IOPscience iopscience.iop.org

Home Search Collections Journals About Contact us My IOPscience

A microscopic approach to the dimerization in frustrated spin- 1/2 antiferromagnetic chains

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1994 J. Phys.: Condens. Matter 6 5965

(http://iopscience.iop.org/0953-8984/6/30/015)

View the table of contents for this issue, or go to the journal homepage for more

Download details:
IP Address: 171.66.16.147
The article was downloaded on 12/05/2010 at 19:02

Please note that terms and conditions apply.

J. Phys.: Condens. Matter 6 (1994) 5965–5980. Printed in the UK

A microscopic approach to the dimerization in frustrated spin-$\frac{1}{2}$ antiferromagnetic chains

Y Xian

Department of Mathematics, UMIST (University of Manchester Institute of Science and Technology), PO Box 88, Manchester M60 1QD, UK

Received 20 April 1994

Abstract. The spontaneous dimerization of frustrated spin-$\frac{1}{2}$ antiferromagnetic chains is studied via a microscopic approach based on a proper set of composite operators (i.e., pseudo-spin operators). Two approximation schemes are developed. Firstly, a spin-wave approximation is made via a Dyson–Maléev-like boson transformation. The ground-state properties and the triplet excitation spectra are obtained as functions of the coupling parameter. Secondly, a microscopic treatment, based on pseudo-spin operators, is formulated within the framework of the powerful, systematic coupled-cluster method (CCM). Comparison between various approximations is made. The advantage of the CCM for the purposes of systematic improvement is emphasized.

1. Introduction

Spontaneous dimerization of theoretical spin–lattice models was perhaps first discovered by Majumdar and Ghosh [1] in 1969. They found that for the one-dimensional (1D) spin-$\frac{1}{2}$ system with nearest-neighbour and next-nearest-neighbour couplings, the perfect dimer state, in which every two adjacent atoms form a spin-singlet valence bond, is the exact ground state of the Hamiltonian at a particular coupling. Obviously, the translational symmetry is spontaneously broken in the dimerized system, and the corresponding ground state of the 1D model is doubly degenerate.

A spin-1 chain, or other higher-order spin chains, can also exhibit dimerized valence-bond structures in their ground states [2]. Very recently, a series of 1D SU(n) spin-s ($n = 2s + 1$) antiferromagnetic model problems have been solved using a Bethe *ansatz* [3], and the corresponding dimerization order parameters have been exactly calculated by the author [4]. In addition to the dimerized spin chains, it is possible that some 1D spin systems (with integer spin quantum numbers) favour the trimer configuration which is produced by a sequence of spin-singlet states formed from every three adjacent spins. Furthermore, dimerization or trimerization may also occur in two or more spatial dimensions. Clearly, a dimerized or trimerized spin system can be viewed as a kind of solid in which the corresponding simple valence bonds are localized and the translational symmetry is broken. The perfect dimer or trimer state is not in general the exact ground state of a given Hamiltonian, but for some systems the ground state may still possess a non-zero solid-like dimer or trimer long-range order and hence show the characteristics of a quantum solid. (It therefore seems more appropriate to refer to these solid-like systems collectively as 'valence-bond lattices' [5].) Due to quantum correlations, one expects that the corresponding long-range order of those quantum solids will be reduced or vanish at certain coupling strengths. A good example is provided by the ground state of the spin-1 SU(3) antiferromagnetic chain [4], where the dimerization order parameter is reduced to 42%.

0953-8984/94/305965+16$19.50 © 1994 IOP Publishing Ltd

One is quite familiar with phonons in ordinary atomic solids and magnons in ferromagnets or antiferromagnets. In particular, the spin-wave theory of Anderson [6] provides a simple and excellent description of the spin correlations in the ground and low-lying excited states for a number of antiferromagnets. Similar approximations have also been developed for some dimerized systems. Parkinson [7] formulated a spin-wave approximation based on a spin-$\frac{1}{2}$ dimer state. He focused on the spin-$\frac{1}{2}$ Heisenberg model and employed the method of equations of motion. A triplet excitation spectrum of $\sqrt{2}\sin k$ was obtained. This compares well with the exact result of a triplet spectrum of $(\pi/2)\sin k$. Chubukov [8] later provided a similar spin-wave theory by using a transformation similar to that of Holstein and Primakoff to study specifically the dimerization of the spin-1 chains with Heisenberg and biquadratic exchanges. Read and Sachdev [9] investigated the dimerization problem within the framework of Schwinger boson field theory. Very recently, the author [10] has extended Parkinson's theory to discuss possible trimerization of a 1D isotropic spin-1 system. The concept of dimerization has also been extended to 2D spin systems, e.g., the J_1–J_2 Heisenberg model on the square lattice [11].

In this article, I intend to investigate the dimerization of spin systems via a systematic, microscopic approach. Because of their simplicity, I focus on the 1D spin-$\frac{1}{2}$ Heisenberg chains with nearest-neighbour and next-nearest-neighbour couplings, for which the dimer state is the ground state at a particular coupling [1]. Following Parkinson [7], I study the dimerization in terms of a proper set of composite operators (pseudo-spin operators). I extend and reformulate Parkinson's theory so that the ground-state properties as well as the excitations can be investigated. Firstly, by employing a Dyson–Maléev-like boson transformation [12] for the pseudo-spin operators, I develop a spin-wave theory. The advantage of using Dyson–Maléev transformation lies in the fact that Hamiltonian can be expressed in a compact form as a finite-order polynomial of boson operators, rather than as an infinite series which is the case when the Holstein–Primakoff-like transformation is used (see, e.g., [8]). Secondly, I apply the powerful, systematic, microscopic many-body theory of the coupled-cluster method (CCM) [13] based on the pseudo-spin operators of the frustrated spin-$\frac{1}{2}$ chain. A systematic approximation scheme within the CCM approach is developed for the ground state. The CCM has recently been successfully applied to the various spin systems with Ising-like long-range order [14] or with planar long-range order [15]. The excellent results produced by the CCM approximations, particularly for the spin systems with Ising-like long-range order, provide the main motivation for the current CCM approach to the dimerization problems.

It should be emphasized that the dimerized or trimerized states are not merely mathematical artifacts. In fact, the 1D frustrated spin-$\frac{1}{2}$ system has recently been shown to be relevant to the reconstruction of FCC metal surfaces at finite temperature, with the spin dimerized phase corresponding to the disordered flat surfaces [16].

The plan of this paper is as follows. In section 2 the composite operators in a matrix representation are introduced and the corresponding boson transformations are given. Section 3 is devoted to the spin-wave approximation for the 1D spin-$\frac{1}{2}$ model. The ground-state energy and excitation spectra are obtained as functions of the coupling constants. In section 4 I describe in detail the microscopic CCM for the ground state of the dimerized spin system. The results from the CCM approximation are compared with the spin-wave theory. I conclude this article with a general discussion in section 5.

