Exact phase diagram for the mixed spin-$1/2$ and spin-$S$ Ising models on the square lattice

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Abstract

We propose a new approach which permits us to obtain exact results for the mixed spin-$1/2$ and Spin-$S$ ($S > 1/2$) Ising models with single ion anisotropy $\Delta$ on the square lattice. We drive an explicit expression for the critical temperature for arbitrary values of $S$. We determine the exact phase diagrams for different values of $S$, and we show that there is no tricritical point. For $S$ integer, there is no long range order when the anisotropy exceeds a critical value which is independent of $S$. Furthermore, the exact Ising transition temperature $T_C$ is always recovered, for any values of $S$, in the limit of $\Delta \to -\infty$. These exact results are based on a conjecture which extend the analyticity of the $n$-spin-1/2 correlations functions (except at $T = T_C(\text{Ising})$) for any finite number $n$, to $n \to \infty$. This is confirmed by our calculations since our results are practically similar to those obtained by Monte Carlo simulations for $S = 1, 2$ and $3/2$.

1. Introduction

The Ising model is certainly the most studied cooperative many-body model in science [1]. Its importance lies in the fact that it has been used for various physical systems, such as a model for certain kinds of highly anisotropic magnetic crystals as well as a lattice model for fluids, alloys, adsorbed monolayers and even more in field theories of elementary particles (lattice gauge theories describing the quark structure of hadrons) [2]. It was also used successfully for biological, chemical systems and in the design of quantum computer based on one dimensional Ising systems [3, 4].

In statistical mechanics, in particular, in phase transitions and critical phenomena, physicists are interested in exactly solved models. On one hand, they permit to describe, to explain and therefore to obtain insight into the behavior of real systems throughout direct comparisons with experimental data. On the other hand, they provide extremely valuable tests of general theories and assumptions, such as scaling and universality hypotheses. Moreover, they serve as benchmarks for testing approximated methods which might be applicable to a wider class of models. The exact solution of the spin-1/2 Ising model on a square lattice, performed by Onsager in 1944 [5], was considered as an initial giant step in statistical mechanics of phase transitions. Due, however, to the intrinsic mathematical complexities of the model, no exact solution have been performed for spin greater than 1/2 or for a mixture of spin 1/2 and spin-S on square lattice. It is appropriate to mention here that recently there has been an intense interest directed to study the magnetic properties of two-sublattices mixed spin Ising systems. They have less translational symmetry than their single spin counterparts. This latter property has a great influence on the magnetic properties of the mixed-spin systems and causes them to exhibit unusual behavior not observed in single-spin Ising models. These mixed spin Ising models are well adapted to study a certain type of ferrimagnetism [6]. Experimentally, it has been shown that the MnNi(EDTA)-6H$_2$O complex [7] is a good example of a mixed system. More interestingly, the two dimensional compounds
A\textsuperscript{3}M\textsuperscript{II}M\textsuperscript{III}(C\textsubscript{2}O\textsubscript{4})\textsubscript{3} could represent good candidates to be described by the mixed spin Ising model since the magnetic metal atoms M\textsuperscript{II} and M\textsuperscript{III} of these polymeric molecular-based magnetic materials constitute more or less a regular honeycomb lattice [8]. The most obvious attempt for solving the two-dimensional mixed spin Ising model is to map the model onto an exactly solved one. Such technique has been used to solve exactly the mixed spin-1/2 and spin-S (S > 1/2) Ising models on honeycomb lattice [9]. It has been shown that, for S integer, no long range order is present in the system when the uniaxial crystal field is greater than a critical value. This latter is independent of the value of S. Using the same procedure, two of us (A D and N B) have solved exactly a two dimensional mixed- spin Ising ferrimagnet with next nearest neighbor (NNN) interaction between pairs of spins1/2. They showed that the NNN interaction is mainly responsible for the appearance of the compensation phenomenon while the crystal field may influence both the existence and the location of the compensation temperature [10]. We also note that M Jašćur \textit{et al} studied the effect of the uniaxial and the biaxial crystal-field potential on the magnetic properties of the mixed spin-1/2 and spin-1 (3/2) on the honeycomb lattice using exact mapping transformation [11, 12]. Apart from this, a considerable work in recent years has been undertaken to investigate the mixed spin-1/2 and spin-S (S > 1/2) Ising models on decorated square lattice [13], which can be mapped onto simpler models. Attention has been devoted to study the phase diagrams and the compensation points. However, the method used in [13] cannot be applied when the mixed-spin model is defined on the square lattice because it generates high order interactions with increasing complexities. The purpose of the present work is to use a new approach to obtain the exact phase diagrams of the tow-dimensional mixed spin-1/2 and spin-S Ising models on square lattice.

