Approximate ground states of some finite clusters having Heisenberg antiferromagnetic exchange interactions

1990 J. Phys.: Condens. Matter 2 10085

(http://iopscience.iop.org/0953-8984/2/50/013)

J. Phys.: Condens. Matter **2** (1990) 10085–10090. Printed in the UK

Approximate ground states of some finite clusters having Heisenberg antiferromagnetic exchange interactions

K W H Stevens
Department of Physics, University of Nottingham, Nottingham NG7 2RD, UK

Received 1 March 1990

Abstract. The purpose of this paper is to attempt to determine how much of the Ising ground state there is in the ground state of an Heisenberg antiferromagnetic. Square planar clusters of 5, 13 and 25 spins of $\frac{1}{2}$ are considered, using approximate ground states chosen as a result of symmetry arguments. Various physical quantities, including bond energies which are more negative than those found with the corresponding Ising states, then become relatively easy to evaluate. We conclude that the chosen states are better approximations than the Ising states. It seems that as the number of spins increases the probability of finding the Ising state tends to zero, while remaining the most probable state, in a site representation.

1. Introduction

The discovery of high temperature super-conductivity in a range of copper oxides has created interest in all copper compounds which contain planar arrangements of copper and oxygen ions, whether or not they are super-conducting. Among these are compounds which are insulating and regarded as containing square planar arrangements of Cu^{2+} ions, each with spin $\frac{1}{2}$, with O^{2-} ions between the nearest neighbour Cu^{2+} ions. It is generally assumed that nearest neighbour Cu^{2+} ions are coupled by antiferromagnetic exchange interactions of the Heisenberg type. The understanding of the nature of the ground and excited states of such a system is then a special class of a widely studied problem: that of the nature of the states of a general antiferromagnetic. This problem is an extremely challenging one, and there is extensive literature on it. (For recent references see [1].)

The purpose of this paper is to describe a conceptually simple idea which may be of interest because it seems to throw light on the nature of the ground states of some finite clusters of spins, without the considerable effort that is required to obtain exact solutions. The investigation began in an attempt to answer a question which arises from the usual model of an antiferromagnet: that in which there are two interpenetrating ferromagnetic sub-lattices. With an Ising type of interaction the ground state has one sub-lattice with all spins having $m_s = \frac{1}{2}$ and another having $m_s = -\frac{1}{2}$, and the interest is in the amplitude of this state in the ground state of the Heisenberg system. To obtain a feeling for the answer it seemed to be of interest to study a small system using the algebraic form of the Lanczos method [2]. The first choice is shown in figure 1—a structure consisting of five

0953-8984/90/5010085 + 06 $03.50

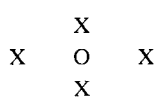

Figure 1. 5 sites, with X denoting up and O denoting down spins.

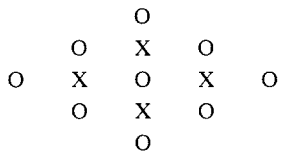

Figure 2. 13 spins, with X denoting up and O denoting down spins.

spins—the problem being to find the ground state and determine the amplitude of the state:

$$|-\rangle|++++\rangle. \tag{1}$$

The first ket represents the m_s of the site at the centre of the cluster (site 0, which will also be labelled as shell A) and the second ket gives the m_s values of the outer four sites (labelled sequentially from 1 to 4 and described as being in shell B). By the method one begins with the above state and applies the Hamiltonian to it. This generates a state that contains the initial state and a remainder, which is used to define a normalized state orthogonal to the initial one. The Hamiltonian is then applied to this new state and the remainder, which is orthogonal to both previous states, is used to define a third orthonormal state, and so on. When the matrix of the Hamiltonian is set up in the basis of the orthonormal states so generated it is tri-diagonal. In the present application it is not necessary to go beyond the first step, for only two orthonormal states are generated. The matrix of the Hamiltonian is 2×2 and is easily diagonalized, giving, as an eigenstate, the low-lying state

$$(\tfrac{4}{5})^{1/2}[|-\rangle|++++\rangle - \tfrac{1}{4}(|+\rangle|-+++\rangle + |+\rangle|+-++\rangle + |+\rangle|++-+\rangle + |+\rangle|+++-\rangle)]. \tag{2}$$

By Kramers' theorem this is degenerate with the state in which all spins are reversed. Also, the mean energy of the interaction of the single spin in the A shell with any spin in the B shell is $-\frac{3}{8}$ in units of J, the exchange interaction which is taken to be positive.

