A THERMODYNAMIC APPROXIMATION OF THE GROUND STATE OF ANTIFERROMAGNETIC HEISENBERG SPIN- $\frac{1}{2}$ LATTICES

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The exact ground state of finite Heisenberg spin- $\frac{1}{2}$ lattices is studied. The coefficients of the so-called Ising configurations contributing to the ground state are approximated by Boltzmann-like expressions. These expressions contain a parameter that may be related to an inverse temperature.

1. Introduction

Long-range order and symmetry breaking in antiferromagnetic Heisenberg systems ($S = \frac{1}{2}$) have been investigated extensively in the past by several authors. Because of the difficulty of the problem, exact results are scarce.

Until now only the linear chain has been solved exactly. Bethe [1] first calculated the eigenvector in 1931 and in 1938 Hulthén [2] found the ground-state energy.

For two and three dimensions the situation is much more complicated. No exact solution exists and one has to use approximations. We will derive from exact finite cell calculations an approximation method for the ground state that can also be used for large spin- $\frac{1}{2}$ systems. In this approximation the ground state will be written as a linear combination of Ising states, as explained in the next section. In this expression the coefficients obey a Boltzmann-like distribution over the secular part of the energy (Ising energy). In this description a

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variational parameter β can be introduced that represents the inverse of a "temperature".

Apart from calculations for non-frustrated lattices, as e.g. the linear chain and the square lattice, we also pay attention to the frustrated triangular lattice.

2. Numerical methods

The Hamiltonian of an antiferromagnetic Heisenberg system can be written as

$$H = H_{xx} + H_{yy} + H_{zz} = \sum J(S_{ix}S_{jx} + S_{iy}S_{jy} + S_{iz}S_{jz}), \quad (1)$$

where the summation is over all nearest-neighbour pairs (i, j) and J is the exchange integral, which is positive. It will be given the value 1 in this paper. $S_{i\xi} = \frac{1}{2}\sigma_{i\xi}$ is a component of the spin operator of particle i and $\sigma_{i\xi}$ is the corresponding Pauli spin matrix ($\hbar = 1$, $\xi = x, y, z$).

The ground state $|\psi_0\rangle$ is given by the equation

$$H|\psi_0\rangle = E_0|\psi_0\rangle \quad (2)$$

where E_0 is the lowest eigenvalue. This state can be expanded in terms of the complete set of eigenfunctions of H_{zz} , which are of the form $|n\rangle = |\dots + - + - \dots\rangle$. We call these states Ising kets. These states are eigenfunctions of the individual S_{iz} , with $S_{iz}|\dots, \pm, \dots\rangle = \pm\frac{1}{2}|\dots, \pm, \dots\rangle$. Lieb and Mattis [3] showed that the ground state of non-frustrated Heisenberg systems is a $S=0$ state. So we may restrict ourselves, for these cases, to the subspace in which the z -component of the total spin S equals zero ($M=0$). We can write

$$|\psi\rangle = \sum c_n |n\rangle, \quad (3)$$

with for every $|n\rangle$

$$\sum_i S_{iz} |n\rangle = 0, \quad (4)$$

and with

$$\sum_n |c_n|^2 = 1, \quad (5)$$

so $|\psi\rangle$ is normalized.

If the number of spins is large the solution of (2) is hard to determine, even with numerical methods. We solved (2) for a small number of spins, up to 16, numerically, and tried to derive from this solution an approximation method for large systems. Every state $|n\rangle$ has an Ising energy E_n that is given by

$$H_{zz}|n\rangle = E_n|n\rangle, \quad (6)$$

and this E_n represents a measure of the number of parallel or antiparallel neighbour pairs. For all states $|n\rangle$ in the exact ground state, with the same Ising energy, we calculate the root mean square of the coefficients of these states $|n\rangle$. This root mean square is defined by

$$\bar{a}(E) = \left(\sum^* |c_n|^2 \right)^{1/2} / \left(\sum^* 1 \right)^{1/2}, \quad (7)$$

in which Σ^* represents the restricted sum over all states $|n\rangle$ with $E_n = E$. This function of the Ising energy will be calculated for different lattices in the next sections.

