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1982 J. Phys. A: Math. Gen. 15 3561

(http://iopscience.iop.org/0305-4470/15/11/034)

J. Phys. A: Math. Gen. **15** (1982) 3561–3569. Printed in Great Britain

Ground state degeneracy of the Ising antiferromagnets in the maximum critical field

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Received 19 April 1982

Abstract. We demonstrate that all Ising antiferromagnets with arbitrary many-neighboured interaction and in the maximum critical field, have highly degenerate ground states accompanied with non-zero residual entropies. The residual entropies vanish when the range of interaction tends to infinity. The proof is realised by an explicit calculation in the case of a one-dimensional many-neighboured Ising antiferromagnet, and by establishing bounds for the residual entropy in the case of an Ising system situated on a lattice with arbitrary number of dimensions. We also show that the established bounds may serve as estimates of frequently unknown actual values of the residual entropy.

1. Introduction

The ground states of the antiferromagnetic Ising systems in an external magnetic field have been studied by many authors. Brooks and Domb (1951) noted that the square Ising model with the antiferromagnetic nearest-neighbour (NN) coupling J in the critical field $H_c = 4J$ should have a non-zero entropy at the absolute zero temperature. They estimated that, at $H = H_c$ and $T = 0$, the model entropy retains more than 50% of its maximum value $Nk_B \ln 2$, where N is the number of spins and k_B is the Boltzmann constant. The residual entropy results from the competition between parallel and antiparallel ordering of spins, caused by the external field and exchange interaction, respectively. In the pioneer review of the theory of cooperative phenomena, Domb (1960) pointed out that the Ising chain with the antiferromagnetic NN coupling J, in the critical field $H_c = 2J$, has also a residual entropy. From an exact expression for the free energy, Domb (1960) found that the entropy per spin in the limit $T \to 0$ is equal to $k_B \ln[(1+\sqrt{5})/2]$, for $H_c = 2J$. On the other hand, Bonner and Fisher (1962) observed that the ground state degeneracy of the Ising chain can be expressed as a term of the Fibonacci sequence, whose Nth term, for large N, can be represented by $[(1+\sqrt{5})/2]^N$ (see also Jarić and Milošević 1974). Subsequently, there have appeared many papers (see, e.g., Bader and Schilling 1981, and references quoted therein), which established ground state phase diagrams for the one-, two- and three-dimensional Ising models with arbitrary first-, second- and third-neighbour interactions. Thereby they also provide critical fields for the Ising models with three successive antiferromagnetic interactions. However, none of these papers furnish an answer to the question about possible values of the corresponding residual entropies.

In the present paper we study the Ising systems with arbitrary antiferromagnetic interactions of range k. Such systems may have many critical fields, whose existence

0305-4470/82/113561+09$02.00

and order depend on the lattice topology and characteristics of the set $\{J_1, J_2, \ldots, J_j, \ldots, J_k\}$, where J_j is the jth-neighbour interaction. However, there always exists a maximum critical field, such that in a higher field *all* spins are aligned in the field direction, whereas in a lower field *some* spins are flipped in the opposite direction. We show that this threshold field is accompanied with a finite residual entropy, provided that the antiferromagnetic interaction is of a finite range. In § 2 we calculate exactly the ground state degeneracy of an Ising chain, with arbitrary antiferromagnetic interaction of range k, and in the presence of the maximum critical field. From the obtained results, it follows that the corresponding residual entropy vanishes when $k \to \infty$. Although in the case of two- and three-dimensional Ising antiferromagnets one can hardly calculate exact values of the residual entropy, we establish in § 3 its lower and upper bound. These bounds confirm that there is no residual entropy when the interaction range tends to infinity. Furthermore, the established bounds provide limits for approximate calculations of the residual entropy. Thus, for instance, it follows that the residual entropy of the NN square Ising antiferromagnet cannot be lower than 51.7% of the entropy maximum, in agreement with the classic result of Brooks and Domb (1951). In § 4 we give further numerical analysis of the obtained results, together with an overall discussion and pertinent conclusions.

2. One-dimensional antiferromagnets

We start our discussion with a general Ising model of an antiferromagnet

$$\mathscr{H} = \sum_{j=1}^{k} J_j \sum_{\langle mn \rangle_j} S_m S_n - H \sum_{n=1}^{N} S_n, \tag{1}$$

where J_j is the jth-neighbour antiferromagnetic interaction ($J_j > 0$), $\langle mn \rangle_j$ stipulates that the summation is restricted over those pairs of lattice sites which are the jth neighbours, and S_m is the conventional Ising-spin variable ($S_m = \pm 1$). The applied field is positive ($H > 0$), and it defines the positive (upward) direction of spins.