At this point it is of interest to pause and ask what the analysis has achieved, for it is obvious that, since the Hamiltonian commutes with the total spin, the Hamiltonian eigenstates are eigenstates of total spin. Thus, beginning with a state which has $M_s = \frac{3}{2}$, the method has generated an eigenstate of total spin. But this could have been done in quite a different way, for the state of the central site (0) is a state belonging to a total spin of $\frac{1}{2}$, while the state $|++++\rangle$ of the B shell is an eigenstate with $S = 2$. But since $D_{1/2} \otimes D_2 = D_{3/2} + D_{5/2}$ it is clear that the ground state either belongs to total spin $\frac{3}{2}$ or $\frac{5}{2}$, and with an antiferromagnetic interaction it is not surprising that the ground level has the lower of the two values, which is what is found. This state could have been written down directly by using the appropriate coupling coefficients for angular momentum operators (which are known variously as Clebsch–Gordan, or Wigner coefficients or as 3-j symbols, see [3]). In fact the state given above is equivalent to

$$(\tfrac{4}{5})^{1/2}(|-\tfrac{1}{2}, 2\rangle - \tfrac{1}{2}|\tfrac{1}{2}, 1\rangle) \qquad (\tfrac{1}{2}, 2). \tag{3}$$

The first number in each ket gives the M_s value for spin $\frac{1}{2}$ (the A shell) and the second the M_s for the spin 2 of the B shell. The $(\frac{1}{2}, 2)$ which follows the ket gives the total spin

values of the two shells in sequence. Two comments can now be made. The first is that the new form is rather simpler to write down than the first, for the string of m_s kets (a 'site representation') is not required. Secondly, the evaluation of expectation values of the whole or parts of the Hamiltonian can be obtained using angular momentum properties, because any site operator can be replaced by an equivalent angular momentum operator acting in a suitable spin space. It is not necessary to give a demonstration at this point, as the technique will be used frequently for the larger clusters to be considered in the following.

2. A cluster of 13 spins

The next cluster of interest is one in which spins in the B shell are coupled to those nearest neighbours which have not been included previously. Such a cluster is shown in figure 2. It contains 13 spins: one in the A shell; 4 in the B shell and 8 in the C shell, with those in the C shell being numbered consecutively from 6 to 13. It is convenient to suppose that the exchange interactions between the B and C shells can be gradually turned on by varying a parameter from zero to unity. When it is zero a ground state of the Hamiltonian is given by an extension of (3)

$$(\tfrac{4}{5})^{1/2}(|-\tfrac{1}{2}, 2\rangle - \tfrac{1}{2}|\tfrac{1}{2}, 1\rangle)|--------\rangle \tag{4}$$

where the ket $|--------\rangle$ is the state in which all the spins in the C shell have $m_s = -\frac{1}{2}$. The state (4) contains the Ising ground state with quite a large amplitude because the part

$$(\tfrac{4}{5})^{1/2}(|-\tfrac{1}{2}, 2\rangle - \tfrac{1}{2}|\tfrac{1}{2}, 1\rangle) \tag{5}$$

has been constructed from an initial state which has the A site with $m_s = -\frac{1}{2}$ and all B sites with $m_s = \frac{1}{2}$. The application of successive powers of the Hamiltonian when the parameter is non-zero (the Lanczos technique) is quite tedious, and it may be thought to be unnecessarily so, because one knows, *a priori*, that the eigenstate is a state of total spin. It would seem sensible, therefore, to avoid beginning with the above state and instead use one which is already an eigenstate of total spin. Since the first part of the state is already an eigenstate of total spin, with $S = \frac{3}{2}$, and the part $|--------\rangle$ is an eigenstate of $S = 4$, with $M_s = -4$, there is the question of which total spin to choose. Now $D_{3/2} \otimes D_4 = D_{5/2} + D_{7/2} + D_{9/2} + D_{11/2}$, so it seems likely, for an antiferromagnetic, that the manifold with the smallest total spin will lie lowest. It therefore seems reasonable to choose, as initial state for the Lanczos method, a state constructed from the $S = \frac{3}{2}$ of the first two shells and an $S = 4$ for the C shell which belongs to an $S = \frac{5}{2}$ and has $M = -\frac{5}{2}$. This is readily done using Clebsch–Gordan coefficients or by using the relation