This paper is organized as follows: in section 2, we define the model system and show how the phase diagram can be exactly solved on the square lattice for arbitrary S. Namely; we give a detailed description of this new method which allows us to overcome the above difficulty related to the four neighbors, generated by the exact mapping transformation. Next, the phase diagrams as a function of the parameters are presented and discussed. In section 3, we focus our attention for the cases S = 1, 2 and 3, and S = 3/2, 5/2 and 7/2. We expect that these cases represent the main qualitatively behavior of S integer and S half-odd-integer respectively. Finally, our concluding remarks are given in section 4.

2. Model and its exact phase diagram

In this paper, we use a new approach to obtain the exact phase diagrams of the two-dimensional mixed spin-1/2 and spin-S Ising models on square lattice.

The model is described by the following Hamiltonian

$$H = -J \sum_{\langle j, k \rangle \in \Lambda_1} \sigma_j S_j + \Delta \sum_{j \in \Lambda_2} (S_j)^2$$ (1)

The lattice consists of two interpenetrating sublattices \(\Lambda_1\) with spins \(\sigma = \pm 1\) and \(\Lambda_2\) with spins S, where S is an arbitrary Ising spin greater than 1/2. The interactions are restricted to the nearest-neighbors and the periodic boundary conditions will be adopted. The partition function is then given by

$$Z = \sum_{\{\sigma, S\}} \exp(-\beta H)$$ (2)

where \(\beta\), as usual, is \((k_B T)^{-1}\).

The first step of the method consists in performing the partial trace over spins S. Then Z can be written as

$$Z = \sum_{\{\sigma\}} \prod_{j=1}^{N} Z_j$$ (3)

\(Z_j\) stands for a trace over spin states of the jth spin belonging to the sublattice \(\Lambda_2\). It is given by

$$Z_j = \sum_{S_j \in \Lambda_2} \exp\left[\frac{K}{2} S_j (\sigma_{jk} + \sigma_{jl} + \sigma_{jm} + \sigma_{jn} - D (S_j)^2)\right] + \sum_{s=-S}^{+S} \exp(-Dm^2) \cosh\left[\frac{K}{2} m (\sigma_{jk} + \sigma_{jl} + \sigma_{jm} + \sigma_{jn})\right]$$ (4)

where \(\sigma_{jk}, \sigma_{jl}, \sigma_{jm}\) and \(\sigma_{jn}\) are the nearest-neighbors of the spin \(S_j\) (figure 1(a)). \(K = \beta J, D = \beta \Delta\) and \(N\) is the total number of sites in \(\Lambda_2\) (or \(\Lambda_1\)). We note that \(Z_j\) can be expanded as follows

$$Z_j = a(K, D) + b(K, D) (\sigma_{jk} \sigma_{jl} + \sigma_{jk} \sigma_{jm} + \sigma_{jl} \sigma_{jm} + \sigma_{jl} \sigma_{jn} + \sigma_{jm} \sigma_{jn} + \sigma_{jm} \sigma_{jn}) + c(K, D) \sigma_{jk} \sigma_{jl} \sigma_{jm} \sigma_{jn}$$ (5)
where the coefficients $a$, $b$ and $c$ are given by

\[
a(K, D) = \frac{1}{8} \sum_{m=-S}^{S} \exp(-Dm^2) \{ \cosh(2Km) + 4 \cosh(Km) + 3 \} \\
b(K, D) = \frac{1}{8} \sum_{m=-S}^{S} \exp(-Dm^2) \{ \cosh(2Km) - 1 \} \\
c(K, D) = \frac{1}{8} \sum_{m=-S}^{S} \exp(-Dm^2) \{ \cosh(2Km) - 4 \cosh(Km) + 3 \}
\]

For the honeycomb lattice, where the coordination number is $z = 3$, one can map the model onto an effective exactly solved spin-1/2 Ising model on triangular lattice [9]. However, following this idea for the square lattice which has four neighbors, this does not work because (4) generates an additional four spins and next-nearest neighbors couplings, so the spin-1/2 model has no exact solution.