In order to study ground states of a system described by the Hamiltonian (1) we introduce the following variables: $n(\downarrow)$ number of spins turned down, and $n_j(-)$ number of negative terms in the sum restricted over the jth neighbours, for a given configuration of spins. Thereby, assuming periodic boundary conditions, the Hamiltonian (1) can be written in the form

$$\mathscr{H} = \sum_{j=1}^{k} [z_j N/2 - 2n_j(-)]J_j - H[N - 2n(\downarrow)], \tag{2}$$

or

$$\mathscr{H} = \text{const.} + Y_1 + Y_2, \tag{3}$$

with

$$Y_1 = -2 \sum_{j=1}^{k} n_j(-) J_j, \tag{4}$$

and

$$Y_2 = 2Hn(\downarrow), \tag{5}$$

where z_j is the number of jth neighbours per site of a given lattice. Henceforth we adopt $n(\downarrow)$ as a basic variable, which may vary from zero to N. Of course, each $n_j(-)$ is a function of both $n(\downarrow)$ *and* arrangement of spins turned down. But, as we are interested in the minimum values of $\mathcal{H}$, we will be concerned only with those arrangements of spins that give the largest possible values of $n_j(-)$, yielding minimal values of Y_1 in (3).

The exchange interaction of a particular spin turned down is effective within a region that contains $\Sigma_{j=1}^{k} z_j$ lattice sites. If this region, for every spin turned down, is filled up by spins turned up, all quantities $n_j(-)$ will have their maximum values

$$n_j(-) = z_j n(\downarrow), \qquad j = 1, 2, \ldots, k. \tag{6}$$

These equations will be satisfied providing that

$$n(\downarrow) \in [0, N/r], \tag{7}$$

where r is characteristic of a given lattice. The ratio N/r represents the maximum possible number of spins turned down distributed on the lattice so that no two of them interact, that is to say r is related to the smallest distance between two non-interacting spins in the lattice. Thus, in the case of a one-dimensional lattice $r = k+1$ for arbitrary k, whereas in the case of the two-dimensional square lattice $r = 2, 4, 6, 9$ for $k = 1, 2, 3, 4$, respectively. Similarly, in the case of the simple cubic lattice $r = 2, 4, 8$ for the three successive ranges of interaction $k = 1, 2, 3$.

Since in the interval (7), all $n_j(-)$ have their maximum values, the function Y_1 has the largest decrement per spin turned down

$$-\Delta Y_1 = 2 \sum_{j=1}^{k} z_j J_j. \tag{8}$$

This decrement of the function Y_1 can be compensated by an increment of the function Y_2 if

$$H = \sum_{j=1}^{k} z_j J_j. \tag{9}$$

Therefore, in an external magnetic field equal to (9), the Hamiltonian (3) will have the constant value for *all* $n(\downarrow)$ in the interval (7). This value of $\mathcal{H}$ is then the ground state energy, as for $n(\downarrow)$ outside the interval (7) the increment of Y_2 is larger than the decrement of Y_1, and consequently $\mathcal{H}$ becomes larger. Similarly, one can deduce that the magnetic field (9) is the maximum critical field (Kanamori 1966). Indeed, for H larger than (9) the increment of Y_2 is larger than the decrement of Y_1, and the Hamiltonian would increase if the spins started to turn down. On the other hand, if H is smaller than (9) the function Y_1 decreases faster than Y_2 increases, and the ground state would be outside the interval (7), where Y_1 should acquire a smaller decrement.

In the case of a one-dimensional Ising chain equations (6) become uniform

$$n_j(-) = 2n(\downarrow), \qquad j = 1, 2, \ldots, k. \tag{10}$$

These new equations are satisfied if every spin turned down is followed by at least k spins turned up. Such a configuration of spins is possible providing that

$$n(\downarrow) \in [0, N/(k+1)]. \tag{11}$$

Therefore, in the critical field

$$H = 2 \sum_{j=1}^{k} J_j, \tag{12}$$

the ground state is highly degenerate. The corresponding degeneracy is a sum of degeneracy elements, each one being the number of ways in which a group of $n(\downarrow)$ spins turned down can break the remaining group of spins aligned up, so that every spin turned down is followed by at least k spins turned up. This number is the binomial coefficient formed of the numbers $N - kn(\downarrow)$ and $n(\downarrow)$. Thus the ground state degeneracy is

$$P = \sum_{n(\downarrow)=0}^{[N/(k+1)]} \binom{N - kn(\downarrow)}{n(\downarrow)}, \tag{13}$$