$$\begin{aligned} 2\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 &= (\boldsymbol{S}_1 + \boldsymbol{S}_2) \cdot (\boldsymbol{S}_1 + \boldsymbol{S}_2) - \boldsymbol{S}_1 \cdot \boldsymbol{S}_1 - \boldsymbol{S}_2 \cdot \boldsymbol{S}_2 \\ &= 2S_1^z S_2^z - S_1^- S_2^+ - S_1^+ S_2^- \end{aligned} \tag{6}$$

and setting

$$|\tfrac{5}{2}, -\tfrac{5}{2}\rangle = a|\tfrac{3}{2}, -4\rangle + b|\tfrac{1}{2}, -3\rangle + c|-\tfrac{1}{2}, -2\rangle + d|-\tfrac{3}{2}, -1\rangle \qquad (\tfrac{3}{2}, 4) \tag{7}$$

where on the right side the numbers in the kets denote M values and the $(\frac{3}{2}, 4)$ denote the associated total spin values in the same order. When (6) is applied to the left side of (7) it produces a multiple of the right side, which can be equated, by the direct application

of (6), to the right side. This produces simple relations between the coefficients a, b etc. It is found that

$$|\tfrac{5}{2}, -\tfrac{5}{2}\rangle = (\tfrac{3}{2})^{1/2}(|\tfrac{3}{2}, -4\rangle - (3/\sqrt{24})|\tfrac{1}{2}, -3\rangle + (3/\sqrt{84})|-\tfrac{1}{2}, -2\rangle$$
$$- (1/\sqrt{56})|-\tfrac{3}{2}, -1\rangle) \quad (\tfrac{3}{2}, 4). \tag{8}$$

If (8) has been expressed in a site representation it would have contained many kets and the application of powers of the Hamiltonian, as is required in the Lanczos method, would have produced even more complicated expressions. Rather than embark on this, it is of interest to ask how good an approximation this state alone is to the ground state? An indication can be obtained by comparing the expectation value of the Hamiltonian in this state with that for the corresponding Ising state.

To demonstrate how this is done it is convenient to begin with s^z_C and evaluate its expectation value for the state (8), using the right hand side of the equation. The second M value in each ket refers to a manifold of states constructed solely from the C shell, and within this manifold s^z_C is equivalent to $\frac{1}{8}S^z_C$. It follows that

$$\langle \tfrac{5}{2}, -\tfrac{5}{2}|s^z_C|\tfrac{5}{2}, -\tfrac{5}{2}\rangle = \tfrac{2}{3}\cdot\tfrac{1}{8}(-4 + \tfrac{9}{24}(-3) + \tfrac{9}{84}(-2) + \tfrac{1}{56}(-1)) = -25/56 \tag{9}$$

The mean value of the z-component of a spin in the C shell is thus of smaller magnitude than would be found for the Ising state. (All other components have zero expectation values.) The evaluation of the expectation value for s^z_B is slightly more complicated. Within the $S = 4$ part of (8) it is equivalent to unity. Within the $S = \frac{3}{2}$ part it can be written as λS_z. To determine λ, use is made of (2). Then

$$\tfrac{3}{2}\lambda = \tfrac{4}{5}(\tfrac{1}{2} + \tfrac{1}{16}) = \tfrac{3}{10}$$