The second step of our approach permits us to avoid this difficulty. It consists to consider the spin-1/2 Ising model on square lattice with coupling constant $K_1$, solved exactly by Onsager [5]. For this model, the sites of the two sublattices $\Lambda_1$ and $\Lambda_2$ are occupied by Ising spins $\sigma = \pm 1$. The idea is to trace over the $\{ \sigma_j \}$ set of spins located on one sublattice. Then its partition function $Z_j^I$ can be written in the form

\[
Z_j^I = \sum_{\{ \sigma \}} Z_j^I
\]

with

\[
Z_j^I = a_j(K_j) + b_j(K_j) \{ \sigma_{jk} \sigma_{jl} + \sigma_{jk} \sigma_{jm} + \sigma_{jl} \sigma_{jm} + \sigma_{jl} \sigma_{jm} + \sigma_{jk} \sigma_{jm} \sigma_{jl} \sigma_{jm} \} + c_j(K_j) \sigma_{jk} \sigma_{jl} \sigma_{jm} \sigma_{jn}
\]

Where $\sigma_{jk}$, $\sigma_{jl}$, $\sigma_{jm}$ and $\sigma_{jn}$ are the nearest-neighbors of the spin $\sigma_j$ (figure 1(b)). The coefficients $a_j$, $b_j$ and $c_j$ are given by

\[
a_j(K_j) = \frac{1}{4} \{ \cosh(4K_j) + 4 \cosh(2K_j) + 3 \} \\
b_j(K_j) = \frac{1}{4} \{ \cosh(4K_j) - 1 \} \\
c_j(K_j) = \frac{1}{4} \{ \cosh(4K_j) - 4 \cosh(2K_j) + 3 \}
\]

one can identify $Z_j$ with $Z_j^I$ as,

\[
Z_j = e^{\delta^*}Z_j^I,
\]
where \( g \) is a regular function of the parameters. From equations (5), (10) and (14), we obtain

\[
\frac{b(K, D)}{a(K, D)} = \frac{b_1(K_1)}{a_1(K_1)},
\]

\[
\frac{c(K, D)}{a(K, D)} = \frac{c_1(K_1)}{a_1(K_1)}.
\]

Unfortunately, these equations give only one solution \( (D = -\infty, K = K_0) \), which is rather uninteresting since the model (1) is then trivially equivalent to the spin-\( -1/2 \) Ising model. To obtain the full phase diagrams in the \( (D, K) \)-plane, we have to reduce the number of equations (15) and (16). To this end we define a new quantity which simultaneously keeps the general expression of (5) not affected and makes the number of the above equations reduced. To achieve this task, we consider the quantity,

\[
\Omega_j = (1 + \sigma_k \sigma_j \sigma_{jm} \sigma_{jm}) Z_j = (a + c) + 2b (\sigma_k \sigma_j + \sigma_k \sigma_{jm} + \sigma_j \sigma_{jm} + \sigma_k \sigma_{jm} + \sigma_j \sigma_{jm} + \sigma_k \sigma_{jm} + \sigma_j \sigma_{jm})
\]

which leads to the following equation

\[
\frac{a + c}{b} = \frac{a_1 + c_1}{b_1}
\]

On the other hand, using equation (18) for the whole systems \( \{j \in \Lambda_2\} \), we obtain

\[
\sum_{\{j \in \Lambda_2\}} \prod_{\{j, k, j_m, j_{jm} \in \Lambda_1\}} (1 + \sigma_k \sigma_j \sigma_{jm} \sigma_{jm}) Z_j = e^{Ng} \sum_{\{j \in \Lambda_1\}} \prod_{\{j, k, j_m, j_{jm} \in \Lambda_1\}} (1 + \sigma_k \sigma_j \sigma_{jm} \sigma_{jm}) Z_j^L.
\]