2. Pseudo-spin operators and their bosonizations

I first consider a two-spin system, each component being a spin $\frac{1}{2}$. For completeness, some

of the analysis given by Parkinson [7] is repeated here. Clearly, there are four states for such a two-spin system. If $|\uparrow\rangle$ and $|\downarrow\rangle$ are used to represent spin-up and spin-down states respectively, the singlet state can be written as

$$|0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) \tag{2.1}$$

and the triplet states with s^z_{total} $(\equiv s^z_1 + s^z_2)$ $= 1, 0, -1$ are given by respectively

$$|1\rangle = |\uparrow\uparrow\rangle \qquad |2\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle) \qquad |3\rangle = |\downarrow\downarrow\rangle. \tag{2.2}$$

Following Parkinson [7], I employ a matrix representation. Each of the four states of equations (2.1) and (2.2) is then represented by a column matrix with a single non-zero element. One can then introduce operators A_{ij} as having only a single non-zero element of a (4×4) matrix, namely $\langle i'|A_{ij}|j'\rangle = \delta_{ii'}\delta_{jj'}$. For example, A_{10} (A_{30}) is an operator which increases (decreases) s^z_{total} by one unit, while A_{20} leaves it unchanged. Their Hermitian conjugates (i.e., transpose matrices) have the opposite effects. Together with other operators, these sixteen operators form a complete set for the spin pair and any operator of the pair can be written as a linear combination of these sixteen.

For a pair of spins, of which has spin greater than one half, similar operators can be defined. For example, there are nine states for a two-spin system, each component having spin 1, and hence there are 81 (9×9) A_{ij} operators which form a complete set for a pair of spin-1 atoms. For a three-spin system, each component having spin 1, the dimensionality of the matrix is 27 [10].

It is worth pointing out that these A_{ij} operators are *non-linear* in terms of the original single-spin operators; for example,

$$A_{00} = \frac{1}{4} - s_1 \cdot s_2 \qquad A_{01} = -\frac{1}{\sqrt{2}}(s^-_1 - s^-_2)(s^z_1 + s^z_2). \tag{2.3}$$

It is in this sense that I have referred to these A_{ij} as *composite operators* [10]. (Notice that A_{00} is the usual spin-singlet projection operator.) Furthermore, it is easy to prove that they obey the following pseudo-spin algebra:

$$\left[A_{ij},\ A_{kl}\right] = A_{il}\delta_{jk} - A_{kj}\delta_{li}. \tag{2.4}$$

Therefore, I also refer to them as *pseudo-spin operators*. My assumption in this paper is that it is more natural to study the dimerization in terms of these composite operators rather than the original single-spin operators. The spin-wave theory in section 3 and the CCM approximations in section 4 are developed on the basis of this assumption.

In a straightforward manner, one can express the single-spin operators in terms of A_{ij} operators. They are given by [7]

$$s^z_1 = \frac{1}{2}(A_{02} + A_{20} + A_{11} - A_{33}) \qquad s^z_2 = \frac{1}{2}(-A_{02} - A_{20} + A_{11} - A_{33}) \tag{2.5a}$$

$$s^-_1 = \frac{1}{\sqrt{2}}(A_{30} - A_{01} + A_{21} + A_{32}) \qquad s^-_2 = \frac{1}{\sqrt{2}}(A_{01} - A_{30} + A_{21} + A_{32}) \tag{2.5b}$$

$$s^+_1 = \frac{1}{\sqrt{2}}(A_{03} - A_{10} + A_{12} + A_{23}) \qquad s^+_2 = \frac{1}{\sqrt{2}}(A_{10} - A_{03} + A_{12} + A_{23}). \tag{2.5c}$$

Recognizing that A_{ij} obeys the pseudo-spin algebra of (2.4), one can make the following Dyson–Maléev transformation [12]:

$$\begin{aligned} A_{00} &= 1 - a_1^+ a_1 - a_2^+ a_2 - a_3^+ a_3 \\ A_{p0} &= a_p^+ A_{00} \qquad A_{0p} = a_p \\ A_{pp} &= a_p^+ a_p \qquad A_{pq} = a_p^+ a_q \end{aligned} \tag{2.6}$$

where $p, q = 1, 2, 3$, and a_p, a_p^+ are three sets of boson operators, obeying the usual boson commutation

$$[a_p,\ a_p^+] = 1 \tag{2.7}$$

and with all other commutators yielding zero.

By definition the singlet state $|0\rangle$ of (2.1) is the vacuum state of the bosons, namely

$$a_p|0\rangle = 0 \qquad p = 1, 2, 3. \tag{2.8}$$

The physical states correspond to the vacuum state $|0\rangle$ and the three states with only one boson excited. Furthermore, as the matrix elements between physical and unphysical subspaces are equal to zero, the transformation given by (2.6) is exact at zero temperature just as in the case of the conventional spin-wave theory [6].

A general spin-$\frac{1}{2}$ Hamiltonian can be expressed in terms of A_{ij} operators according to (2.5) and then in terms of those three sets of boson operators according to (2.6). In section 3 I consider a spin-wave theory for the frustrated 1D model using these pseudo-spin operators and their bosonizations. In section 4 I develop a microscopic formalism within the framework of the CCM also based on these pseudo-spin operators.

3. Spin-wave theory

The 1D spin-$\frac{1}{2}$ isotropic model with nearest-neighbour and next-nearest-neighbour couplings is described by the Hamiltonian

$$H = \sum_{l=1}^{N} (s_l \cdot s_{l+1} + J s_l \cdot s_{l+2}) \tag{3.1}$$

where J is the coupling constant and N is the total number of spins. I use the periodic boundary condition and choose even N for convenience. I have also taken the lattice spacing to be unity. At $J = \frac{1}{2}$, the Hamiltonian of (3.1) becomes the well known Majumdar–Ghosh model [1], which has two degenerate dimer ground states, with one of them given by

$$|D\rangle = \prod_{r=1}^{N/2} |0\rangle_{2r-1,2r} \tag{3.2}$$

where the notation $|0\rangle_{i,j}$ represents the singlet-paired state of (2.1). This dimer state $|D\rangle$ is shown graphically in figure 1. After choosing the dimer indices as shown in figure 1, the Hamiltonian can be written as

$$\begin{aligned} H = \sum_{r=1}^{N/2} \big[& s_1(r) \cdot s_2(r) + s_2(r) \cdot s_1(r+1) + J s_1(r) \cdot s_1(r+1) \\ & + J s_2(r) \cdot s_2(r+1) \big]. \end{aligned} \tag{3.3}$$