As a first approximation to the exact solution we make a variational state

$$|\Psi_{tr}\rangle = \sum_n (-)^p a(E_n) |n\rangle, \quad a(E_n) > 0, \quad (8)$$

in which all states $|n\rangle$ with the same Ising energy have the same variational coefficient $a(E_n)$. Here we have applied the Marshall rule. This Marshall rule [4] says that if two neighbouring spins in state $|n\rangle$ have opposite orientation e.g. $|\dots +, - \dots\rangle$ and the spins are interchanged to give $|n'\rangle = |\dots -, + \dots\rangle$ the signs of the coefficients of $|n\rangle$ and $|n'\rangle$ in the ground state are opposite. In (8) p is the number of $(+, -)$ interchanges needed to get from one of the Néel states to the state $|n\rangle$. The Néel states are the two ground states of the Ising part of the Hamiltonian, i.e. the states of maximum sublattice magnetization. For the linear chain one of the Néel states can be written as: $|+ - + - \dots\rangle$. This Marshall rule is applicable to non-frustrated lattices (e.g. square, cubic) [7]. For frustrated lattices (e.g. triangular) it leads to contradictions, so there it cannot be used. We minimize the energy by varying the coefficients $a(E_n)$ to get the energy

$$E_{var} = \langle \Psi_{tr} | H | \Psi_{tr} \rangle / \langle \Psi_{tr} | \Psi_{tr} \rangle. \quad (9)$$

We call this the global approximation.

The following approximation we make is the assumption that we may write $a(E_n)$ as

$$a(E_n) = K \exp(-\tfrac{1}{2} \beta E_n), \quad (10)$$

with $H_{zz}|n\rangle = E_n|n\rangle$. The constant K is a normalization constant chosen so that $\langle\Psi_{\text{tr}}|\Psi_{\text{tr}}\rangle = 1$. We use β as a variational parameter to minimize the energy. This parameter β may be interpreted as an inverse temperature ($\beta = 1/k_B T$) as will be explained in section 8. We call this approximation (10) the Boltzmann approximation.

3. Linear chain

For the linear chain we have calculated the exact ground state and ground state energy for 8 and 12 spins with periodic boundary conditions. In fig. 1, we show the logarithmic behaviour of the average coefficients $\bar{a}(E)$. In table I we give the energy $\varepsilon_0 = E_0/NJ$ for the exact solutions and the approximations. From fig. 1 it is obvious that for low Ising energies $\ln \bar{a}(E)$ is rather well approximated by a linear function of E_n . For higher values of E_n the coefficients in the exact ground state deviate a little from this linear function. But the contribution of these states to the ground state is very small as may be seen from fig. 2. In this figure we depicted the cumulative weight function

$$P(E) = \sum_{\substack{n \\ E_n \leq E}} |a(E_n)|^2. \quad (11)$$

For large numbers of spins in the chain an exact numerical solution for the

Fig. 1. The dependence of $\ln \bar{a}(E_n)$ on the Ising energy E_n for the linear chain of 8 and 12 spins for the global (point) and Boltzmann approximation (line).

Table I

	ε_0			β
	exact	variational	Boltzmann	
8 spins	-0.4564	-0.4509	-0.4497	1.986
12 spins	-0.4489	-0.4355	-0.4350	2.160
48 spins		-0.4144	-0.4140	2.390

ground state is impossible. However, it is possible to derive some relations between coefficients of connected states. The Hamiltonian (1) can be written as

$$H = H_{zz} + H_{xy} = H_{zz} + \sum_{i,j} \frac{1}{2} (S_i^+ S_j^- + S_i^- S_j^+). \tag{12}$$

The operator

$$H_{xy} = \sum_{i,j} \frac{1}{2} (S_i^+ S_j^- + S_i^- S_j^+) \tag{13}$$

connects the different Ising states (kets) by means of a transition, i.e. the interchange of two neighbouring antiparallel spins. Suppose we have $2N$ spins in a chain, with N spins up (+) and N spins down (−) and spin $(2N + 1)$ is identified with spin 1 (periodic boundary conditions). The number of parallel neighbours equals $2p$, the number of (+ +) pairs as well as the number of (− −) pairs equals p . The number $2q$ indicates the number of antiparallel

Fig. 2. The fraction $P(E)$ of the states in the ground state with Ising energy smaller than or equal to the value E , for 8 and 12 spins in a linear chain.

neighbouring pairs. We notice that: $2p + 2q = 2N$. The Ising energy of a configuration with given p and q equals

$$E = \sum S_{i,z} S_{i+1,z} = \frac{1}{4}(2p - 2q) = p - \frac{1}{2}N. \quad (14)$$