Using (3) and (9), the partition function of the mixed spin-\( 1/2 \) and spin-\( S \) Ising model on square lattice is related to the partition function \( Z_j \) of the spin-\( 1/2 \) Ising model on square lattice as follows

\[
Z \left( \prod_{\{j, k, j_m, j_{jm} \in \Lambda_1\}} (1 + \sigma_k \sigma_j \sigma_{jm} \sigma_{jm}) \right)_H = e^{Ng} Z^L \left( \prod_{\{j, k, j_m, j_{jm} \in \Lambda_1\}} (1 + \sigma_k \sigma_j \sigma_{jm} \sigma_{jm}) \right)_{H'}
\]

which can be expressed in terms of free energies \( -\beta F = \ln Z, -\beta F^L = \ln Z^L \) as

\[
-\beta F + \ln \left( \prod_{\{j, k, j_m, j_{jm} \in \Lambda_1\}} (1 + \sigma_k \sigma_j \sigma_{jm} \sigma_{jm}) \right)_H = -\beta F^L + Ng
\]

\[
+ \ln \left( \prod_{\{j, k, j_m, j_{jm} \in \Lambda_1\}} (1 + \sigma_k \sigma_j \sigma_{jm} \sigma_{jm}) \right)_{H'}.
\]

where \( \langle \ldots \rangle_H \) and \( \langle \ldots \rangle_{H'} \) denote the canonical ensemble average defined by \( H \) and \( H' \), respectively. They are sums of even (greater than 2) numbered correlations functions on the \( \sigma \)-sublattices \( \Lambda_2 \) of the mixed spins Ising model and the spin-\( 1/2 \) Ising model, respectively.

In order to use the equation (20) to locate the critical line, we have to show that the singularities of the free energy \( F \), expressed in equation (23), coincide with those of \( F^L \) when the condition (20) is satisfied. This is the case since, as is well known, the correlations, expressed in equation (23), are analytic except at \( T = T_c \) for any finite number \( n \) of spins [14]. This behavior is extended even for \( n \to \infty \), which constitutes our conjecture. This later is confirmed by our calculations since our results are practically similar to those predicted by Monte Carlo approach for various integer and half-odd-integer values of \( S \). So the singularities of \( F \) are those of \( F^L \) (equation (23)). Therefore, the critical line is the solution of the equation (20) where \( K_j = K_{j1} = \frac{1}{2} \log (1 + \sqrt{2}) \) [5].
3. Results and discussions

In this section, let us discuss the exact phase diagrams of the mixed spin-1/2 and spin-S Ising model in the presence of a quadratic single-ion term acting upon S-spins. The transition temperature for a given S as a function of the reduced crystal field interaction $\Delta/J$ can be determined by solving numerically (20). Before proceeding to a discussion of the most interesting results of these phase diagrams, it is convenient to stress out that for all integer spins $S$ the phase diagrams are qualitatively similar and so are those corresponding to half-odd-integer spins $S$. Indeed, in figure 2, we represent the phase diagrams in the $(\Delta/J, K_BT_C/J)$ plane, for integer spins $S = 1, S = 2$ and $S = 3$. As can be expected, the magnetic domain is enlarged when the value of $S$ increases, and then the critical temperature is spin-dependent but when $\Delta/J$ approaches its critical value 2, the critical temperature becomes not sensitive to the value of $S$.

Further, as a result of competition between the crystal field ($\Delta > 0$) and the bilinear exchange interaction, the critical lines decrease and the critical temperature vanishes at $\Delta/J = 2$, which can be also be obtained by comparing the ground state energy of the ferromagnetic and paramagnetic phases. Therefore, the long range order is absent when $\Delta/J > 2$ for any value of the temperature. It is interesting to note when $\Delta/J = 2$, the critical temperature vanishes regardless of $S$ (integer). We should note that the system trivially reduces to a two states Ising model when $\Delta/J \to -\infty$. This situation is clearly seen in figure 3 in which $T^*$ tends to the Ising critical value $(K_J^2)^{-1}$ as $\Delta/J \to -\infty$, where we defined $T^*$ as $T_C/S$.