One can then express H in terms of the composite operators A_{ij} by (2.5) as

$$H = \sum_{r=1}^{N/2} (H_r + \tfrac{1}{4} H_{r,r+1} + \tfrac{1}{2} J H'_{r,r+1}) \tag{3.4}$$

with

$$H_r \equiv \frac{1}{4} - A^r_{00} \tag{3.5a}$$

$$\begin{aligned} H_{r,r+1} \equiv {} & (-A^r_{02} + A^r_{11} - A^r_{20} - A^r_{33})(A^{r+1}_{02} + A^{r+1}_{11} + A^{r+1}_{20} - A^{r+1}_{33}) \\ & + (A^r_{01} + A^r_{21} - A^r_{30} + A^r_{32})(A^{r+1}_{03} - A^{r+1}_{10} + A^{r+1}_{12} + A^{r+1}_{23}) \\ & + (-A^r_{03} + A^r_{10} + A^r_{12} + A^r_{23})(-A^{r+1}_{01} + A^{r+1}_{21} + A^{r+1}_{30} + A^{r+1}_{32}) \end{aligned} \tag{3.5b}$$

$$\begin{aligned} H'_{r,r+1} \equiv {} & (A^r_{02} + A^r_{20})(A^{r+1}_{02} + A^{r+1}_{20}) + (A^r_{11} - A^r_{33})(A^{r+1}_{11} - A^{r+1}_{33}) \\ & + (A^r_{01} - A^r_{30})(A^{r+1}_{10} - A^{r+1}_{03}) + (A^r_{21} + A^r_{32})(A^{r+1}_{12} + A^{r+1}_{23}) \\ & + (A^r_{10} - A^r_{03})(A^{r+1}_{01} - A^{r+1}_{30}) + (A^r_{12} + A^r_{23})(A^{r+1}_{21} + A^{r+1}_{32}). \end{aligned} \tag{3.5c}$$

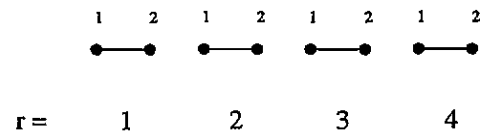

Figure 1. The perfect dimer state and dimer indexing. Each bond represents a singlet configuration as given by equation (2.1).

By (2.6), one can further express H in terms of the three sets of boson operators. For clarity, I use different notations for these three sets of bosons

$$a \equiv a_1,\ a^+ \equiv a_1^+ \qquad b \equiv a_3,\ b^+ \equiv a_3^+ \qquad c \equiv a_2,\ c^+ \equiv a_2^+. \tag{3.6}$$

Now the Hamiltonian of (3.4) can be written as

$$H = H_0 + V \tag{3.7}$$

where H_0 contains only the quadratic terms and a constant

$$\begin{aligned} H_0 = -\frac{3}{8}N + \sum_{r=1}^{N/2} \Big\{ & a_r^+ a_r + b_r^+ b_r + c_r^+ c_r + \frac{1}{4}(2J-1)\big[(a_r - b_r^+)(a_{r+1}^+ - b_{r+1}) \\ & + (a_r^+ - b_r)(a_{r+1} - b_{r+1}^+) + (c_r + c_r^+)(c_{r+1} + c_{r+1}^+)\big] \Big\} \end{aligned} \tag{3.8}$$

and V contains higher-order terms up to the sixth:

$$V = V_3 + V_4 + V_5 + V_6. \tag{3.9}$$

There is a close analogy between the bosonizations in the present case and in Anderson's spin-wave theory [6], where the Néel state is the vacuum state for the two sets of boson operators. Clearly, A_{00} corresponds to s^z in the conventional spin-wave theory, while A_{n0} (A_{0n}) corresponds to s^+ (s^-) etc. Notice that there are three sets of independent boson operators in the present case, but there are only two in the conventional spin-wave theory. This is because the symmetry-broken vacuum state (the Néel state) in the conventional spin-wave theory is in the subspace of zero s^z_{total} ($\equiv \sum_l s^z_l$) while the symmetry-broken vacuum state (the dimer valence-bond state) in the present case is in the subspace of zero vector s_{total} ($\equiv \sum_l s_l$). It is already clear that one should expect a triplet excitation of spin 1 for the present dimerized system.

After Fourier transforming for the three sets of boson operators

$$
\begin{aligned}
a_k &= \sqrt{\frac{2}{N}} \sum_{r=1}^{N/2} \exp(-2\mathrm{i}kr) a_r \\
b_k &= \sqrt{\frac{2}{N}} \sum_{r=1}^{N/2} \exp(-2\mathrm{i}kr) b_r \\
c_k &= \sqrt{\frac{2}{N}} \sum_{r=1}^{N/2} \exp(-2\mathrm{i}kr) c_r
\end{aligned}
\tag{3.10}
$$

(the factor of 2 in the exponential functions is due to the double spacings in the dimer index r), H_0 of (3.8) can be diagonalized by obtaining the usual Bogoliubov transformations

$$
\begin{aligned}
a_k &= \cosh\theta_k \alpha_k - \sinh\theta_k \beta^+_{-k} \\
b_k &= -\sinh\theta_k \alpha^+_{-k} + \cosh\theta_k \beta_k \\
c_k &= \cosh\theta_k \gamma_k + \sinh\theta_k \gamma^+_{-k}
\end{aligned}
\tag{3.11}
$$

where θ_k is given by

$$
\tanh 2\theta_k = \frac{(1-2J)\cos 2k}{2-(1-2J)\cos 2k}. \tag{3.12}
$$

The diagonalized Hamiltonian H_0 can be simply written as

$$
H_0 = \sum_k \omega_k (\alpha^+_k \alpha_k + \beta^+_k \beta_k + \gamma^+_k \gamma_k) + E_0 \tag{3.13}
$$

where the triplet spectrum is given by

$$
\omega_k = \sqrt{1-(1-2J)\cos 2k} \tag{3.14}
$$

which agrees with Parkinson [7] at $J = 0$, and where E_0 is defined as

$$
\frac{E_0}{N} = \frac{3}{4} \sum_k \left[\sqrt{1-(1-2J)\cos 2k} - 1\right] - \frac{3}{8}. \tag{3.15}
$$

In equations (3.13) and (3.15) the summation over k is defined as

$$
\sum_k \equiv \frac{1}{2\pi} \int_{-\pi}^{\pi} \mathrm{d}k. \tag{3.16}
$$

It is well known [17] that the ground state $|\Phi_0\rangle$ of the quadratic Hamiltonian H_0 is given by the two-body form as

$$|\Phi_0\rangle = \exp\left[-\sum_k \tanh\theta_k \left(a_k^+ b_{-k}^+ - \frac{1}{2}c_k^+ c_{-k}^+\right)\right]|D\rangle \tag{3.17}$$

where θ_k is determined by (3.12) and $|D\rangle$ is the boson vacuum state of (3.2).