It can be derived that the number of Ising states corresponding with a given value of p , so with the same Ising energy, is given by

$$\eta(p) = 2 \binom{N}{p} \binom{N-1}{p}. \quad (15)$$

The transitions from one state to another will take place by interchange of two antiparallel neighbouring spins and is caused by H_{xy} (13). With this interchange the Ising energy of the state can change with an amount of $\Delta E = -1, 0$ or $+1$, cf. (14). The total number of transitions from a state with energy E to a state with energy $E + \Delta E$ can be expressed in terms of the number p :

$$\Delta E = 1 \quad (\Delta p = 1) \quad a_+(p) = 2 \times 2N \times \binom{N-2}{p} \binom{N-2}{p}, \quad (16)$$

$$\Delta E = 0 \quad (\Delta p = 0) \quad a_0(p) = 2 \times 2 \times 2N \binom{N-2}{p} \binom{N-2}{p-1}, \quad (17)$$

$$\Delta E = -1 \quad (\Delta p = -1) \quad a_-(p) = 2 \times 2N \binom{N-2}{p-1} \binom{N-2}{p-1}. \quad (18)$$

With the help of these relations and the use of global coefficients, $a(E_n)$, which are the same for all states with a given Ising energy, it is possible to determine variationally the minimum energy of the Heisenberg Hamiltonian.

We made this calculation for 48 spins and table I gives the result. In fig. 3 we show the behaviour of $\ln a(E_n)$ as a function of E_n . In this figure we see that for a large region of E_n , $\ln a(E_n)$ is a linear function of E_n with small deviations for higher E_n . In fig. 4 we see that the states with higher E_n form a rather small part of this state. The variational ground state has an energy per spin that differs from the Hulthén value by about 6.5 percent.

4. The square lattice

For the square lattice we have chosen a system of 4×4 spins with periodic boundary conditions (p.b.c.).

First we solved (2) exactly and found, of course, the same result as Oitmaa

Fig. 3. $\ln \bar{a}(E_n)$ for a linear chain of 48 spins in the global approximation.

and Betts [5] did, i.e. $\varepsilon_0 = -0.7018$. The Marshall rule was applied in the construction of the variational (=global) wave function and in the Boltzmann approximation. The global approximation differs only 1.2% from the exact value of ε_0 whereas the Boltzmann approximation gives a minimal energy that is 1.5% higher. We have drawn the functions $\ln \bar{a}(E_n)$ and $P(E)$ for the exact solutions in figs. 5 and 6, respectively.

Fig. 4. $P(E)$ for a linear chain of 48 spins in the global approximation.

Fig. 5. $\ln \bar{a}(E_n)$ for the square lattice of 4×4 spins in the global approximation.

Suzuki and Miyashita [6] used a correlated trial function for a spin- $\frac{1}{2}$ X - Y magnet and applied this wave function to the isotropic Heisenberg model. Though this wave function was constructed for the X - Y model they found a rather good ground-state energy. However, from our results it can be concluded that there exists an adaptation of their method to the isotropic Heisenberg model.

Fig. 6. $P(E)$ for the square lattice of 4×4 spins in the global approximation.

To get an impression of the accuracy of this method it seems worthwhile to compare the results with other approximations. As an example we can take the RVB (resonating valence bonds) approximation [7], which gives a ground-state energy that is 4.7% above the exact value. (See table II.)

5. The $2 \times 2 \times 2$ cube

We have also studied the $2 \times 2 \times 2$ cube. This cube is not really a 3D system but a unit cell in 3 dimensions. The results for this case look like those for the square lattice, as can be seen in figs. 7 and 8 and in table II.

6. The triangular lattice

The triangular lattice is an example of a fully frustrated lattice, which means that the Marshall rule cannot be applied.

For a set of 12 spins with p.b.c. we solved (2) numerically and found the same energy as that given by Marland and Betts [8], i.e. $\varepsilon_0 = -0.6102$. We calculated the exact ground state too. The phases of the Ising kets in this exact ground state were used for the global and the Boltzmann approximation. In fig. 9 we show the global (points) and the Boltzmann approximation (line). Though we see that the Boltzmann and the global approximation differ, the energies of these approximations are only about 6.7% above the exact value.

Fig. 7. $\ln \bar{a}(E_n)$ for the $2 \times 2 \times 2$ cube in the global approximation.

Fig. 8. $P(E)$ for the $2 \times 2 \times 2$ cube in the global approximation.

Also in this case we see that the Boltzmann approximation is much better than the RVB approximation of Inoue [9], which is 11% above the exact value. To decide whether the Boltzmann approximation is reliable for the triangular lattice or not, we need the results for larger numbers of spins.

Fig. 9. $\ln \bar{a}(E_n)$ for the triangular lattice with 12 spins (p.b.c.) in the global (point) and Boltzmann approximation (line).