Thus

$$\langle|s^z_B|\rangle = \tfrac{3}{10}\cdot\tfrac{2}{3}[\tfrac{3}{2} + \tfrac{9}{24}(-3) + \tfrac{9}{84}(-2) + \tfrac{1}{56}(-1)] \tag{10}$$
$$= \tfrac{9}{28} \tag{11}$$
$$\langle|s^z_C s^z_B|\rangle = \tfrac{1}{8}\cdot\tfrac{3}{10}\langle|s^z_1 s^z_2|\rangle \tag{12}$$

where S_1 acts in the $S = \frac{3}{2}$ manifold of (8) and S_2 in the $S = 4$ manifold. Thus

$$\langle|s^z_B s^z_C|\rangle = \tfrac{1}{8}\,\tfrac{3}{10}[\tfrac{3}{2}(-4) + \tfrac{9}{24}(\tfrac{1}{2})(-3) + \tfrac{9}{84}(-\tfrac{1}{2})(-2) + \tfrac{1}{56}(-\tfrac{3}{2})(-1)]$$
$$= -\tfrac{9}{56} \tag{13}$$

The Ising value would be $-\frac{1}{4}$, so there is a substantial difference. However, in the Heisenberg case $\frac{1}{2}(s^+_B s^-_C + s^-_B s^+_C)$ must also be included. The same equivalence factors can be used, so:

$$\langle|\tfrac{1}{2}(s^+_B s^-_C + s^-_B s^+_C)|\rangle = \tfrac{1}{8}\,\tfrac{3}{10}\langle|S^+_1 S^-_2|\rangle \tag{14}$$

with $|\ \rangle$ written as the righthand side of (8) and with $S_1 = \frac{3}{2}$ and $S = 4$. It follows that

$$\langle|s^+_C s^-_B|\rangle = \tfrac{1}{8}\,\tfrac{3}{10}\,\tfrac{2}{3}(-3\sqrt{3.8}/\sqrt{24} - 9\sqrt{4.14}/\sqrt{24.84} - 3\sqrt{3.18}/\sqrt{84.56})$$

or

$$\langle|s_C\cdot s_B|\rangle = -\tfrac{9}{32} \tag{15}$$

which has larger magnitude than that for the Ising case. Using similar reasoning the mean values of s^z_A, $s^z_A s^z_B$ and $s_A\cdot s_B$ are respectively $-\frac{3}{14}$, $\frac{9}{35}$ and $-\frac{3}{8}$. Every bond gives

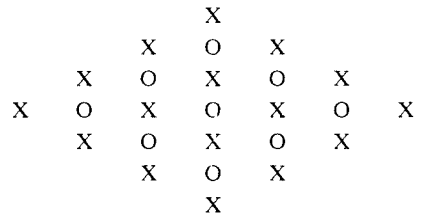

Figure 3. 25 spins, with X denoting up and O denoting down spins.

a lower energy than would be found for the Ising state and so suggests that (8) is a better choice for either the Lanczos or a variational method.

The expectation value of $s_A \cdot s_B$ was found to be identical in the 13 site and in the 5 models. This could have been expected, for $s_A \cdot s_B$ is an invariant, and so could have been replaced by an equivalent constant within a manifold of total spin. In using (8) $s_A \cdot s_B$ is equivalent to unity in any $S = 4$ parts, and so it is as if it acts solely in the $S = \frac{3}{2}$ manifold, which is just the case considered in the 5 site model.

3. 25 sites

The method is only slightly more complicated when extended to the 25 sites shown in figure 3, a problem which is outside the range of the present finite cluster calculations. In this case the state with $S = \frac{5}{2}$ for the A, B and C shells is coupled to the $S = 6$ of a D shell, yielding an overall $S = \frac{7}{2}$. The state with $M = \frac{7}{2}$ is

$$(\tfrac{8}{13})^{1/2}[|-\tfrac{5}{2}, 6\rangle - (5/\sqrt{60})|-\tfrac{3}{2}, 5\rangle + (5/\sqrt{11.15})|-\tfrac{1}{2}, 4\rangle - 1/\sqrt{22}|\tfrac{1}{2}, 3\rangle$$
$$+ (4/3\sqrt{22.8})|\tfrac{3}{2}, 2\rangle - (1/6\sqrt{22})|\tfrac{5}{2}, 1\rangle] \qquad (\tfrac{5}{2}, 6) \tag{16}$$

and with the states already given in (3) and (8) and a knowledge of the matrix elements of S within a variety of spin manifolds [4] it is straightforward to calculate any required expectation values. Thus $\langle|s_A^z|\rangle = -\frac{1}{6}$, $\langle|s_B^z|\rangle = \frac{1}{4}$, $\langle|s_C^z|\rangle = -\frac{25}{72}$ and $\langle|s_D^z|\rangle = \frac{49}{108}$. A check is provided because

$$\langle|s_A^z + 4s_B^z + 8s_C^z + 12s_D^z|\rangle = \left\langle\left|\sum s_z\right|\right\rangle = \tfrac{7}{2} \tag{17}$$

The mean values of $s_A \cdot s_B$ and $s_B \cdot s_C$ are exactly the same as the values for the 13 site model, and $s_C \cdot s_D$ is $-\frac{25}{96}$.