The ground-state energy E_g within this spin-wave theory is given by the expectation value of the full Hamiltonian of (3.7) with respect to $|\Phi_0\rangle$. Clearly, the odd-body terms yield zero, and one has $E_g = E_0 + \langle V_4\rangle + \langle V_6\rangle$, where $\langle V_4\rangle$ and $\langle V_6\rangle$ can be calculated by Wick's theorem. If one ignores V_4 and V_6 which represent spin-wave interactions, the ground-state energy is then approximated by E_0 of (3.15). This is the result shown in figure 2, where I also include for comparison the numerical results of Tonegawa and Harada [18], obtained by extrapolating the finite-size calculations for $J < 1/2$, and the exact results by Parkinson [19] of the $N = 20$ system for $J > 1/2$. Notice that at $J = 1/2$ (the Majumdar–Ghosh point), equation (3.15) gives the exact result, namely $E_0/N = -3/8$. This is not surprising because $|\Phi_0\rangle = |D\rangle$ at this point. At $J = 0$ (Heisenberg), one has $E_0/N = -0.4498$, whereas the exact result by the Bethe *ansatz* [14] is -0.4432; but one should be careful here because $J = 0$ corresponds to one of the terminating points at which the spin-wave theory is most unreliable, as discussed below.

From the triplet spectrum of (3.14) one sees that there are two terminating points, $J = 0$ and 1, beyond which (i.e., for $J < 0$ and $J > 1$) the spin-wave excitations are unstable. In figure 3, the triplet excitation spectrum is shown schematically for several values of the coupling constant J. It is clear that in the region $0 < J < 1$, there is a non-zero gap, and that this gap collapses at the terminating points. In particular, the triplet spectrum is flat with a gap of 1 at $J = 1/2$. The flatness reflects the fact that at $J = 1/2$, H_0 of (3.8) contains no coupling at all between pairs of spins (dimers). More realistic calculations for the excitations at $J = 1/2$ were performed by Shastry and Sutherland and others [20]. They obtained a spectrum of soliton-like excitations with the minimum gap of 0.25 at $k = 0$ and π and the maximum gap of 1 at $k = \pi/2$. Tonegawa and Harada's numerical calculations [18] confirmed the non-zero gap at $J = 1/2$ and in the nearby region. They predicted that the gap collapses at $J \approx 0.3$, while Haldane [21], who used a fermion representation, predicted this value to be about 1/6. This gapless point seems to signal a phase transition from the dimerized phase to a critical phase similar to what is predicted by the Heisenberg model at $J = 0$. In any case, the triplet spectrum of $\sqrt{2}\sin k$ from (3.14) at $J = 0$ seems to agree well with the exact result of $(\pi/2)\sin k$, as pointed out by Parkinson [7].

A more intriguing situation occurs for $J > 1/2$, where the spin-wave spectrum has a minimum at $k = \pi/2$. In particular, at $J = 1$, the spectrum is gapless with a cusp at $k = \pi/2$. Whether or not this suggests a phase change in the spatial periodicity of the system from double to fourfold, for example, is still unclear. The numerical calculations of the structure factor by Tonegawa and Harada [18] certainly showed a complicated feature for $J > 1/2$. There is also numerical evidence in the excitation spectra, which suggests that the spatial periodicity is not twofold in the region near $J = 1$ [19]. Clearly, the spin-wave theory described here is not adequate for this task and higher-order calculations are needed.

One can also straightforwardly calculate the long-range dimerization order parameter within the present spin-wave theory. The dimer order parameter D is defined as

$$D \equiv \langle s_{l-1}\cdot s_l\rangle - \langle s_l \cdot s_{l+1}\rangle = \langle s_1(r)\cdot s_2(r)\rangle - \langle s_2(r)\cdot s_1(r+1)\rangle \tag{3.18}$$

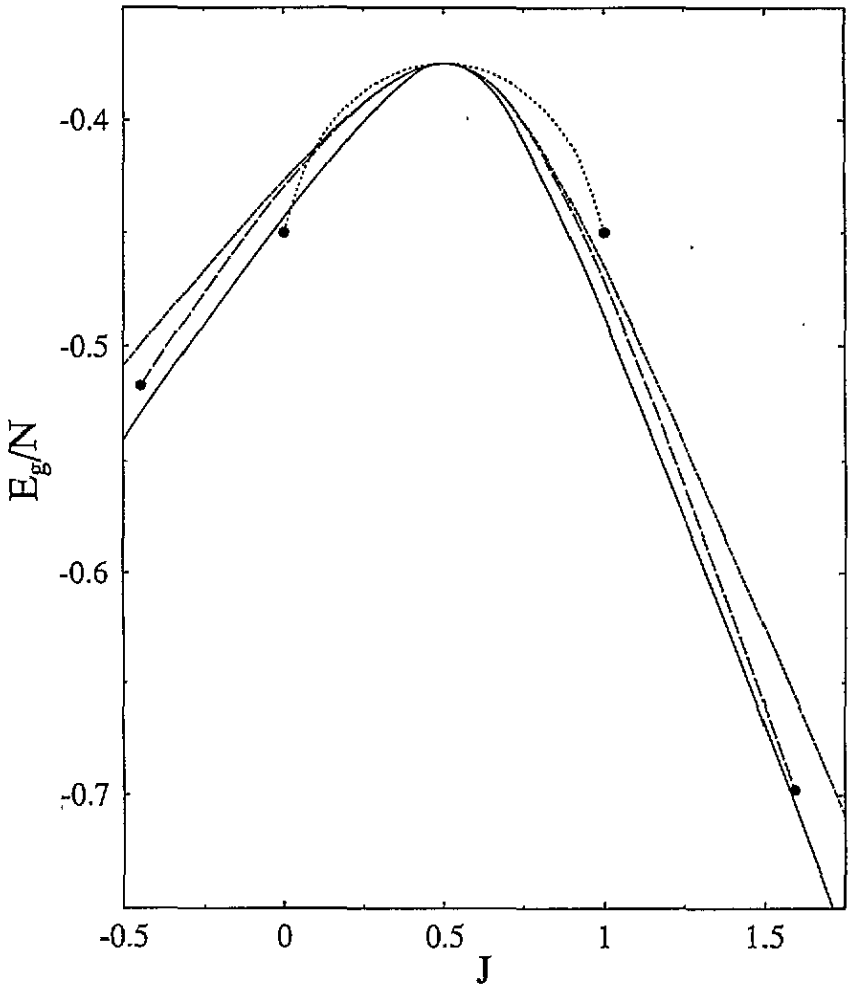

Figure 2. Ground-state energy per spin as a function of the coupling constant J. Shown are results from the spin-wave theory (dotted), the SUB2-2 scheme (dashed), and the full SUB2 scheme (long dashed). The terminating points of the SUB2 scheme and spin-wave theory are indicated. The numerical results from [19, 20] are also included (solid).

where the expectation is with respect to the ground state of the system. From (2.5) and (2.6), using the spin-wave ground state $|\Phi_0\rangle$ of (3.17), it is easy to show that in the spin-wave approximation, D is non-zero in the region $0 < J < 1$ and gradually diminishes when J moves toward the two terminating points. But at the terminating points ($J = 0, 1$), D diverges to $-\infty$, implying a breakdown of the spin-wave theory. In the following section, I provide an alternative approach to the dimerization problem by applying the microscopic CCM.

