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Entropic order in frustrated antiferromagnets

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Abstract. We show that in frustrated spin-S Ising antiferromagnets the entropy can stabilize a partially-ordered ground state. In the model on the decorated square lattice partial long-range order exists for $S \ge 1$. For the model on the union jack lattice the partially-ordered ground state exists even for S = 1/2. For this lattice the ground-state problem is shown to be equivalent in a certain case to the staggered six-vertex model. We suggest that the model on the triangular lattice for $S > S_C \sim 3$ might exhibit a novel kind of partial ordering.

1. Introduction

The emergence of order is a very interesting phenomenon. Usually it is the minimization of energy which induces order at a sufficiently low temperature; however, in some systems the ordering appears through maximization of the entropy. Sometimes this leads to the so-called re-entrant phenomenon where a disordered ground state is replaced at a higher temperature by an ordered and more strongly degenerate phase [1]. Since entropic effects seem negligible at low temperatures, one may think that this mechanism cannot order the system at the ground state. However, there is a number of examples where the temperature does not seem to play an essential role, but nevertheless the ordering probably has an entropic origin. This is the case of the so-called Alder condensation in a system of hard disks [2] or its analogue in the system of electrons [3]. Such systems are, however, very complicated and their precise description is still lacking. Thus, it would be desirable to examine some simpler models which, hopefully, can mimic at least some of the features of the above-mentioned systems and which are still tractable with satisfactory precision. The relatively good understanding that we have of lattice spin models suggests that such simple models might be sought among them.

In this paper we provide some examples of frustrated antiferromagnetic models with partially ordered ground states, i.e. they contain a finite fraction of disordered spins. Such 'free' spins give a dominant contribution to the entropy of the ground state and actually stabilize the partially-ordered structures. We hope that such models, although far from being realistic, can shed some light on the interesting problem of entropic ordering.

In section 2 we examine the frustrated spin-S antiferromagnetic Ising model on the decorated square lattice. In the presence of the one-ion anisotropy, which restricts the 'free' spins to the decorating sites, the ground-state problem after decimation is equivalent to the S = 1/2 antiferromagnetic Ising model and, thus, the existence of partial long-range order can be inferred from the well known exact solution [4]. This example violates the conjecture made some years ago by Hoever *et al* [5] about the non-existence of spontaneous symmetry

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breaking in systems where ground-state configurations can be locally transformed one into another. Transfer-matrix calculations show that without one-ion anisotropy the system exhibits very similar behaviour.

In section 3 we study the frustrated model on the union jack lattice. Under a similar restriction on the location of the 'free' spins the problem is equivalent to the staggered six-vertex model. The existence of partial long-range order at any S > 0 is, thus, a simple consequence of the fact that the vertex model has a non-zero staggered polarization.

In section 4 we briefly describe the same model on the triangular lattice. This is the most interesting and, at the same time, the most difficult case. We map the ground-state problem into a certain SOS model. The flat phase in the SOS model, which exists above the roughening point, corresponds to the partially-ordered ground state. However, the mapping leaves some ambiguity concerning the nature of the partial order and we suggest that in this model it might be a novel kind of ordering. More detailed accounts concerning this case can be found elsewhere [6, 7]. Section 5 contains our conclusions.

2. Decorated square lattice

Let us consider the frustrated spin-S antiferromagnetic Ising model on the decorated square lattice. For the reasons specified below, we apply one-ion anisotropy on non-decorating sites. The Hamiltonian of this model is written as

$$H = J \sum_{(i,j)} [s_i s_j + t_{(i,j)} (s_i + s_j)] + D \sum_i s_i^2$$
(2.1)

where s_i , $t_{(i,j)} = -S$, -S + 1, ..., S denote spin operators on the non-decorating and decorating sites, respectively. The one-ion anisotropy constant is assumed to be non-positive $(D \leq 0)$. The symbol (i, j) denotes the pair of nearest neighbours on the square lattice.

