A Microscopic Approach to the Dimerization in the Frustrated Spin-$\frac{1}{2}$ Antiferromagnetic Chains

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Abstract. The spontaneous dimerization of the frustrated spin-$\frac{1}{2}$ antiferromagnetic chains is studied by 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 by a Dyson-Maléev-like boson transformation. The ground-state properties and the triplet excitation spectra are obtained as functions of the coupling parameter. Secondly, based on the pseudo-spin operators, a microscopic treatment 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.

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I. 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 models have been solved by 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 higher-order 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 nonzero solid-like dimer or trimer long-range order and hence show the characteristics of a quantum solid. (It therefore seems more appropriate to call 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%.

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 equation-of-motions. A triplet excitation spectrum of $\sqrt{2}\sin k$ was obtained. This compares well with the exact result of triplet spectrum of $\frac{7}{2}\sin k$. Chubukov [8] later provided a similar spin-wave theory by using a transformation similar to that of Holstein-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 the 2D spin systems, e.g., the $J_1$-, $J_2$ Heisenberg model on the square lattice [11], etc.

In this article, I intend to investigate the dimerization of spin systems by a systematic, microscopic approach. Because of its 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, a spin-wave theory is developed. 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 (e.g. Ref. [6]). 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, is 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 ar-
tifacts. In fact, the 1D frustrated spin-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 Sec. II the composite operators in a matrix representation are introduced and the corresponding boson transformations are given. Sec. III is devoted to the spin-wave approximation for the 1D spin-1/2 model. The ground-state energy and excitation spectra are obtained as functions of the coupling constants. In Sec. IV 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 by a general discussion in Sec. V.

II. Pseudo-Spin Operators and Their Bosonizations

I first consider a two-spin system, each with spin 1/2. For completeness, some of the analysis by Parkinson [7] is repeated here. Clearly, there are four states for such a two-spin system. If notations |↑⟩ and |↓⟩ are used to represent spin up and down states respectively, the singlet state can be written as

$$|0⟩ = \frac{1}{\sqrt{2}}(|↑↓⟩ - |↓↑⟩),$$  

(2.1)

and the triplet states with $s_{total}^z (≡ s_1^z + s_2^z) = 1, 0, -1$ are given by respectively

$$|1⟩ = |↑↑⟩,$$

$$|2⟩ = \frac{1}{\sqrt{2}}(|↑↓⟩ + |↓↑⟩),$$

$$|3⟩ = |↓↓⟩.$$  

(2.2)

Following Parkinson [7], I employ a matrix representation. Each of the four states of Eqs. (2.1) and (2.2) is then represented by a column matrix with a single nonzero element. One can then introduce operators $A_{ij}$ as having only a single non-zero element of a $(4 \times 4)$ matrix, namely $⟨i'|A_{ij}|j’⟩ = δ_{i'i}δ_{jj'}$. For example, $A_{10}$ ($A_{30}$) is an operator which increases (decreases) $s_{total}^z$ 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, each 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 with spin 1, and hence there are eighty-one (9 × 9) $A_{ij}$ operators which form a complete set for a pair of spin-1 atoms. For three-spin system each with spin 1, the dimensionality of the matrix is twenty-seven [10].

It is worth pointing out that these $A_{ij}$ operators are nonlinear in terms of the original single spin operators, for example,

$$A_{00} = \frac{1}{4} - s_1 \cdot s_2,$$

$$A_{01} = -\frac{1}{\sqrt{2}}(s_1^+ - s_2^+)(s_1^z + s_2^z),$$  

(2.3)

etc. 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,

$$[A_{ij}, A_{kl}] = A_{il}δ_{jk} - A_{kj}δ_{li}.$$  

(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 Sec. III and the CCM approximations in Sec. IV are developed based on this assumption.

In a straightforward manner, one can express the single spin operators in terms of $A_{ij}$ operators [7]. They are given by [17]

$$s_1^z = \frac{1}{2}(A_{02} + A_{20} + A_{11} - A_{33}),$$

$$s_2^z = \frac{1}{2}(-A_{02} - A_{20} + A_{11} - A_{33}),$$  

(2.5a)

$$s_1^+ = \frac{1}{\sqrt{2}}(A_{30} - A_{01} + A_{21} + A_{32}),$$

$$s_2^+ = \frac{1}{\sqrt{2}}(A_{01} - A_{30} + A_{21} + A_{32}),$$  

(2.5b)