4. The coupled-cluster approach

The CCM is widely recognized nowadays as providing one of the most universally applicable,

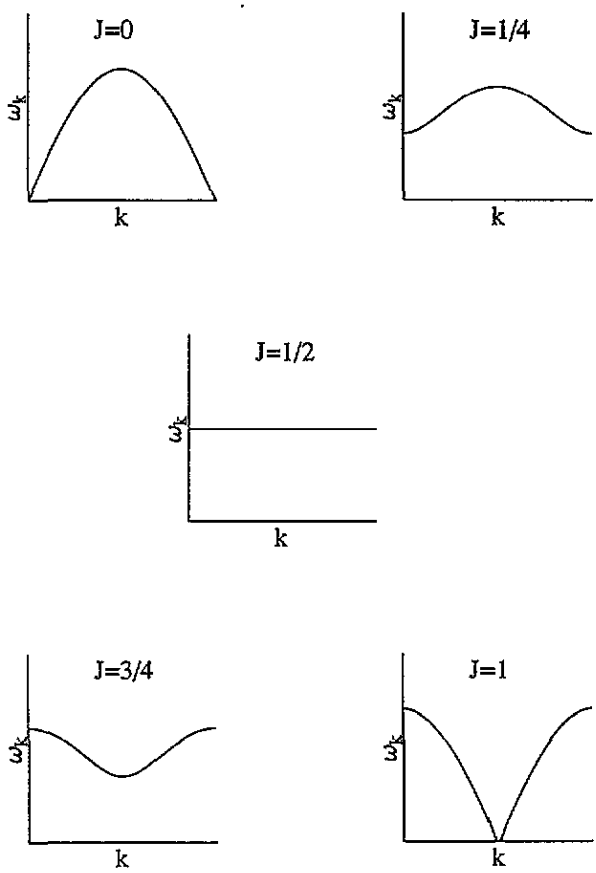

Figure 3. Schematic plots of the triplet excitation spectrum of (3.14) for various values of the coupling constant J.

most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The recent application of the CCM to various spin models has produced excellent numerical results [14, 15]. It therefore seems appropriate and timely to apply the CCM to the dimerization problem. The interested reader is referred to [13] for the general formalism of the CCM and to [14] for its particular application to the spin systems with an anticipated Ising-like long-range order.

Generally speaking, the CCM starts with a proper model state $|\Phi\rangle$, which is usually a simple, uncorrelated many-body wavefunction, and incorporates many-body correlations on top of $|\Phi\rangle$ by acting on it with an exponentiated correlation operator S. This operator S consists of purely so-called configuration creation operators with respect to the model state $|\Phi\rangle$, and is partitioned by one-body, two-body, ..., up to N-body correlation operators with N the number of particles in the system. Thus, the CCM *ansatz* for the ground ket state is

$$|\Psi_g\rangle = e^S|\Phi\rangle. \tag{4.1}$$

The Schrödinger equation of the ground state, after a simple manipulation, can then be written as

$$e^{-S}He^S|\Phi\rangle = E_g|\Phi\rangle \tag{4.2}$$

where E_g is the ground-state energy, and where the similarity-transformed Hamiltonian can be expressed as a series of nested commutators

$$e^{-S}He^{S} = H + [H, S] + \frac{1}{2!}[[H, S], S] + \cdots \tag{4.3}$$

which usually terminates at the fourth-order for most Hamiltonians with pair-interaction potentials [13]. Equations (4.1)–(4.3) are the hallmarks of the CCM.

For the case of dimerization under consideration, it is natural to choose the dimer state $|D\rangle$ of (3.2) as the model state, namely $|\Phi\rangle = |D\rangle$. The configuration creation operators with respect to this model state $|D\rangle$ are clearly given by any combinations of the three operators A^r_{10}, A^r_{20}, and A^r_{30}. Their Hermitian conjugates are the corresponding annihilation operators. Since the antiferromagnetic ground state is definitely in the sector of zero s^z_{total}, the correlation operator S is in general written in the form

$$S = \sum_{n=1}^{N/2} S_n \tag{4.4}$$

where $N/2$ is the total number of valence bonds of the spin-$\frac{1}{2}$ chain, and the n-body correlation operators S_n are given respectively by

$$S_1 \equiv \sum_{r=1}^{N/2} \mathcal{S}_r A^r_{20} \tag{4.5a}$$

$$S_2 \equiv \sum_{r,r'}{}' \left[\mathcal{S}^{(1)}_{r,r'} A^r_{10} A^{r'}_{30} - \frac{1}{2!} \mathcal{S}^{(2)}_{r,r'} A^r_{20} A^{r'}_{20} \right] \tag{4.5b}$$

$$S_3 \equiv \sum_{r,r',r''}{}' \left[\mathcal{S}^{(1)}_{r,r',r''} A^r_{10} A^{r'}_{30} A^{r''}_{20} - \frac{1}{3!} \mathcal{S}^{(2)}_{r,r',r''} A^r_{20} A^{r'}_{20} A^{r''}_{20} \right] \tag{4.5c}$$

etc. In (4.5b) and (4.5c) the primes on the summations imply exclusion of the terms with any pair of indices equal. We notice the similarity between the spin-wave ground state $|\Phi_0\rangle$ of (3.17) and the CCM state $|\Psi_g\rangle$ of (4.1) if S is replaced by S_2. W also notice the similarity between the present CCM analysis and that of [15] where the spin-1 model state is given by the simple planar configuration with $s^z_l = 0$ for all sites l.

The ground-state energy is obtained by taking the inner product of the Schrödinger equation (4.2) with the model state $|D\rangle$ itself, namely

$$E_g = \langle D|e^{-S}He^{S}|D\rangle. \tag{4.6}$$

The correlation coefficients $\{\mathcal{S}_{r,r',\ldots}\}$ are determined from the coupled set of equations obtained by taking inner products of (4.2) with states constructed from the corresponding annihilation operators, namely

$$\langle D|A^r_{02}e^{-S}He^{S}|D\rangle = 0 \qquad \text{for all } r \tag{4.7}$$

for the one-body equation;

$$\langle D|A^r_{01}A^{r'}_{03}e^{-S}He^{S}|D\rangle = 0 \qquad \text{for all } r, r'(\neq r) \tag{4.8}$$

and

$$\langle D|A_{02}^{r}A_{02}^{r'}\mathrm{e}^{-S}H\mathrm{e}^{S}|D\rangle = 0 \qquad \text{for all } r, r'(\neq r) \tag{4.9}$$

for the two-body equations. The three-body equations and higher-order many-body equations are obtained in a similar fashion.