The summation over bonds (i, j) can be regarded as a summation over triplets $(s_i, s_j, t_{(i,j)})$. Model (2.1) is fully frustrated since there is no configuration of a triplet $(s_i, s_i, t_{(i,j)})$ which would saturate all three of its bonds: $s_i s_i, s_i t_{(i,j)}, s_j t_{(i,j)}$.

Configurations $\{s_i, t_{(i,j)}\}\$ which minimize (2.1) for D < 0 are specified as follows (the case D = 0 will be considered separately).

(i) Spins on non-decorated sites can take only extremal values: $s_i = \pm S$. This condition minimizes the second term of the Hamiltonian.

(ii) If the neighbouring spins s_i , s_j are of the same sign, then the decorated spin $t_{(i,j)}$ has to be extremal and of the opposite sign (of s_i , s_j). When s_i , s_j are of opposite signs, then the decorated spin is 'free' and can take any admissible value.

One can easily perform the summation over decorating spins. Thus, the degeneracy of the ground state $\Omega(D < 0)$ can be written as

$$\Omega(D < 0) = \sum_{\{s_i, t_{(i,j)}\}} 1 = \sum_{\{s_i = \pm S\}} (2S + 1)^{k(\{s_i\})}$$
(2.2)

where $k(\{s_i\})$ is the number of antiferromagnetic pairs in the configuration $\{s_i\}$.

Let us notice that there is no restriction on the spins s_i , except that they have to be extremal. Thus, the last term is equivalent (up to an unimportant constant) to the partition function of the square lattice S = 1/2 antiferromagnetic Ising model with the coupling $\tilde{J} = \frac{1}{2} \ln(2S+1)$. As is well known [4], this model is critical for $\tilde{J}_C = \frac{1}{2} \ln(\sqrt{2}+1)$, which corresponds to $S_C = \sqrt{2}/2 \sim 0.7071...$ Thus, we obtain that only for S = 1/2 is the

ground state of the model disordered ($\tilde{J} < \tilde{J}_{C}$). For larger S the ground state is partially ordered; it consists of the ordered antiferromagnetic square backbone and 'free' decorating spins. The long-range order saturates in the limit $S \rightarrow \infty$. Moreover, let us notice that the spin S enters model (2.2) as a weight factor which, thus, might be regarded as a continuous parameter (model (2.1) is meaningful only for integer and half-integer S).

The emergence of the long-range order or, equivalently, spontaneous symmetry breaking in model (2.1) for $S \ge 1$ violates the conjecture made some years ago by Hoever *et al* [5]. On the basis of the results obtained for a certain class of frustrated models, they conjectured that if any two ground-state configurations can be transformed into one another through a sequence of local, energy-invariant transformations, then there is no spontaneous symmetry breaking in such a system. In other words, there should be no spontaneous condensation onto a particular configuration or set of configurations. It is easy to realize that any two ground-state configurations of model (2.1) can be transformed into one another through such local transformations by changing, simultaneously, at most five spins (one non-decorating and its four surrounding decorating spins).

Is the condition D < 0 essential for the appearance of long-range order in the ground state of model (2.1)? In the following we will consider the case D = 0. Since nondecorating spins s_i are no longer restricted to the extremal values the problem becomes more complicated. Ground-state configurations are specified as follows. For any triplet $(s_i, s_j, t_{(i,j)})$ at least one of its bonds has to be 'minimal', i.e. has to contribute energy $-JS^2$; condition (ii) is actually a special case of this condition. Of course, every groundstate configuration for D < 0 is a ground-state configuration for D = 0.

Although the summation over $t_{(i,j)}$ can be performed in the same way as in (2.2), the lack of condition (i) makes the resulting model more complicated:

$$\Omega(D=0) = \sum_{\{s_i\}} (2S+1)^{k(\{s_i\})}.$$
(2.3)

The only restriction which is imposed on the configurations $\{s_i\}$ is that at least one of the two neighbouring spins has to be extremal.