where $[N/(k+1)]$ is the integral part of $N/(k+1)$. Knowing P we can calculate the concomitant dimensionless entropy in the thermodynamic limit

$$\sigma = \lim_{N\to\infty} \left(\frac{1}{N} \ln P\right). \tag{14}$$

It suffices to find the largest term in the sum (13). This could be the binomial coefficient with $n(\downarrow)$ that follows from $2n(\downarrow) = N - kn(\downarrow)$. However, since $N - kn(\downarrow)$ is a decreasing function of $n(\downarrow)$, it should be a binomial coefficient with $n(\downarrow)$ less than $(N - kn(\downarrow))/2$. For such $n(\downarrow)$ the inequality

$$\binom{N - kn(\downarrow)}{n(\downarrow)} < \binom{N - k(n(\downarrow)+1)}{n(\downarrow)+1} \tag{15}$$

should turn into the opposite inequality. Hence, if we denote the ratio of the correct $n(\downarrow)$ and N by x, the latter should be the smallest positive root of the equation

$$x(1-kx)^k = (1-kx-x)^{k+1}, \tag{16}$$

which follows from (15) for very large N. When we know the solution of (16), we obtain the entropy

$$\begin{aligned} \sigma &= \ln\left\{ \lim_{N\to\infty} \binom{N - kxN}{xN}^{1/N} \right\} \\ &= \ln\left\{ \frac{(1-kx)^{1-kx}}{x^x (1-kx-x)^{1-kx-x}} \right\}, \end{aligned} \tag{17}$$

or, by using equation (16),

$$\sigma = \ln\left\{ \frac{1-kx}{1-kx-x} \right\}. \tag{18}$$

Therefore, we have obtained an exact formula for the residual entropy of the antiferromagnetic Ising chain, with arbitrary many-neighboured interaction, in the maximum critical field. When the range of the interaction tends to infinity, it follows from (18) that the residual entropy vanishes. In the special case $k = 0$, i.e. when there is no interaction between spins, equation (16) and formula (18) give the expected result $\sigma = \ln 2$. For $k = 1$ one can find that the smaller solution of (16), $x = (5-\sqrt{5})/10$, gives $\sigma = \ln\{(1+\sqrt{5})/2\}$, in agreement with the result of Domb (1960).

3. Bounds of the residual entropy

In the case of an Ising antiferromagnet situated on a lattice with dimension $d>1$, one can hardly calculate exact values of the residual entropy. However, we shall demonstrate that its lower and upper bound can be found. The idea springs from the conditions (6). They are satisfied if every spin turned down succeeds in forbidding other spins turned down to occupy any of its $\Sigma_{j=1}^{k} z_j$ neighbouring sites. If $n(\downarrow)=1$ then the next spin flipped down finds $p=\Sigma_{j=1}^{k} z_j+1$ forbidden sites in the lattice. This means that the number of forbidden sites per spin turned down cannot be larger than p. On the other hand, the same number, which we will hereafter denote by f, cannot be smaller than r, where r is defined by (7). In fact, when $n(\downarrow)=N/r$ all sites in the lattice are forbidden, and thus the minimum of f is r. Shortly, when $f=p$ the forbidden zones that surround the spins turned down do not overlap, whereas for $f=r$ there is the largest possible overlapping between the zones. As we are dealing with the isotropic exchange interactions, it is appropriate to term these zones the forbidden spheres.

Knowing the two extreme values of f (p and r) one can calculate the lower and upper bound of the residual entropy. Let $n(\downarrow)$ be a number from the interval (7). If every spin turned down forbids exactly p lattice sites to be occupied by any other spin turned down, then the number of possible configurations, for fixed $n(\downarrow)$, is given by

$$C(n(\downarrow))=\frac{N(N-p)(N-2p)\ldots(N-p(n(\downarrow)-1))}{n(\downarrow)!}. \tag{19}$$

This number is smaller than the actual number of possible configurations for a given $n(\downarrow)$, since it does not include, for instance, the apparent possibility that two spins turned down may have a common forbidden site, which cannot be occupied by a third spin turned down. Thus $C(n(\downarrow))$ is an element of the lower bound of the ground state degeneracy

$$P_l=\sum_{n(\downarrow)=1}^{[N/r]} C(n(\downarrow)). \tag{20}$$

In order to calculate the corresponding lower bound for the residual entropy, in the thermodynamic limit, we look for the maximum $C(n(\downarrow))$. It turns out to be $C(n(\downarrow))$ for the smallest of $n(\downarrow)$ that cause violation of the inequality $C(n(\downarrow))<C(n(\downarrow)+1)$. One can verify that, for large N, $n(\downarrow)=[N/(p+1)]$ satisfies this requirement. The corresponding degeneracy element can be written in the form