One sees that the similarity-transformed Hamiltonian of (4.3) is needed in all of the above equations. I leave details of the derivation to the appendix, and only point out that, as expected, the otherwise infinite expansion series of (4.3) does indeed terminate at the fourth-order term. In fact, the exact energy equation (4.6) can be straightforwardly derived as

$$\frac{E_g}{N} = \frac{1}{8}[(1-2J)(2b_1^{(1)} + b_1^{(2)} - a) - 3] \tag{4.10}$$

where I have used the translational and reflectional symmetries, setting accordingly

$$S_r = a \qquad S_{r_1,r_2}^{(i)} = S_{r_2,r_1}^{(i)} = b_r^{(i)} \qquad \text{with } i = 1,2 \text{ and } r = r_2 - r_1 \tag{4.11}$$

It is also obvious that $b_{-r}^{(i)} = b_r^{(i)}$.

The exact one-body equation of (4.7) can also be easily derived. It couples only to the two-body coefficients. Similarly, the two-body equations of (4.8) and (4.9) couple only to the one-body and the three-body coefficients, and so on. From the one-body equation, it is interesting to note that the physical solution is given by $a = 0$, implying no one-body correlations for the dimerization problem. This is not surprising because the model state $|D\rangle$ is in the sector of zero total spin vector (i.e., $s_{\text{total}} = 0$), and the one-body correlation operator S_1 of (4.5*a*) will take the state out of this sector. Furthermore, if one assumes that the two sets of the two-body correlation coefficients are identical, i.e.

$$b_r^{(1)} = b_r^{(2)} \equiv b_r \tag{4.12}$$

the two-body correlation operator S_2 then commmutes with the total spin vector s_{total}. This is a necessary condition if one requires the CCM ground state to be in the sector of zero s_{total}. (Actually, one requires every correlation operator S_n to commute with s_{total}.) The energy equation is now reduced to

$$\frac{E_g}{N} = \frac{3}{8}[(1-2J)b_1 - 1]. \tag{4.13}$$

One clearly needs to employ an approximation scheme for any practical calculation. The most common approximation scheme in the CCM is the so-called SUBn scheme, in which one keeps up to n-body correlation operators and sets all the higher-order many-body correlation operators S_m $(m > n)$ to zero. I consider the SUB2 scheme here. I find that within the SUB2 scheme, the condition (4.12) is indeed satisfied. After simplification, the two identical two-body equations are given by

$$\frac{1}{2}\sum_{\rho=\pm 1}\left(K_3\delta_{r\rho} + K_2 b_r - 2K_1 b_{r+\rho} + K_1\sum_{r'\neq 0} b_{r'}b_{r+\rho-r'}\right) = 0 \qquad r \neq 0 \tag{4.14}$$

and

$$K_1 \equiv 1-2J \qquad K_2 \equiv 4(1-2K_1b_1) \qquad K_3 \equiv K_1(1+4b_1^2) - 2(1+2J)b_1. \tag{4.15}$$

A simpler approximation can be made from the full SUB2 equation (4.14), namely the so-called SUB2-2 scheme in which one keeps only the single coefficient, b_1, setting all other b_r equal to zero ($|r| > 1$). Equation (4.14) then reduces to

$$1 - 2J + 2(3 - 2J)b_1 - 9(1 - 2J)b_1^2 = 0 \tag{4.16}$$

with the physical solution

$$b_1 = \frac{1}{9(1-2J)}[3 - 2J - \sqrt{40J^2 - 48J + 18}]. \tag{4.17}$$

The full SUB2 equation (4.14) can also be solved via a Fourier transformation exactly similar to (3.9) of [15]. Here I only quote the final result given by the following self-consistency equation for b_1:

$$b_1 = \frac{1}{3K_1}\left(2 - \frac{K_2}{2}\frac{1}{2\pi}\int_{-\pi}^{\pi} \mathrm{d}q\, \sqrt{1 - k_1\cos 2q + k_2\cos^2 2q}\right) \tag{4.18}$$

where the constants k_1 and k_2 are defined by

$$k_1 \equiv \frac{1}{K_2^2}(4K_1K_2 + 8K_1^2 b_1 - 4K_1^2 X) \qquad k_2 \equiv \frac{4K_1(K_1 - K_3)}{K_2^2} \tag{4.19a}$$

and where X is defined by

$$X \equiv \sum_{r=1}^{N/2} b_r b_{r+1} = \frac{1}{2\pi}\int_{-\pi}^{\pi} \mathrm{d}q\, \frac{1}{4K_1^2\cos 2q} \times \left(2K_1\cos 2q - K_2 + K_2\sqrt{1 - k_1\cos 2q + k_2\cos^2 2q}\right)^2. \tag{4.19b}$$

After obtaining b_1 as a function of J from (4.17) or (4.18), the ground-state energy is then given by substituting b_1 into (4.13). These ground-state energies are shown in figure 2, together with the results of the spin-wave theory and of the numerical calculations [19, 20] for comparison. As in the spin-wave theory described in section 3, at $J = 1/2$ (the Majumdar–Ghosh point) the exact result is recovered for both the SUB2-2 and full SUB2 schemes—namely, $b_1 = 0$ and $E_g/N = -3/8$. At the Heisenberg point ($J = 0$), the SUB2-2 and full SUB2 schemes give $E_g/N = -0.4268, -0.4298$ respectively, slightly higher values than the exact result of -0.4432. Furthermore, it is interesting to observe that in the full SUB2 scheme, there are also two terminating points, $J_c^{(1)} = -0.4443$ and $J_c^{(2)} = 1.591$, beyond which, namely for $J < J_c^{(1)}$ and $J > J_c^{(2)}$, there is no real solution in (4.18). The corresponding energy values are -0.5172 and -0.6977 respectively. It has previously been argued that CCM SUB2 terminating points may correspond to the phase transition critical points in the past [14, 15]. It seems reasonable to consider that possibility again here. From our past experience, I believe that the higher-order approximations for the ground state, and the calculations of the low-lying excitation spectra and of spin correlation functions will reveal more information about the possible phase transitions. For present purposes, one sees from figure 2 that the extremely simple SUB2 scheme gives much better results for a wide range of the coupling constant J than does the spin-wave theory, at least as far as the ground-state energy is concerned.

5. Discussion

In this paper, I have studied the dimerization problem via a microscopic approach, employing the proper set of composite operators of Parkinson [7]. Two approximation schemes, namely the spin-wave theory and a CCM analysis, have been applied to the 1D frustrated spin-$\frac{1}{2}$ model. The ground-state and low-lying excited energies are obtained as functions of the coupling constant. The implications of possible phase transitions at the naturally arising terminating points of the solutions have been discussed. Another approach may be provided by using a variational trial wave function of the type of equations (4.1) and (4.5); this is similar to what was done in the calculation of Sachdev for the spin-$\frac{1}{2}$ Heisenberg model [22].