Table II

	ϵ			β
	exact	variat.	Boltzmann	
Square lattice, $N = 16$	-0.7018	-0.6931	-0.6911	1.074
$2 \times 2 \times 2$ cube	-0.6025	-0.5999	-0.5982	1.250
Triangular lattice, $N = 12$	-0.6102	-0.5842	-0.5687	2.000

7. Numerical results

In table II we have collected all relevant results for the square lattice, triangular lattice and the cube. From this table we conclude that the approximation of the coefficients in the ground state by means of an exponential function of the Ising energy offers a very good value for the energy (at most 1.5% too high, for the square and the cubic lattice).

8. Discussion

The proposal to approximate the coefficients in the ground state by a function of a few parameters including the Ising energy E_n was already made by Marshall [4] in 1955. We think, however, that the dominant variable is the Ising energy.

We have given two approximations to the exact finite cell calculations. The accuracy of the first approximation can be measured by a quantity Δ_{glob} , which gives the difference between the approximated global value and the exact value for the energy per spin,

$$\Delta_{\text{glob}} = \epsilon_{\text{glob}} - \epsilon_{\text{exact}}. \quad (19)$$

If we calculate $\Delta_{\text{glob}}/\epsilon_{\text{exact}}$ for the linear chain with 12 spins we get 0.029. For the square lattice with 16 spins it has the value 0.012. These numbers suggest that Δ_{glob} becomes smaller when the dimension increases. The limit of maximum dimension is represented by the so-called spherical lattice. This lattice is subdivided into two sublattices A and B, and each spin in sublattice A is connected with every spin in sublattice B. For such a lattice it is easy to show by symmetry arguments that all Ising states with the same Ising energy have the same coefficient in the ground state, so the global approximation becomes exact, hence $\Delta_{\text{glob}} = 0$. Because each spin on the A sublattice is connected with each spin on the B sublattice (e.g. N) we could call the spherical lattice

Fig. 10. $\ln a(E_n)$ for 48 spins in the spherical lattice.

$N/2$ -dimensional. Summarizing one may expect that as the dimension increases, the global approximation improves.

To illustrate the case of the spherical lattice we give in fig. 10 the dependence of $\ln a(E_n)$ on the Ising energy, for a system of 48 spins.

The description of the ground state by a global approximation is further approximated by the Boltzmann approximation. In this case too we measure the accuracy of this approximation by means of the quantity Δ_{Boltz} with

$$\Delta_{\text{Boltz}} = \varepsilon_{\text{Boltz}} - \varepsilon_{\text{glob}}. \quad (20)$$

One gets:

$$\text{linear chain: } \Delta_{\text{Boltz}}/\varepsilon_{\text{glob}} = 0.0011 \text{ (12 spins) ,}$$

$$\text{square lattice: } \Delta_{\text{Boltz}}/\varepsilon_{\text{glob}} = 0.0029 \text{ (16 spins) .}$$

For the spherical lattice the Boltzmann approximation is not applicable as a consequence of large degeneracy of the states with high Ising energy (see fig. 10) and the fact that the difference in energy for adjacent states for large $|E|$ is of the order of the number of spins. For such a spectrum one could not expect a Boltzmann-like distribution. This indicates that with increasing dimension, the accuracy of the Boltzmann approximation will decrease.

The best approximation will be found if both Δ_{glob} and Δ_{Boltz} are small. It seems that the best candidates for this to happen are precisely the most

interesting cases from a physical point of view, i.e. the two- and three-dimensional lattices.

In the Boltzmann approximation we used a parameter β , which can be related to an inverse temperature. To justify this relation we look at the Hamiltonian of an X - Y antiferromagnetic spin- $\frac{1}{2}$ system with a variable Y -interaction. The Hamiltonian of such a system is given by

$$H = H_{xx} + \delta H_{yy} \quad (21)$$

$$\boxed{X} \rightleftharpoons \boxed{Y}$$

$$= \sum (S_{ix}S_{jx} + \delta S_{iy}S_{jy}). \quad (22)$$

If $\delta = 0$, H represents the Ising Hamiltonian and $\delta = 1$ corresponds with the isotropic X - Y Hamiltonian. When the interaction is switched on ($\delta \neq 0$) energy may pass from one subsystem (X) to the other subsystem (Y). The energy is fluctuating between these X and Y parts and so we can speak of two coupled systems. We suppose that the energy of the interaction between the two systems is small compared to the total energy of one system and that the systems are in thermal equilibrium. Further we suppose that we are dealing with a canonical ensemble for each subsystem, and that we can assign statistical weights to the states in phase space with

$$G(|n\rangle) = \alpha \exp[-\beta E(|n\rangle)] \quad \text{with } \beta = 1/k_B T, \quad (23)$$

with k_B the Boltzmann constant and T the temperature of system and bath.