$$s_1^- = \frac{1}{\sqrt{2}}(A_{03} - A_{10} + A_{12} + A_{23}),$$

$$s_2^- = \frac{1}{\sqrt{2}}(A_{10} - A_{03} + A_{12} + A_{23}),$$  

(2.5c)
Recognizing that $A_{ij}$ obeys the pseudospin algebra of Eq. (2.4), one can make the following Dyson-Maléev transformation [12],

$$A_{00} = 1 - a_{1}^{+} a_{1} - a_{2}^{+} a_{2} - a_{3}^{+} a_{3};$$
$$A_{ph} = a_{p}^{+} A_{00}, \quad A_{0p} = a_{p};$$
$$A_{pp} = a_{p}^{+} a_{p}, \quad A_{pq} = a_{p}^{+} a_{q};$$

(2.6)

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

$$[a_{p}, a_{p}^{+}] = 1,$$

(2.7)

and with all other commutators yielding zero.

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

$$a_{p}|0\rangle = 0, \quad p = 1, 2, 3.$$

(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 Eqs. (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 by Eqs. (2.5) and then in terms of those three sets of boson operators according to Eqs. (2.6). In Sec. III I consider a spin-wave theory for the frustrated 1D model using these pseudo-spin operators and their bosonizations. In Sec. IV I develop a microscopic formalism within the framework of the CCM also based on these pseudo-spin operators.

III. Spin-Wave Theory

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

$$H = \sum_{i=1}^{N}(s_{i} \cdot s_{i+1} + J s_{i} \cdot s_{i+2}),$$

(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 Eq. (3.1) becomes the well-known Majumdar-Ghosh model [1], which has two degenerate dimer ground-states, with one of them given by

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

(3.2)

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

$$H = \sum_{r=1}^{N/2}[s_{1}(r) \cdot s_{2}(r) + s_{2}(r) \cdot s_{1}(r + 1) + J s_{1}(r) \cdot s_{1}(r + 1) + J s_{2}(r) \cdot s_{2}(r + 1)].$$

(3.3)

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

$$H = \sum_{r=1}^{N/2}(H_{r} + \frac{1}{4}H_{r,r+1} + \frac{1}{2}J H_{r,r+1}'),$$

(3.4)
with
\[ H_0 = \frac{1}{4} - A_{00}^2, \]
\[ H_{r,r+1} = (-A_{02} + A_{20}^r - A_{02}^r)(A_{02}^{r+1} + A_{10}^{r+1} + A_{20}^{r+1} - A_{02}^{r+1}) + \ldots \]
\[ + (A_{00} - A_{00}^r)(A_{00}^{r+1} - A_{00}^{r+1}) + (A_{11} - A_{11}^r)(A_{11}^{r+1} - A_{11}^{r+1}) + \ldots \]
\[ + (A_{10} - A_{00}^r)(A_{10}^{r+1} - A_{00}^{r+1}) + (A_{12} - A_{20}^r)(A_{12}^{r+1} + A_{20}^{r+1}) + \ldots \]
\[ + (A_{12} - A_{20}^r)(A_{12}^{r+1} + A_{20}^{r+1}) + (A_{12} - A_{20}^r)(A_{12}^{r+1} + A_{20}^{r+1}). \]

By Eqs. (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,
\[ a \equiv a_1, \quad a^+ \equiv a_1^+; \quad b \equiv a_3, \quad b^+ \equiv a_3^+; \quad c \equiv a_2, \quad c^+ \equiv a_2^+. \]

Now the Hamiltonian of Eq. (3.4) can be written as
\[ H = H_0 + V, \]
where \( H_0 \) contains only the quadratic terms and a constant,
\[ H_0 = -\frac{3}{8} N + \sum_{r=1}^{N/2} \left\{ a_r^+ a_r + b_r^+ b_r + c_r^+ c_r + \frac{1}{4} (2J - 1) [(a_r - b_r)^+ (a_{r+1}^+ - b_{r+1}) + (a_r^+ - b_r)(a_{r+1} - b_{r+1}) + (c_r + c_r^+)(c_{r+1} + c_{r+1})] \right\}, \]
and \( V \) contains higher-order terms up to the sixth,
\[ V = V_5 + V_4 + V_5 + V_6. \]

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 \( A_{00} \) (\( A_{0n} \)) 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 \( s_{\text{total}}^0 \) (\( \equiv \sum_i s_i \)) while the symmetry-broken vacuum state (the dimer valence-bond state) in the present case is in the subspace of zero vector \( s_{\text{total}} \) (\( \equiv \sum_i s_i \)). It is already clear that one should expect a triplet excitation of spin 1 for the present dimerized system.