From the present preliminary attempt to formulate a microscopic theory for the dimerization problem, it is clear that higher-order calculations within the present analysis are needed for both the ground and excited states. The very successful applications [14] of the CCM to the spin systems with an Ising-like long-range order seem to suggest that the CCM can also provide a systematic and potentially accurate approximation scheme for the dimerization problem. Furthermore, within the formalism presented in this paper, it is straightforward to extend the same analysis to both higher-order dimensionality and/or spin systems with spin quantum number greater than one half. In particular, the 2D spin-$\frac{1}{2}$ Heisenberg model on the square lattice with J_1–J_2 couplings [9, 11] has been under intensive study for its possible dimerization. An equally interesting Hamiltonian model is provided by the 1D spin-1 Heisenberg biquadratic systems, where it is known the ground state is dimerized at a particular coupling constant [4], and where trimerization is also possible in another region [10].

Acknowledgments

I am grateful to J B Parkinson for drawing my attention to [7] which forms the basis of this work and for providing some numerical results of finite-size calculations prior to publication, and to R F Bishop for suggesting application of the CCM to this problem. I also thank C E Campbell, N J Davidson, C Zeng and D J J Farnell for many useful discussions.

Appendix

In this appendix, I derive the similarity transformations within the SUB1 and SUB2 schemes of the CCM, described in section 4.

Notice that in the similarity transformation of (4.3), any quadratic term in the Hamiltonian of equations (3.4)–(3.5) can be transformed as

$$\mathrm{e}^{-S} A_{ij}^{r} A_{kl}^{r+1} \mathrm{e}^{S} = \left(\mathrm{e}^{-S} A_{ij}^{r} \mathrm{e}^{S}\right)\left(\mathrm{e}^{-S} A_{kl}^{r+1} \mathrm{e}^{S}\right) \tag{A1}$$

and each similarity-transformed operator can be expanded as a series of nested commutators

$$\mathrm{e}^{-S} A_{ij}^{r} \mathrm{e}^{S} = A_{ij}^{r} + [A_{ij}^{r}, S] + \frac{1}{2!}[[A_{ij}^{r}, S], S] + \cdots. \tag{A2}$$

Since the correlation operator S consists only of the creation operators A_{10}, A_{30} and A_{20}, the expansion series of (A2) terminates at first order in S for all A_{00} and A_{nm} with $n, m \neq 0$,

and at second order in S for the pure annihilation operators A_{0n} with $n \neq 0$, by the pseudo-spin algebra of (2.4). Therefore the similarity-transformed Hamiltonian of (4.3) terminates at fourth order.

In the SUB1 scheme, one makes the replacement $S \to S_1$, where S_1 is given by (4.5a). From (A2), it is straightforward to derive the following SUB1 similarity transformations:

$$\begin{aligned} &\tilde{A}^r_{n0} = A^r_{n0} \qquad \tilde{A}^r_{nm} = A^r_{nm} \\ &\tilde{A}^r_{00} = A^r_{00} - \mathcal{S}_r A^r_{20} \qquad \tilde{A}^r_{n2} = A^r_{n2} + \mathcal{S}_r A^r_{n0} \\ &\tilde{A}^r_{02} = A^r_{02} + \mathcal{S}_r (A^r_{00} - A^r_{22}) - \mathcal{S}_r^2 A^r_{20} \end{aligned} \tag{A3}$$

where $n = 1, 2, 3$ and $m = 1, 3$, and where the definition

$$\tilde{A}^r_{ij} \equiv \mathrm{e}^{-\mathcal{S}_1} A^r_{ij} \mathrm{e}^{\mathcal{S}_1} \tag{A4}$$

is used.

In the SUB2 scheme, one makes the replacement $S \to S_1 + S_2$. One can firstly make the SUB1 similarity transform by using (A3), and then apply the following SUB2 similarity transform for each A^r_{ij}:

$$\bar{A}^r_{n0} = A^r_{n0} \tag{A5a}$$

$$\bar{A}^r_{n1} = A^r_{n1} + \sum_{r'}{}' \mathcal{S}^{(1)}_{r,r'} A^r_{n0} A^{r'}_{30} \tag{A5b}$$

$$\bar{A}^r_{n2} = A^r_{n2} - \sum_{r'}{}' \mathcal{S}^{(2)}_{r,r'} A^r_{n0} A^{r'}_{20} \tag{A5c}$$

$$\bar{A}^r_{n3} = A^r_{n3} + \sum_{r'}{}' \mathcal{S}^{(1)}_{r',r} A^r_{n0} A^{r'}_{10} \tag{A5d}$$

$$\bar{A}^r_{00} = A^r_{00} - \sum_{r'}{}' (\mathcal{S}^{(1)}_{r,r'} A^r_{10} A^{r'}_{30} + \mathcal{S}^{(1)}_{r',r} A^{r'}_{10} A^r_{30} - \mathcal{S}^{(2)}_{r,r'} A^r_{20} A^{r'}_{20}) \tag{A5e}$$

$$\begin{aligned} \bar{A}^r_{01} = A^r_{01} &+ \sum_{r'}{}' \big(\mathcal{S}^{(1)}_{r,r'} A^{r'}_{30} (A^r_{00} - A^r_{11}) - \mathcal{S}^{(1)}_{r',r} A^{r'}_{10} A^r_{31} + \mathcal{S}^{(2)}_{r,r'} A^{r'}_{20} A^r_{21}\big) \\ &+ \frac{1}{2} \sum_{r',r''}{}' \mathcal{S}^{(1)}_{r,r'} A^{r'}_{30} \big(\mathcal{S}^{(2)}_{r,r''} A^r_{20} A^{r''}_{20} - 2\mathcal{S}^{(1)}_{r,r''} A^r_{10} A^{r''}_{30} - \mathcal{S}^{(1)}_{r'',r} A^{r''}_{10} A^r_{30}\big) \\ &- \frac{1}{2} \sum_{r',r''}{}' \big(\mathcal{S}^{(1)}_{r',r} \mathcal{S}^{(1)}_{r,r''} A^{r'}_{10} A^r_{30} A^{r''}_{30} - \mathcal{S}^{(2)}_{r,r'} \mathcal{S}^{(1)}_{r,r''} A^{r'}_{20} A^r_{20} A^{r''}_{30}\big) \end{aligned} \tag{A5f}$$

$$\begin{aligned} \bar{A}^r_{02} = A^r_{02} &- \sum_{r'}{}' \big(\mathcal{S}^{(2)}_{r,r'} A^{r'}_{20} (A^r_{00} - A^r_{22}) + \mathcal{S}^{(1)}_{r',r} A^{r'}_{10} A^r_{32} + \mathcal{S}^{(1)}_{r,r'} A^{r'}_{30} A^r_{12}\big) \\ &- \frac{1}{2} \sum_{r',r''}{}' \mathcal{S}^{(2)}_{r,r'} A^{r'}_{20} \big(2\mathcal{S}^{(2)}_{r,r''} A^r_{20} A^{r''}_{20} - \mathcal{S}^{(1)}_{r,r''} A^r_{10} A^{r''}_{30} - \mathcal{S}^{(1)}_{r'',r} A^{r''}_{10} A^r_{30}\big) \\ &+ \frac{1}{2} \sum_{r',r''}{}' \big(\mathcal{S}^{(1)}_{r',r} \mathcal{S}^{(2)}_{r,r''} A^{r'}_{10} A^r_{30} A^{r''}_{20} + \mathcal{S}^{(1)}_{r,r''} \mathcal{S}^{(2)}_{r,r'} A^{r''}_{30} A^r_{10} A^{r'}_{20}\big) \end{aligned} \tag{A5g}$$