$$C(m)=\prod_{i=0}^{m-1}\{(N-pi)/(i+1)\} \tag{21}$$

where

$$m=[N/(p+1)],\qquad p=\sum_{j=1}^{k} z_j+1. \tag{22}$$

Hence we find the entropy lower bound

$$\begin{aligned}\sigma_l&=\lim_{N\to\infty}\left\{\frac{1}{N}\ln C(m)\right\}\\&=\lim_{N\to\infty}\left\{\frac{1}{N}\sum_{i=0}^{m-1}(\ln(N-pi)-\ln(i+1))\right\},\end{aligned} \tag{23}$$

which is equivalent to

$$\sigma_l = \lim_{N\to\infty} \left\{ \frac{1}{N} \int_0^m (\ln(N-pt) - \ln(t+1))\, \mathrm{d}t \right\} \tag{24}$$

so that, after a few steps of a simple calculation, we finally obtain

$$\sigma_l = (1/p)\ln(p+1). \tag{25}$$

From this formula we can infer that *all* Ising antiferromagnets described by (1), in the appropriate critical fields (9), *have non-zero residual entropies*, which might tend to zero when the range of interaction tends to infinity. Indeed, p is of order of k, and according to (25) σ_l vanishes when $p \to \infty$. This would mean that residual entropies vanish as well, if their upper bounds also tend to zero when the range of interaction tends to infinity. We shall demonstrate that the latter condition is satisfied.

Let us now assume that, for arbitrary $n(\downarrow)$ in the interval (7), every spin turned down forbids exactly r lattice sites to be occupied by any other spin turned down. Furthermore, we assume that the diminished forbidden spheres ($r<p$) do not overlap. As r is the lowest number of f introduced above, the assumptions imply a strong tendency of the spins turned down to approach each other at the smallest allowed distances. This brings about a great number of possible configurations, and results in an overestimation of the ground state degeneracy. The latter, for a given $n(\downarrow)$, can be evaluated in the same way as in the case $f=p$. Repeating all neccesary steps we obtain an upper bound for the residual entropy

$$\sigma_\mathrm{u} = (1/r)\ln(r+1). \tag{26}$$

As we have already observed, the precise value of r depends on the lattice geometry. In the case of a one-dimensional lattice $r=k+1$. However, in the general case it is evident that r cannot be smaller than the range of the antiferromagnetic interaction k, since r is in fact related to the smallest distance between two non-interacting spins in the lattice. Thus, when the range of interaction tends to infinity, r also becomes infinite, and, according to (26), the upper bound of the residual entropy vanishes. This vindicates the conclusion that the residual entropy itself vanishes when $k\to\infty$.

4. Discussion

The first result we have obtained in this paper is an exact formula for the residual entropy of the antiferromagnetic Ising chain with many-neighboured interaction in the maximum critical field (see equations (12), (16) and (18)). This result confirmed an interesting conceptual fact, that consists in the vanishing of the residual entropy when the range of interaction k tends to infinity. This fact is interesting in relation to the theoretical foundations of the third law of thermodynamics, and may be useful in the study of the frustration phenomena (Toulouse 1977), since an Ising antiferromagnet in the critical field is a fully frustrated system. It should also be observed that calculation of the residual entropy as the zero-temperature limit of the first derivative of the corresponding free energy for $k>2$ is practically futile, for such a calculation requires analysis of an eigenvalue problem of order 2^k (Dobson 1969). In that respect our first result has its own virtue. It corroborates the statement of Aizenmann and Lieb (1981), that the zero-temperature entropy should be computable by a direct counting of the ground state configurations, although the calculation is not usually trivial.

The direct calculation of the residual entropy in the case of two- and three-dimensional Isihg antiferromagnets is almost insuperable. Thus, we searched for the entropy bounds, in order to extend results established for one-dimensional systems. The obtained lower and upper entropy bound, equations (25) and (26) respectively, substantiate the statement that residual entropy of an arbitrary Ising antiferromagnet, in the maximum critical field, vanishes when the range of interaction tends to infinity.

Here one can raise the question as to whether the entropy bounds may serve the purpose of estimating the residual entropy of a given Ising antiferromagnet. Since the established bounds (25) and (26) are quite general, we shall look at the one-dimensional Ising antiferromagnets first. Comparison of the exact values for the residual entropies, obtained from numerical solutions of equation (16), and the corresponding boundary values reveals that the bounds impose acceptable intervals for the actual values (see table 1). It can be also noticed that the upper bound is always worse than the lower one. For instance, when $k=8$ the deviation of the lower bound from the exact value is about 12%, whereas the deviation of the upper bound is more than 32%. However, a simple argument can be brought forward so as to establish a better formula for the entropy upper bound.