Let us notice that in (2.3) the weight of the configuration $\{s_i\}$ does not depend on the values of the non-extremal spins. Since the non-extremal spins cannot be nearest neighbours, we can easily perform summation over the non-extremal states. Thus, we arrive at the following three-state problem (zero represents a non-extremal state and ± 1 correspond to the extremal $\pm S$ states):

$$\Omega(D=0) = \sum_{\{u_i\}} (2S+1)^{k(\{u_i\})} (2S-1)^{l(\{u_i\})}$$
(2.4)

where $u_i = 0, \pm 1$ and at least one of the two neighbouring spins has to be non-zero. The symbol $l(\{u_i\})$ denotes the number of zero spins in the configuration $\{u_i\}$.

Due to the above 'hard-core' condition, the spin-1 model with a partition function which is equivalent to (2.4) has to possess infinite interactions. Nevertheless, the properties of model (2.4) can easily be examined by numerical methods [8]. Let us place model (2.4) on the strip of width L and infinite length, and with a toroidal boundary condition. The partition function in such a geometry is given as the largest eigenvalue λ_L of the corresponding transfer matrix. Moreover, the inverse of the correlation length can be written as $\xi_L^{-1} = \ln(\lambda_L/\lambda'_L)$, where λ'_L is the second largest eigenvalue. From renormalization-group arguments [9] one expects that the critical point S_C is approximately given as a solution of the equation

$$\frac{\xi_L}{L} = \frac{\xi_{L-1}}{L-1}$$
(2.5)

and the difference in widths of strips is unity due to the expected best convergence in this case. Condition (2.5) implies that the critical exponent ν which describes the divergence of the correlation length is given as

$$\frac{1}{\nu} = \frac{\ln\left(\frac{\partial\xi_L}{\partial S} / \frac{\partial\xi_{L-1}}{\partial S}\right)}{\ln(L/(L-1))} - 1$$
(2.6)

and the derivatives are calculated at the critical point obtained from (2.5). Although model (2.4) is antiferromagnetic, there is no magnetic field and, thus, it can be mapped into the ferromagnetic system by means of a simple gauge transformation. This procedure enables us to avoid alternation of results caused by the parity of L. Thus the elements of the transfer matrix T are given as

$$T(\alpha, \alpha') = (2S+1)^{k_1+k_2/2}(2S-1)^{l/2}$$
(2.7)

where k_1, k_2 denote the numbers of, respectively, vertical and horizontal ferromagnetic bonds between rows α and α' (the matrix T transfers in the vertical direction). The letter *l* denotes the total number of zero states in α and α' . Moreover, using the rotational and magnetic (up-down) symmetry [10] one can easily quasi-diagonalize the transfer matrix T. For L = 9 the largest block which has to be diagonalized numerically has the dimension 474.

Numerical results, i.e. the values of S_C and ν , are shown in table 1.

L	S _C	ν	η	с
3	0.688 408	0.930 67	0.26418	0.605 19
4	0.710993	0.949 10	0.25778	0.56439
5	0.724119	0.966 84	0.25477	0.537 54
6	0.730 127	0.978 57	0.25328	0.523 18
7	0.732 873	0.985 36	0.252.45	0.515 55
8	0.734238	0.989 36	0.25195	0.51120
9	0.734 986	0:991_88	0.25163	0.508 49
Extr.	0.73632	0.9988	0.2507	0.5015

Table 1. Values of S_C , ν , η and c calculated for several values of L. The extrapolations are based on the power-law convergence (2.10).

1.....

Assuming conformal invariance, one can derive [11] the relation between the critical exponent η , which describes the asymptotic behaviour of the correlation functions $\langle u_i u_j \rangle \sim |i - j|^{-\eta}$, and the correlation length

$$\eta = \frac{1}{\pi} \lim_{L \to \infty} \frac{L}{\xi_L}.$$
(2.8)

Moreover, the finite-size corrections to the free energy f_{∞} can be used [12] to obtain the central charge c:

$$-\frac{\ln\lambda_L}{L} = f_\infty - \frac{\pi c}{6L^2}.$$
(2.9)

The central charge is determined from the two estimations of the free energy for L and L-1.