$$\bar{A}^r_{03} = A^r_{03} + \sum_{r'}{}' \big(\mathcal{S}^{(1)}_{r',r} A^{r'}_{10} (A^r_{00} - A^r_{33}) - \mathcal{S}^{(1)}_{r,r'} A^{r'}_{30} A^r_{13} + \mathcal{S}^{(2)}_{r,r'} A^{r'}_{20} A^r_{23}\big)$$

$$+\frac{1}{2}\sum_{r',r''}{}' \mathcal{S}^{(1)}_{r',r} A^{r'}_{10}\left(\mathcal{S}^{(2)}_{r,r''} A^{r}_{20} A^{r''}_{20} - 2\mathcal{S}^{(1)}_{r'',r} A^{r}_{30} A^{r''}_{10} - \mathcal{S}^{(1)}_{r,r''} A^{r''}_{30} A^{r}_{10}\right)$$

$$-\frac{1}{2}\sum_{r',r''}{}' \left(\mathcal{S}^{(1)}_{r,r'} \mathcal{S}^{(1)}_{r'',r} A^{r'}_{30} A^{r}_{10} A^{r''}_{10} - \mathcal{S}^{(2)}_{r,r'} \mathcal{S}^{(1)}_{r'',r} A^{r'}_{20} A^{r}_{20} A^{r''}_{10}\right) \tag{A5h}$$

where $n = 1, 2, 3$, and the primes on the summations imply exclusion of any pair of indices that are equal, and where the operators with a bar represent the similarity transform for the S_2 correlation operator

$$\bar{A}^{r}_{ij} \equiv \mathrm{e}^{-S_2} A^{r}_{ij} \mathrm{e}^{S_2}. \tag{A6}$$

In deriving (A5), I have used the fact that

$$\mathcal{S}^{(1)}_{r,r} = \mathcal{S}^{(2)}_{r,r} = 0 \qquad \mathcal{S}^{(2)}_{r,r'} = \mathcal{S}^{(2)}_{r',r}. \tag{A7}$$

After making the similarity transformations of (A3) and (A5), the CCM equations (4.6)–(4.9) can be derived by using the pseudo-spin algebra of (2.4) to move all the creation operators A_{10}, A_{20}, A_{30} in each term to the left, and all the annihilation operators A_{01}, A_{02}, A_{03} (also A_{nm}, $n, m \neq 0$) in each term to the right, and by using the fact that

$$\langle D|A^{r}_{n0} = A^{r}_{mn}|D\rangle = 0 \qquad n \neq 0 \tag{A8}$$

and

$$\langle D|A^{r}_{00} = \langle D| \qquad A^{r}_{00}|D\rangle = |D\rangle \tag{A9}$$

for all r.

References

[1] Majumdar C K and Ghosh D K 1970 *J. Phys. C: Solid State Phys.* **3** 91; 1969 *J. Math. Phys.* **10** 1388, 1399

[2] Affleck I, Kennedy T, Lieb E and Tasaki H 1987 *Phys. Rev. Lett.* **59** 799

[3] Parkinson J B 1988 *J. Phys. C: Solid State Phys.* **20** 21
Barber M N and Batchelor M T 1989 *Phys. Rev.* B **40** 4621
Klümper A 1989 *Europhys. Lett.* **9** 815
Affleck I 1990 *J. Phys.: Condens. Matter* **2** 405

[4] Xian Y 1993 *Phys. Lett.* **183A** 437

[5] The term 'valence-bond lattices' is used in this article in order to distinguish what is considered here from the well known 'valence-bond solids' defined in [2]; it indicates a particular valence-bond configuration of a spin-1 many-spin system, which has no ordinary symmetry breaking.

[6] Anderson P W 1952 *Phys. Rev.* **86** 694
Kubo R 1952 *Phys. Rev.* **87** 568
Oguchi T 1960 *Phys. Rev.* **117** 117

[7] Parkinson J B 1979 *J. Phys. C: Solid State Phys.* **12** 2873

[8] Chubukov A V 1991 *Phys. Rev.* B **43** 3337

[9] Read N and Sachdev S 1989 *Phys. Rev. Lett.* **62** 1694; 1990 *Phys. Rev.* B **42** 4568

[10] Xian Y 1993 *J. Phys.: Condens. Matter* **5** 7489

[11] Singh R R P and Narayanan R 1990 *Phys. Rev. Lett.* **65** 1072
Chubukov A V and Jolicoeur Th 1991 *Phys. Rev.* B **44** 12 050

[12] Dyson F J 1956 *Phys. Rev.* **102** 1217, 1230
Maléev S V 1958 *Sov. Phys.–JETP* **6** 776
For the pseudo-spin operators in the present context, the Holstein–Primakoff transformations were given by

Mead L R and Papanicolaou N 1983 *Phys. Rev.* B **28** 1633
[13] Bishop R F and Kümmel H 1987 *Phys. Today* **40** 52
Bishop R F 1991 *Theor. Chim. Acta* **80** 95
[14] Bishop R F, Parkinson J B and Xian Y 1991 *Phys. Rev.* B **43** 13782; 1991 *Phys. Rev.* B **44** 9425; 1992 *J. Phys.: Condens. Matter* **4** 5783
[15] Bishop R F, Parkinson J B and Xian Y 1993 *J. Phys.: Condens. Matter* **5** 9169
[16] den Nijs M and Rommelse K 1989 *Phys. Rev.* B **40** 4709
Santoro G and Fabrizio M 1994 Disordered flat phase in a solid on solid model of FCC(110) surface and dimer states in quantum spin-$\frac{1}{2}$ chains *Preprint*
[17] Blaizot J P and Ripka G 1986 *Quantum Theory of Finite Systems* (London: MIT)
[18] Tonegawa T and Harada I 1987 *J. Phys. Soc. Japan* **56** 2153
[19] Courtesy of J B Parkinson.
[20] Shastry S S and Sutherland B 1981 *Phys. Rev. Lett.* **47** 964
Caspers W J, Emmett K M and Magnus W 1984 *J. Phys. A: Math. Gen.* **17** 2697
[21] Haldane F D M 1982 *Phys. Rev.* B **25** 4925
[22] Sachdev S 1989 *Phys. Rev.* B **39** 12232