The present model is also interesting for $S$ half-integer. In figure 4, we represent the phase diagrams for half-odd-integer spins, $S = 3/2, S = 5/2$ and $S = 7/2$. As it is seen from the figure, the critical temperature $K_BT_C/J$...
decreases rapidly but doesn’t vanish for any value of $\Delta$. Therefore, the long range order exists for any value of the crystal field, below the critical line. Here, we can also notice that the system trivially reduces to two states Ising models for $\Delta / J \rightarrow \pm \infty$.

As is seen in figure 6, for relatively high values of the crystal field ($\Delta / J \gtrsim 1$), the critical temperature becomes not sensitive to the value of $S$. As depicted in figure 5, the normalized critical temperature $T^*$ tends to the exact value $T_C$ of the nearest neighbor spin-1/2 Ising model obtained by Onsager as $\Delta / J \rightarrow -\infty$. It indicates that our approach gives exact results and particularly it reproduces the exact solution performed by Onsager [5] in 1944 in the limit $\Delta / J \rightarrow -\infty$.

It is interesting to see that it is obvious if one can solve exactly the Hamiltonian (1) for $S = 1/2$, the solution has to be $K_C = 4K^C_l$. Following our procedure, for $S = 1/2$, the equation (20)

$$\frac{1}{4} \cosh (K_C) + \frac{1}{4} = \frac{1}{4} \cosh (4K^C_l) + \frac{1}{4}$$

has the same solution $K_C = 4K^C_l$. Also, the results shown in figure 6 for $S = 3/2$ are in complete agreement with the Monte Carlo studies [15]. On the other hand, we confirm that there is no tricritical point for any value of $\Delta$ and any integer value of $S$. In particular, we have to mention that our exact calculations confirm the result obtained previously in the case of spin $S = 1$ by Monte Carlo simulations [16–18] and, as one of us (N. Benayad) has predicted using renormalization-group method [19]. These results are depicted in figure 7. However, effective field theory (EFT) [20] and finite cluster approximation (FCA) [21, 22] give wrong results for a
coordination number greater than 3, since they predict the existence of a tricritical behavior. These wrong results arise from the decoupling approximations used for treating correlations between spins in those theories. Again, for $S = 2$, the previous simulation results $[23]$ are closely tied to those determined by our exact approach, which is shown in figure 8. We have to note that the figures 6, 7 and 8 drawn, respectively, for $S = 3/2$, $S = 1$ and $S = 2$ show a clear similitude between transition lines obtained in the framework of our approach and those obtained by Monte Carlo simulations. This gives a confirmation to our conjecture.

4. Conclusions

In this work we introduce a new approach, which enables us to investigate exactly the phase diagram of the mixed spin-1/2 and spin-$S$ Ising models on square lattice. It has been obtained for arbitrary spins $S (S > 1/2)$ in the presence of a quadratic single-ion term acting upon $S$-spins. We showed that, in both cases ($S$ integer and half-odd-integer), the current system exhibits only second order transition without tricritical behavior as suggested previously by approximated methods EFT $[20]$ and FCA $[21, 22]$. On the other hand, our exact results confirm those obtained using Monte Carlo simulations $[16–18]$ and renormalization group method $[19]$ did for $S = 1$. This is also the case for $S = 2$ $[23]$ and $S = 3/2$ $[15]$ using Monte Carlo simulations. Further, the phase diagrams of the present system exhibited a number of interesting phenomena, which depend on $S$. We found

Figure 6. Comparison between our results and those in $[15]$. The solid line expresses our exact solution of the present system ($S = 3/2$).

Figure 7. Comparison between our results and those in $[16]$ and $[19]$. The solid line expresses our exact solution of the present system ($S = 1$).
that, for $S$ integer, there is no long-order when the crystal field exceeds the value $\Delta/J = 2$; while for $S$ half-odd-integer the system remains order up to a certain temperature regardless of $\Delta/J$. Moreover, we showed that our new method recovers the Onsager’s solution when $\Delta/J \to -\infty$.

It is straightforward that our new exact approach presented in this work can be naturally extended to certain higher dimensional lattice situations where the critical point of the corresponding spin-1/2 Ising model is known with high accuracy. It is the substance of our next work, which will be submitted soon. Finally, we conclude that our method can be also used to solve exactly other models in statistical mechanics, unsolved up to now.

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Figure 8. Comparison between our results and those in [23]. The solid line expresses our exact solution of the present system ($S = 2$).