The values of η and c calculated at $S_{\rm C} = 0.73632$ are also shown in table 1. Extrapolated values in the bottom row of table 1 are obtained using the data for L = 7, 8 and 9 and assuming power-law convergence:

$$x(L) = x(\infty) + AL^{-B}.$$
(2.10)

Our calculations strongly suggest that model (2.4) belongs to the Ising universality class with $\nu = 1$, $\eta = 0.25$ and c = 0.5. The small shift of S_C when comparing with the D < 0 case is easy to understand: allowing the non-extremal states on the non-decorating sites increases the fluctuations and thus a larger S is needed to induce partial long-range order.

3. Union jack lattice

In this section we describe the ground-state properties of the fully frustrated spin-S Ising antiferromagnet on the union jack (centred square) lattice. As in the previous section, the ground-state problem considerably simplifies in the presence of one-ion anisotropy. The Hamiltonian of the model is similar to (2.1) and is written as

$$H = J \sum_{(i,j)} s_i s_j + J \sum_{(i,k)} s_i t_k + D \sum_i s_i^2$$
(3.1)

where s_i and t_k are spin operators on eight- and four-coordinated sites respectively (see figure 1). The summation extends over nearest neighbours.



Figure 1. Elementary cell of the union jack lattice. Open and black circles denote eight- and four-coordinated sites respectively.

Since the summation over bonds (of equal strength) is equivalent to the summation over elementary triangles, the ground-state comfigurations are specified as follows.

(i) Spins on eight-coordinated sites can take only extremal values: $s_i = \pm S$.

(ii) On each elementary triangle, spins are such that they form at least one 'minimal' bond.

Decimating over four-coordinated sites, we find that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the ground state can be written as a partition function of the eight-vertex r such that the degeneracy of the degree can be written as a partition function of the degree can be written as a partition function of the degree can be written as a partition function of the degree can be written as a partition function of the degree can be written as a partition of the degree can be written

$$\Omega(D < 0) = 2 \sum_{\{\text{vert.}\}} \prod_{i=1}^{8} w_i^{k_i(\{\text{vert.}\})}$$
(3.2)

where the summation is over all vertex configurations {vert.}. The symbol k_i ({vert.}) denotes the number of vertices of the *i*th kind in a given configuration {vert.}. The factor two comes from the fact that configurations of opposite spins have the same vertex representation.

The weights w_i can easily be deduced from figure 2, which shows the standard assignment of vertex and spin configurations [13]. For the second vertex the decimated spin is 'free'. For the third and fourth vertices there is no value of the decimated spin which would satisfy (ii); for other vertices there is only one value of the decimated spin which satisfies (ii). Thus, we obtain

Figure 2. Assignment of vertex and spin configurations. Spin configurations with opposite spins map into the same vertex configuration. The decimated spins (not shown) are at the vertex.

Using certain symmetry properties [13], this eight-vertex model can be mapped into a staggered six-state vertex model [14]. On sublattices A and B the weights of this model are given as

A:
$$w'_1 = w'_2 = w'_3 = w'_4 = w'_6 = 1$$
 $w'_5 = 2S + 1$ $w'_7 = w'_8 = 0$ (3.4)

B:
$$w'_1 = w'_2 = w'_3 = w'_4 = w'_5 = 1$$
 $w'_6 = 2S + 1$ $w'_7 = w'_8 = 0.$ (3.5)

Let us notice that unequal weights w'_5 , w'_6 reflect the presence of some kind of staggered quadrupolar field. This field induces a non-zero polarization and the system has no phase transition (similarly to the Ising model in the magnetic field). The non-zero polarization of the vertex model translates in the spin language as an excess of antiferromagnetic configurations over the ferromagnetic ones. It implies that for any $S \ge 0$ the ground state of (3.1) has a spontaneously broken symmetry and is partially ordered.

It seems plausible that model (3.1) is ordered even at finite temperatures. Moreover, since the antiferromagnetic phase has a large entropy, we expect that this phase might appear at finite temperatures even in the case when the model is not fully frustrated (for example, when the interactions between $s_i s_j$ and $s_i t_k$ are slightly different) and the ground state has a different structure (e.g. ferromagnetic).