Table 1. The residual entropy of the one-dimensional Ising antiferromagnet in the critical field (12), with an interaction of range k. σ_l, σ_{exact}, σ'_{u} and σ_{u} are calculated according to formulae (25), (18), (29) and (26), respectively.

k	σ_l	σ_{exact}	σ'_{u}	σ_{u}
1	0.4621	0.4812	0.5011	0.5493
2	0.3584	0.3822	0.4024	0.4621
3	0.2971	0.3223	0.3403	0.4024
4	0.2558	0.2812	0.2971	0.3584
5	0.2259	0.2509	0.2649	0.3243
6	0.2030	0.2275	0.2398	0.2971
7	0.1848	0.2087	0.2196	0.2747
8	0.1700	0.1932	0.2030	0.2558

The ground state degeneracy can be considered as a sum of elements which are certain functions of $n(\downarrow)$. For the residual entropy, in the thermodynamic limit, it is the largest of these elements that matters the most. Such an element is defined by a particular value of $n(\downarrow)$, which, on the other hand, defines a particular f, i.e. a definite number of forbidden lattice sites per spin turned down. We shall argue that this number should satisfy the inequality

$$f>(3p+1)/4, \tag{27}$$

where p is given by (22). To this end let us define the quantity

$$\omega(n(\downarrow))=(pn(\downarrow)-fn(\downarrow))/n(\downarrow)=p-f \tag{28}$$

which measures the overlapping of the forbidden spheres. For two spheres the maximum of ω would be $\omega(2)=(p-1)/4$, since two spheres can have in common at most $(p-1)/2$ forbidden lattice sites. When there are many spheres, $\omega(n(\downarrow))$ should not be larger than $(p-1)/4$ (or f smaller than $(3p+1)/4$), since other values of $\omega(n(\downarrow))$

imply appreciable clustering of spheres, which has smaller possibilities of producing a large number of configurations. Thus, values of f that may bring about large degeneracy should satisfy (26). If we now assume that every spin turned down is surrounded by a sphere that contains $f=(3p+1)/4$ forbidden sites, and if we assume that these new spheres do not overlap at all, we will allow for an overestimation of the ground state degeneracy. The corresponding residual entropy can be calculated in the same way as σ_l and σ_u in the preceding section. The final result is

$$\sigma_u' = [4/(3p+1)] \ln \{[(3p+1)/4]+1\}. \tag{29}$$

In table 1 we present values of σ_u' calculated for the one-dimensional Ising antiferromagnets. One can readily notice that formula (29) provides a fairly acceptable upper estimate of the residual entropies. Thus, on the whole, table 1 makes us confident to use (25) and (29) for estimation of the residual entropy in cases when its exact value is not known. In table 2 we present the corresponding estimates in the case of the square Ising antiferromagnet. We can see that for $k=1$ the lower and upper estimates, in perfect agreement with the classic prediction of Brooks and Domb (1951), retain respectively 51.7% and 58% of the entropy maximum (ln 2). In table 3 we give, for the sake of completeness, similar estimates for several standard two-dimensional and three-dimensional lattices. The importance of these estimates, beside the physical insights they may provide, lies in the fact that they cannot easily be obtained by the series expansion methods about zero temperature (see, e.g., Domb 1974). The latter are based on counting the configurations that deviate from a perfect order, and this does not exist in the critical field.

Table 2. The lower (σ_l) and upper (σ_u') bounds of residual entropy of the square Ising antiferromagnet in the maximum critical field (9), with an interaction of range k.

k	σ_l	σ_u'
1	0.3584	0.4024
2	0.2558	0.2971
3	0.2030	0.2398
4	0.1472	0.1771

Table 3. The lower and upper bounds of the residual entropies of the Ising antiferromagnets situated on standard lattices ($k=1$).

Lattice	σ_l	σ_u'
Triangular	0.2971	0.3403
Kagomé	0.3584	0.4024
Honeycomb	0.4024	0.4452
Simple cubic	0.2971	0.3403
Body-centred cubic	0.2558	0.2971
Face-centred cubic	0.2030	0.2398

Acknowledgments

The authors are grateful to Dr M Mijatović for his kind help in the numerical analysis of the results obtained in this work. One of the authors (SM) is indebted to Professors

H W Capel, J H Perk, P W Kasteleyn, and to Dr G R W Quispel, for stimulating discussions during his visit to the Lorentz Institute (Leiden).

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