimates follow from the location of the intersection temperatures of the cumulants for different lattice sizes [20]. Finite-size corrections often turn out to be rather small. Actually, this is also true for the present model, as shown in figure 5 for $D/J = 3.95$. We find very good agreement with the estimates of $T_c$ based on the susceptibility and the specific heat. Note that the value of $U$ at the intersection temperature is, already for fairly small systems sizes, close to the accurately known [21] critical Binder cumulant $U^* = U(T_c, L = \infty)$ for isotropic Ising models, $U^* = 0.6069\ldots$. One may emphasize that anisotropic interactions and correlations may lead to non-trivial dependencies of $U^*$ on such interactions [22, 23]. However, here we are dealing with an isotropic system, and excellent agreement with the known critical value is observed, demonstrating that the transition belongs to the Ising universality class.

Additional insight into the phase transition is provided by the histograms for the total magnetization, $p(m)$. An example is displayed in figure 6. As expected for a continuous transition, $p(m)$ shows, in the ferromagnetic low-temperature phase, two symmetric peaks, at $\pm m_0$, moving closer and closer to each other on approach to $T_c$ and when increasing the lattice size. Above $T_c$, $p(m)$ tends to acquire a Gaussian shape [20].

We emphasize that figure 6 refers to the case $D/J = 3.98$, i.e. very close to the degeneracy point. There is no indication of a transition of first order, which might be signalled by a central peak, in addition to the two peaks at $\pm m_0$, as would be the case for coexistence of the disordered and ordered phases. Accordingly, we may safely conclude, based on the analysis of several quantities, that we have clear evidence for continuous transitions of Ising-type along the boundary of the ferromagnetic phase, at least for the region $D/J \leq 3.98$.

### 3. The model on the simple cubic lattice

Let us now turn to the analysis of the mixed-spin model, equation (1), on a simple cubic lattice. In complete analogy to the two-dimensional case, we did standard Monte Carlo simulations, applying the Metropolis algorithm. We studied lattices with $L^3$ sites, with $L$ ranging from 4 to 32. Full periodic boundary conditions were employed. Typically, runs of $2 \times 10^5 - 5 \times 10^6$ Monte Carlo steps per spin were performed, averaging over a few, at least three, such runs to estimate thermal averages and error bars.
As for the square lattice, the energy \( E \), the specific heat \( C \), magnetizations \( |m_A|, |m_B|, \) and \( |m| \), as well as corresponding susceptibilities, the Binder cumulant \( U \), and histograms for the total magnetization, \( p(m) \), were recorded. Typical Monte Carlo equilibrium configurations were generated to illustrate the microscopic behaviour.

For the cubic lattice, one has a ferromagnetic ground state at \( D/J < 6 \). The degeneracy point occurs now at \( D/J = 6 \), with ground states comprising local ferromagnetic plaquettes of neighbouring A and B spins as well as B spins in the state 0 with surrounding A spins being randomly oriented. For \( D/J > 6 \), a high, but reduced degeneracy prevails, with all B spins being zero, and the A spins pointing randomly ‘up’ or ‘down’.

For \( D/J \) small or negative, a continuous transition of Ising-type is expected to occur, as we confirm in simulations with moderate efforts. Most of our work has been done for \( 3.5 \leq D/J < 6 \), to identify possible deviations from that kind of transition. Indeed, significant deviations from Ising universality have been observed for \( D/J \geq 5.9 \), while for smaller values of \( D/J \) the simulational data are consistent with an Ising-like transition. In addition, we identified and located a line of compensation points in the range \( 5.5 < D/J < 6 \). The main features of the phase diagram are summarized in figure 7. The phase transition line is based on analysing various quantities and taking into account finite-size effects, as for the square lattice. Details of our Monte Carlo findings will be discussed in the following.