For S = 1/2 model (3.1) can be solved exactly, even at finite temperatures and such behaviour is indeed found [15, 16] (in this case the one-ion anisotropy term is, of course, irrelevant).

The solvability of model (3.1) for S = 1/2 comes from the fact that in this case the model satisfies the so-called 'free-fermion' condition [17]. It is easy to check that for S = 1/2 the vertex model (3.2) also satisfies this condition. Using known results [17], the residual entropy (per site) of model (3.1) can be written as

$$s(S = \frac{1}{2}) = \frac{1}{8\pi} \int_0^{2\pi} d\phi \ln\left[\frac{5}{2} + \sqrt{\frac{25}{4} - 4\cos^2\phi}\right] \sim 0.376\,996\dots$$
 (3.6)

Similar to the decorated square lattice, the ground-state problem becomes much more complicated when D = 0. However, we do not expect a qualitative difference with the case D < 0 at least for $S \ge 1/2$.

4. Triangular lattice

In this section we briefly describe our results concerning the ground-state structure of the spin-S antiferromagnetic Ising model on the triangular lattice. Our study was motivated by Monte Carlo simulations [18] of this model which indicate that the exponent η decreases for higher values of S (as shown by Stephenson [19], for S = 1/2 we have $\eta = 1/2$). These simulations suggest that above a certain value S_C the exponent η is presumably zero and the ground state is partially ordered: two sublattices form a honeycomb antiferromagnetic backbone and the third sublattice remains disordered.

The basic mechanism of this ordering is the same as that described in the previous sections: in an ordered structure the number of 'free' spins is the largest and, thus, such configurations prevail. In the case of the triangular lattice a given spin is 'free' when its six neighbours take the extremal values $\pm S$ alternately. We do not discuss the case when the external anisotropy fixes positions of 'non-extremal' spins; it seems to us that in this case it would not lead to a simpler problem.

The results of the previous sections suggest that by decimating over 'free' spins we can relate this ground-state problem with some other, hopefully simpler, model. Since spins taking non-extremal values cannot be nearest neighbours, there is $(2S - 1)^{k([s_i])}$ configurations which have the same 'extremal' backbone and differ only in values of non-extremal spins, $k(\{s_i\})$ is the number of non-extremal spins in the ground-state configuration $\{s_i\}$. Thus, we obtain

$$\Omega = \sum_{\{u_i\}} (2S - 1)^{k(\{u_i\})}$$
(4.1)

where $u_i = 0$ corresponds to a non-extremal state and $u_i = \pm 1$ correspond to $s_i = \pm S$ respectively. The summation is performed over all configurations $\{u_i\}$ which satisfy the (ground-state) condition that each elementary triangle contributes the energy -J, where J is a coupling constant.

The emergence of partial ordering is equivalent to the phase transition in the threestate model (4.1). However, this model is very complicated and it is difficult to predict its behaviour, but we can express the degeneracy of the ground state Ω in another way.

First let us notice that we are led in a natural way to the representation (4.1) by the following mapping: every extremal state $s_i = \pm S$ is mapped into $u_i = \pm 1$, respectively, and every non-extremal state is mapped into $u_i = 0$. The weight $(2S-1)^{k(\{u_i\})}$ gives the number of the configurations $\{s_i\}$ which are mapped into the same configuration $\{u_i\}$. Now, let us modify the second rule of this transformation and map non-extremal states into $u_i = \pm 1$ depending on the type of the surrounding of the non-extremal state and let us notice that for a non-extremal spin there are only two types of surroundings (they are related by the up-down symmetry). In this mapping the weight is given as $(2S)^{k'(\{u_i\})}$, where $k'(\{u_i\})$ is the number of 'favoured' spins in a given configuration $\{u_i\}$ (for a more detailed formulation see our other papers [6, 7]). Thus, we can write

$$\Omega = \sum_{\{u_i\}} (2S)^{k'(\{u_i\})}.$$
(4.2)

In (4.2) the summation is performed over all ground-state configurations of the S = 1/2 model. For S > 1/2 they are, however, unequally weighted. Although multi-spin correlations of spin operators are needed to determine $k'(\{u_i\})$, such a transformation has the important merit that as a result we obtain a two-state model $(u_i = \pm 1)$.