The expectation value of a thermodynamic quantity A in this canonical ensemble is given by

$$\langle A \rangle = \frac{\sum_n A(n) \exp(-\beta E_n)}{\sum_n \exp(-\beta E_n)}. \quad (24)$$

This expression is identical with the expectation value of an operator A in a linear combination of Ising states with the coefficient given by $\exp(-\frac{1}{2}\beta E_n)$ with E_n the Ising (secular) energy of state $|n\rangle$, cf. formula (10).

Lieb, Schultz and Mattis [10] solved the linear antiferromagnetic chain for the X - Y model exactly and calculated the spin-correlation functions for both the Ising model (X - X) and the X - Y model. From their results it can be derived that the X - X spin-correlation in the ground state of the isotropic X - Y model is given by

$$\langle S_{ix}S_{i+1,x} \rangle = -\frac{1}{2\pi}. \quad (25)$$

The X - X spin-correlation for an Ising system that is in thermal equilibrium at temperature T is [13]:

$$\langle S_{ix} S_{i+1,x} \rangle = -\frac{1}{4} \tanh\left(\frac{1}{2} \beta\right). \quad (26)$$

The inverse Ising temperature of the ground state in the X - Y model for the linear chain can now be defined as that β_0 for which formulas (25) and (26) are equal; $\beta_0 = 3.0098$ for this case.

In the same way we can divide the Heisenberg Hamiltonian in two parts, ZZ and X - Y , and consider the ZZ reservoir in thermal equilibrium with the X - Y bath. We can, in that case also attribute an Ising temperature to the whole system bath + reservoir, which is supposed to be in thermal equilibrium.

We have derived (see ref. [11]) a relation between the dimension d of the lattice, the energy per spin $\varepsilon_0(d, N)$ in the ground state and the inverse temperature β^* . In this derivation we make use of the property of regular lattices, such as the chain, square and cubic lattice, that the coefficient a_1 of the states that emerge from the Néel states by one $(+, -)$ interchange, are the same. If one has $2N$ spins in a d -dimensional bipartite lattice the relation between the coefficient of the Néel states, $a_{N\dot{e}}$ and the coefficients a_1 of the above-mentioned states can be written as

$$\frac{a_{N\dot{e}}}{a_1} = \frac{d}{-\frac{1}{2}d - 2\varepsilon_0(d, N)}, \quad (27)$$

which follows from the eigenvalue problem for the ground state. If the Boltzmann approximation is used, then this same ratio reads

$$\frac{a_{N\dot{e}}}{a_1} = \exp\left[+\frac{1}{2}\beta(2d - 1)\right]. \quad (28)$$

In this formula $(2d - 1)$ is the difference between the Ising energy of the Néel state and the energy of a connected state (a state made out of the Néel state by an $(+ -)$ interchange) in a d -dimensional lattice. This leads to the conclusion that β^* is given by

$$\beta^* = 2\{\ln d - \ln[-\frac{1}{2}d - 2\varepsilon_0(d, N)]\} / (2d - 1). \quad (29)$$

This β^* differs a little from the β derived with the variational method because in the derivation of β^* , only two coefficients of the ground state (i.e. $a_{N\dot{e}}$, a_1) appear while in the calculation of β we used all coefficients to minimize the energy.

For the triangular lattice such a relation is not easy to derive because not all coefficients a_1 have the same value.

Table III

Dim:	1	2	3	Triangular	2
β^*	1.904	1.177	0.932 ^{a)}	β_{Heis}	2.000
$\beta_c^{(\text{Ising})}$	∞	1.761	0.887 ^{b)}	$\beta_c^{(\text{Ising})}$	∞

For $d = 2$ we used the Kramers–Wannier results [14].

^{a)} See P.L. Iske [11]. ^{b)} See ref. [12].

In table III we compare the temperatures β^* with the known critical inverse temperatures for the Ising systems that correspond with it, which concerns the linear chain, square and the cube. For the three-dimensional case we see that $\beta^* > \beta_c$ (Ising); this means that the corresponding T^* is lower than the critical temperature in the Ising model. We think that this is indicative of ordering in three-dimensional Heisenberg cubic lattice.

9. Conclusion

From the exact numerical solutions of finite size systems we can conclude that the Boltzmann approximation of the ground state gives a good value for the energy, especially in the two-dimensional case. For the three-dimensional case there is only a weak indication that this conclusion is valid. We expect an increasing accuracy for large numbers of spins. The variational parameter β in the Boltzmann approximation can be connected with an inverse temperature.

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