The specific heat \( C(T) \) shows for small and negative values of \( D/J \) a single maximum, giving rise to critical behaviour in the thermodynamic limit. In case of an Ising-like transition, its height is expected [24] to grow like \( C_{\text{max}} \propto L^{\beta/\nu} \) with the critical exponents of the Ising universality class, \( \alpha \approx 0.11 \) and \( \nu \approx 0.63 \) [25]. Our simulational findings confirm this scenario. As in the case of the square lattice, upon increasing \( D/J \), one encounters, eventually, three maxima in \( C(T) \), see figure 8. In complete analogy to the two-dimensional case, the peak at the lower temperature, \( T_u \), is rather sharp and depends only very weakly on lattice size. It signals the partial disordering of the B sublattice, with B spins being flipped thermally from the ferromagnetic (‘+’ or ‘−’) state to 0. The maximum occurs at \( k_B T_u/J \approx 0.6(6-D/J) \). The upper, rather broad maximum, at \( T_u \), is non-critical as well, stemming from dissolving the, at criticality still quite large spin clusters on the A sublattice. \( T_u \) is only very weakly affected by the strength of \( D \), being determined by the ferromagnetic coupling \( J \). In between the two non-critical maxima in \( C(T) \), a critical peak shows up. It signals the transition, at which both sublattice magnetizations vanish, with quite pronounced local spin order on the A sublattice.

The type of the transition may be inferred from the size dependence of the critical peak, \( C_{\text{max}}(L) \). Indeed, for single-ion terms up to \( D/J = 5.8 \), we find agreement with an Ising-type transition, \( \alpha/\nu \approx 0.17 \). On further approach to the degeneracy point, accurate Monte Carlo data with a fine temperature resolution are required, due to the rather large nonanalytic background term in \( C \) and the sharpness of the peak. In fact, other quantities may provide more easily and clearly reliable clues on the type of transition for that part of the transition line of the ferromagnetic phase.

Before discussing further the type of the phase transition close to the degeneracy point, we shall deal with the compensation points. Indeed, we identified such points in the range \( 5.5 < D/J < 6 \). The resulting line is depicted in figure 7. Two concrete examples are shown in figure 9, for \( D/J = 5.7 \) and 5.9. As may be inferred from that figure, the sublattice magnetization at the compensation point decreases monotonically with decreasing single-ion term. Therefore, when the compensation occurs at low magnetizations, the accurate location of the compensation point is difficult, because of strong finite-size effects in the critical region. On the other hand, with increasing \( D/J \), the compensation point moves towards lower temperatures, and finite-size effects play usually no significant role. In any event, in contrast to the two-dimensional case, we find a line of compensation points for the simple cubic lattice. Obviously, the decrease in the magnetization of the B sublattice, \( |m_B| \), occurs in three dimensions more drastically than for the square lattice, while \( |m_A| \) changes there rather mildly in both cases.
Let us now turn back to the discussion on the type of phase transition. For $D/J \leq 5.8$, the data on the susceptibility $\chi$ confirm the Ising-like character of the transition. In particular, the size dependence of the height of the maximum in $\chi$, $\chi_{\text{max}}(L)$, is found to be consistent with Ising criticality, $\chi_{\text{max}} \propto L^{\nu/\gamma}$ where $\nu \approx 1.24$ and $\gamma \approx 0.63$, thus $\gamma/\nu \approx 1.97$. Indeed, from our simulational data we obtain characteristic exponents close to 2. However, at $D/J = 5.9$, we observe, for systems sizes ranging from $L = 8$ to 32, a substantially lower (effective) exponent, of about 1.7. Because the peak in $\chi$ gets extremely sharp, very accurate simulational data with a very fine temperature mesh are needed to arrive at safe conclusions. A more convenient way to monitor the possible change in the type of the transition will be discussed below.

Interestingly, our analysis of the Binder cumulant $U$ seems to indicate substantial deviations from an Ising-like transition at about $D/J \approx 5.9$ as well. For smaller values of $D/J$ the intersection values of the cumulant curves for different system sizes, already for fairly small systems, seem to agree with the expected asymptotic value of the critical Binder cumulant for isotropic Ising systems [26], $U^* \approx 0.465$. An example is depicted in figure 10(a), for $D/J = 5.5$, with the intersection points, for the simulated finite lattices, approaching the asymptotic value from below, when increasing the system size. At larger single-ion anisotropy, $D/J \geq 5.9$, the intersection points of the curves are appreciably lower than $U^*$, as shown in figure 10(b) for $D/J = 5.9$. However, it is not completely clear, whether the tendency reflects stronger finite-size effects or a change in the type of the phase transition.