After Fourier transformations for the three sets of boson operators,
\[ a_k = \sqrt{\frac{2}{N}} \sum_{r=1}^{N/2} \exp(-2i kr) a_r, \quad b_k = \sqrt{\frac{2}{N}} \sum_{r=1}^{N/2} \exp(-2i kr) b_r, \]
\[ c_k = \sqrt{\frac{2}{N}} \sum_{r=1}^{N/4} \exp(-2i kr) c_r, \]
the factor of 2 in the exponential functions is due to the double spacings in the dimer index \( r \). \( H_0 \) of Eqs. (3.8) can be diagonalized by the usual Bogoliubov transformations,
\[ a_k = \cosh \theta_k a_k - \sinh \theta_k b_k, \quad b_k = -\sinh \theta_k a_k^+ + \cosh \theta_k b_k, \]
\[ c_k = \cosh \theta_k c_k + \sinh \theta_k c_k, \]
\[ a_k = \cosh \theta_k a_k - \sinh \theta_k b_k, \quad b_k = -\sinh \theta_k a_k^+ + \cosh \theta_k b_k, \]
\[ c_k = \cosh \theta_k c_k + \sinh \theta_k c_k, \]
where $\theta_k$ is given by
\[
\tanh 2\theta_k = \frac{(1 - 2J) \cos 2k}{2 - (1 - 2J) \cos 2k}.
\]  

(3.12)

The diagonalized Hamiltonian $H_0$ can be simply written as
\[
H_0 = \sum_k \omega_k (\alpha_k^+ \alpha_k + \beta_k^+ \beta_k + \gamma_k^+ \gamma_k) + E_0,
\]

(3.13)

where the triplet spectrum is given by
\[
\omega_k = \sqrt{1 - (1 - 2J) \cos 2k},
\]

(3.14)

which agrees with Parkinson [7] at $J = 0$, and where $E_0$ is defined as
\[
\frac{E_0}{N} = \frac{3}{4} \sum_k \left[ \sqrt{1 - (1 - 2J) \cos 2k} - 1 \right] - \frac{3}{8}.
\]

(3.15)

In Eqs. (3.13) and (3.15) the summation over $k$ is defined as
\[
\sum_k \equiv \frac{1}{2\pi} \int_{-\pi}^\pi dk.
\]

(3.16)

It is well known [18] that the ground state $|\Phi_0\rangle$ of the quadratic Hamiltonian $H_0$ is given by the two-body form as
\[
|\Phi_0\rangle = \exp \left[ -\sum_k \tanh \theta_k \left( a_k^+ b_k^+ - \frac{1}{2} c_k^+ c_k^+ \right) \right] |D\rangle,
\]

(3.17)

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

The ground-state energy $E_g$ within this spin-wave theory is given by the expectation value of the full Hamiltonian of Eq. (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 Eq. (3.15). This is the result shown in Fig. 2, where I also include for comparison the numerical results of Tonegawa and Harada [19], obtained by extrapolating the finite-size calculations for $J < 1/2$, and the exact results by Parkinson [20] of the $N = 20$ system for $J > 1/2$. Notice that at $J = 1/2$ (the Majumdar-Ghosh point), Eq. (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 Eq. (3.14) one sees that there are two terminating points, $J = 0$ and 1, beyond which (i.e., $J < 0$ and $J > 1$) the spin-wave excitations are unstable. In Fig. 3, the triplet excitation spectrum is schematically shown for several values of the coupling constant $J$. It is clear that in the region of $0 < J < 1$, there is a nonzero gap, and this gap collapses at the terminating points. In particular, the triplet spectrum is flat with the gap of 1 at $J = 1/2$. The flatness reflects the fact that at $J = 1/2$, $H_0$ of Eq. (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 [21]. They obtained the spectrum of soliton-like excitations with the minimum gap of 0.25 at $k = 0$ and $\pi$ and the maximum gap of 1 at $k = \pi/2$. Tonegawa and Harada’s numerical calculations [19] confirmed the nonzero gap at $J = 1/2$ and in the nearby region. They predicted that the gap collapses at $J \approx 0.3$, while Haldane [22], 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 the Heisenberg model at $J = 0$. In any case, the triplet spectrum of $\sqrt{2} \sin k$ from Eq. (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 four-fold, for example, is still unclear. The numerical calculations of the structure factor by Tonegawa and Harada [19] 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 two-fold in the region near $J = 1$ [20]. 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

$$D \equiv \langle s_{i-1} \cdot s_i \rangle - \langle s_i \cdot s_{i+1} \rangle = \langle s_1(r) \cdot s_2(r) \rangle - \langle s_2(r) \cdot s_1(r+1) \rangle,$$

(3.18)

where the expectation is with respect to the ground state of the system. By Eqs. (2.5) and (2.6), using the spin-wave ground state $|\Phi_0\rangle$ of Eq. (3.17), it is easy to show that in the spin-wave approximation, $D$ is nonzero 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.