4. Discussion and conclusion

The original purpose of this work was to try to discover how much of the Ising state there is in the ground state of a Heisenberg system by studying a finite cluster. Whether or not this goal has been achieved is not entirely clear, for the determinations of actual ground states present formidable problems. What has been done is that we have obtained states which are probably better approximations to the actual ground states than the Ising states, for they certainly produce lower expectation values for the Hamiltonians. If one is prepared to ask the same questions of these states as one would like to ask about the true ground states, a number of interesting conclusions can be reached. For the 5 site

problem and the approximate ground state the probability that the system will be in the Ising state is $\frac{4}{5}$, which is the square of the coefficient of $|-\rangle|++++\rangle$ in (2), or of $|-\frac{1}{2}, 2\rangle$ in (3); for the 13 site problem it is $\frac{8}{15}$ (obtained from the square of the product of the normalizing factors in (4) and (8)); and for the 25 site problem it is $\frac{64}{195}$. It is therefore quite clear that as the clusters become larger the probability of finding the Ising state decreases, tending to zero as the number of spins increases. Apart from equations (1) and (2) none of the states which have been used have been written in a site representation. In principle this would be possible, but in practice they would be very lengthy, for with 25 sites, for instance, there are a vast number of such states which could be expected to be present. Since the bulk of the probability has already been absorbed in the Ising state it is to be expected that each such site state will appear with an extremely small amplitude. So it can be expected that although the Ising amplitude tends to zero as the number of spins increases, it is always going to be more probable than any other site state. But this is not to say that the other site states can be ignored because even in the 13 site problem the major contributions to the mean values of $\boldsymbol{s}_A \cdot \boldsymbol{s}_B$ and $\boldsymbol{s}_B \cdot \boldsymbol{s}_C$ come from the spin-flip terms in the Hamiltonian. The inference is that the multitude of states with very small amplitudes have correlated phases and amplitudes, such that their neglect would be disastrous to the determination of the bond energies. Indeed it seems likely that if anything is to be neglected it should be the Ising-like part of the Hamiltonian. It seems, also, that another requirement is met by these relatively small terms. Throughout the analysis it has not been necessary to specify the axis of quantization, and indeed it is arbitrary. But how can it be arbitrary if only the Ising parts of the states are retained? The rotational symmetry associated with the total spin would be lost, and one can only assume that the states with the relatively small amplitudes are necessary to ensure the rotational symmetry.

Finally there is the question of a cooperative transition into the antiferromagnetic phase. It is often assumed that below the transition temperature the system is not in an eigenstate of the Hamiltonian being used because in practice there are other perturbations, of lower symmetry, present which have been omitted and which determine alignments. Indeed in some studies such perturbations are deliberately added and then allowed to tend to zero towards the end of the theory. In the present case it would seem that such a procedure might leave the system in an aligned state, but one which is still an eigenstate of the Heisenberg Hamiltonian!

References

[1] Gross M, Sanchez-Velasev E and Siggia E D 1989 *Phys. Rev.* B **40** 11328
Lui Z and Manousakis E 1989 *Phys. Rev.* B **40** 11437
Neuberger H and Ziman T 1989 *Phys. Rev.* B **39** 2608
Reger J D, Riera J A and Young A P 1989 *J. Phys.: Condens. Matter* **1** 1855
[2] Fletcher J R 1980 *J. Phys. A: Math. Gen.* **13** 2029
Lanczos C 1950 *J. Res. NBS* **45** 255
[3] Judd B R 1963 *Operator Techniques in Atomic Spectroscopy* (New York: McGraw-Hill) p 10
[4] Edmonds A R 1957 *Angular Momentum in Quantum Mechanics* (Princeton, NJ: Princeton University Press)