To get more evidence for a possible change of the nature of the transition, the histograms for the magnetization, $p(m)$, turned out to be most instructive. Already for small lattices, $L = 4$, one sees, close to the transition, a qualitative change of the histograms. We did simulations close to the transitions in the range $5.85 \geq D/J \geq 5.98$, using an increment of 0.01. We observe a dramatic change in the form of the histograms around $D/J \approx 5.91$. Below that value, there is no central peak and thus no indication of phase coexistence when crossing the transition, in contrast to the situation closer to the degeneracy point, where a central peak, in addition to the symmetric peaks at $\pm m_0$, indicates coexistence of the ordered and disordered phases and, accordingly, a transition of first order. That distinction persists for larger system sizes. Examples are displayed in figures 11(a), for $D/J = 5.85$, and 11(b), for $D/J = 5.975$. Based on these observations, we may tentatively locate the tricritical point at $D/J = 5.91 \pm 0.03$. Note that such a change in the form of the histograms does not occur in two dimensions, as has been discussed above, see also figure 6.

In summary, the present analysis on the mixed-spin model on a simple cubic lattice shows clearly a line of compensation points, and allows to locate approximately the tricritical point.

4. Summary

We have studied a mixed-spin Ising model with ferromagnetic couplings, $J$, between spins $1/2$ and $1$ on neighbouring sites of square and simple cubic lattices, the two types of spins forming a bipartite lattice. An additional quadratic single-ion term, $D$, acts upon the $S = 1$ spins. We mainly used standard Monte Carlo simulations to compute various thermodynamic properties as well as the Binder cumulants and histograms of the total magnetization.

The model on the square lattice has been shown to display a continuous phase transition of Ising-type, presumably up to the degeneracy point at $D/J = 4$. No compensation point

![Figure 9](image9.png)

**Figure 9.** Sublattice magnetizations $|m_A|$ and $|m_B|$ for the simple cubic lattice with $L = 20$ at $D/J = 5.7$ (circles) and 5.9 (squares).

![Figure 10](image10.png)

**Figure 10.** (a) Binder cumulant $U$ versus temperature at $D/J = 5.5$ for lattices with $L = 8$ (circles), 12 (squares), 16 (diamonds) and 20 (triangles). (b) $U$ versus temperature at $D/J = 5.9$ for lattices with $L = 10$ (circles), 16 (squares), 20 (diamonds), and 32 (triangles). The horizontal lines indicate the critical Binder cumulant of an isotropic three-dimensional Ising model in the thermodynamic limit [26].
has been found. Close to the degeneracy point, the model displays an intriguing three-peak structure in the specific heat as a function of temperature. The sharp, but non-critical anomaly at low temperatures arises from flipping $S = 1$ spins into the state 0, while the broad non-critical maximum at high temperatures stems from thermal activation of spins in fairly large clusters of $S = 1/2$ spins persisting above the phase transition. At temperatures in between, the critical peak shows up. Both anomalies may cause difficulties in low- and high-temperature expansions, which have predicted, incorrectly, the existence of a tricritical point. The suggestion on the absence of a compensation point has been confirmed, albeit the magnetization on the $S = 1$ sublattice decreases rapidly near the anomaly of the specific heat at low temperatures.

In the case of the simple cubic lattice, the specific heat displays a similar three-peak structure, with two non-critical maxima and the critical peak in between. Sufficiently far away from the degeneracy point, $D/J = 6$, this transition seems to be of first order. The evidence for that kind of transition is mainly based on the type of the histograms of the magnetization, showing phase coexistence. We tentatively locate the tricritical point at $D/J = 5.91 \pm 0.03$. In addition, we determined a line of compensation points, arising from the degeneracy point. Thus, in three dimensions, the mean-field theory appears to give at least qualitatively correct predictions. However, in two dimensions the mean-field theory is found to be incorrect, even qualitatively.

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