Following Blöte and Hilhorst [20], we can map configurations $\{u_i\}$ into a certain sos model. Although model (4.2) is unsolvable, the relation with the SOS model enables us to predict the behaviour of model (4.2). First, let us notice that by increasing S we favour flat configurations (actually, the corrugated ones). Thus, we decrease the fluctuations of the SOS model and, consequently, the parameter S can be related with the inverse of the temperature of the renormalized Gaussian model (we make the commonly accepted assumption that the SOS model in the rough phase renormalizes into the Gaussian model). Moreover, in our mapping the correlations functions $\langle u_i u_j \rangle$ are Fourier transforms of the height variables of the sos model. Since the spin-wave operators of the Gaussian model can be related [21] to the spin model (4.2), the precise dependence of the renormalized Gaussian temperature on the spin S can be eliminated and a number of results concerning the stability of model (4.2) can be obtained [6, 21]. In particular, we find that at the roughening transition we have $\eta = 1/9$. The partial ordering on the triangular lattice thus translates as a flat phase of this SOS model.

The Monte Carlo simulations [18] and our transfer-matrix calculations show that $\eta = 1/9$ for $S \sim 3$, and stability analysis suggests that the critical value of spin has to be close to this value. However, for S = 7/2 Monte Carlo simulations give $\eta = 0.069 \pm 0.003$, which according to the stability analysis is impossible. In the Monte Carlo simulations the measured quantities were spin-S correlation functions $\langle S_i S_j \rangle$. One possible explanation of such a discrepancy might be the inaccuracy of the Monte Carlo method or a failure of the assumption about the renormalization into the Gaussian model; but we can propose yet another explanation. Namely, the relation between order parameters in the original spin-S antiferromagnet is critical with $1/9 > \eta > 0$, and at the same time the SOS model (4.2) is in the long-range-ordered phase. This kind of order would be in some sense similar to the chiral order where the single spin averages are zero and we have a multi-spin order parameter. Although our arguments are rather speculative, the possibility of the existence of such a novel kind of ordering seems to be very interesting and certainly worth further studies.

5. Conclusions

The ground-state configurations of all frustrated spin-S Ising antiferromagnets studied in this paper have to satisfy only one rule: each elementary triangle has to have at least one pair of extremal but opposite spins. This simple rule leads to the strong degeneracy of the ground state, and for a sufficiently large S partially ordered structures appear.

However, more detailed properties of the ground state strongly depend on the geometry (i.e. lattice structure) in which this rule has to be satisfied. In some cases the ground-state problems of models studied in this paper are equivalent to some other models with behaviour that is known exactly or can be predicted: the S-dependent finite temperature square lattice Ising model, the staggered six-vertex model or the SOS model.

Of course, the frustrated models examined in this paper cannot be regarded as being realistic models of entropic ordering. We can only suggest a certain link using some speculative arguments. Let us represent occupied and empty sites by $s_i = \pm S$ respectively. It is possible to imagine that in certain systems, due to, for example, geometrical constraints, certain configurations of particles (e.g. when occupied and empty regions are alternately placed) might create regions where particles obtain some extra freedom and other (non-extremal) states are needed to describe such favourable configurations. At the crude level

such systems might be described by models similar to the ones considered in this paper. Let us notice that in physical continuous systems the accessible space is usually isotropic. Keeping in mind the strong dependence on the geometry, we have to look for the lattice model which also should be, in some sense, isotropic. Among the examined models, only in the model on the triangular lattice are all sites equivalent and, thus, in the context of entropic ordering, this model seems to be the 'least unrealistic'. However, at the same time this is the most difficult case and even the very nature of the partial order might turn out to be surprising.

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