IV. The Coupled-Cluster Approach

The CCM is widely recognized nowadays as providing one of the most universally applicable, 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 very appropriate and timely to apply the CCM to the dimerization problem. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13]. The CCM is widely recognized nowadays as providing one of the most universally applicable, most powerful and most accurate of all microscopic formulations of quantum many-body theory [13].
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

\[
S_1 = \sum_{r=1}^{N/2} S_r A_{20}^r, \tag{4.5a}
\]

\[
S_2 = \sum_{r,r'}^{N/2} \left[ S_{r,r'}^{(1)} A_{10}^r A_{30}^{r'} - \frac{1}{2!} S_{r,r'}^{(2)} A_{20}^r A_{20}^{r'} \right], \tag{4.5b}
\]

\[
S_3 = \sum_{r,r',r''}^{N/2} \left[ S_{r,r',r''}^{(1)} A_{10}^r A_{30}^{r'} A_{20}^{r''} - \frac{1}{3!} S_{r,r',r''}^{(2)} A_{20}^r A_{20}^{r'} A_{20}^{r''} \right], \tag{4.5c}
\]

etc. In Eq. (4.5b) and (4.5c) the primes on the summations imply exclusion of the terms with any pair of indices being equal. We notice the similarity between the spin-wave ground state in Eq. (4.5) and the CCM state \( |\Phi_0\rangle \) of Eq. (3.17) and the CCM state \( |\Psi_0\rangle \) of Eq. (4.1) if \( S \) is replaced by \( S_2 \). We also notice the similarity between the present CCM analysis and that of Ref. 15 where the spin-1 model state is given by the simple planar configuration with \( s_i^z = 0 \) for all sites.

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

\[
E_g = \langle D | e^{-S} H e^S | D \rangle; \tag{4.6}
\]

the correlation coefficients \( \{S_{r,r',\ldots}\} \) are determined by the coupled set of equations obtained by taking inner products of Eq. (4.2) with states constructed from the corresponding destruction operators, namely

\[
\langle D | A_{02}^r e^{-S} H e^S | D \rangle = 0, \quad \text{for all } r, \tag{4.7}
\]

for the one-body equation;

\[
\langle D | A_{01}^r A_{03}^{r'} e^{-S} H e^S | D \rangle = 0, \quad \text{for all } r, r'(\neq r) \tag{4.8}
\]

and

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

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 Eq. (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 Eq. (4.3) indeed terminates at the fourth-order term. In fact, the exact energy equation (4.6) can be straightforwardly derived as

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

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

\[
S_r = a, \quad S_{r_1,r_2}^{(i)} = S_{r_2,r_1}^{(i)} = b_{r}^{(i)}, \quad \text{with } i = 1, 2 \quad \text{and } r = r_2 - r_1. \tag{4.11}
\]

It is also obvious that \( b_{r}^{(i)} = b_{r}^{(i)} \).

The exact one-body equation of Eq. (4.7) can also be easily derived. It couples only to the two-body coefficients. Similarly, the two-body equations of Eqs. (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 Eq. (4.5a) will take the state out of this sector. Furthermore, if one assumes that the two sets of the two-body correlation coefficients are identical, namely

$$b_1^{(1)} = b_1^{(2)},$$  \hspace{1cm} (4.12)

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

$$E_n \equiv \frac{3}{8}(1 - 2J)b_1 - 1.$$  \hspace{1cm} (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 SUB$n$ 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 of Eq. (4.12) is indeed satisfied. After simplification, the two identical two-body equations are given by

$$\frac{1}{2} \sum_{\rho=\pm 1} (K_3 \delta_{\rho \rho} + K_2 b_\rho - 2K_1 b_{r+\rho} + K_1 \sum_{r' \neq 0} b_r b_{r+\rho-r'}) = 0, \quad r \neq 0$$  \hspace{1cm} (4.14)

and

$$K_1 \equiv 1 - 2J, \quad K_2 \equiv 4(1 - 2K_1 b_1), \quad K_3 \equiv K_1(1 + 4b_1^2) - 2(1 + 2J)b_1.$$  \hspace{1cm} (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 $b_r = 0 \ (|r| > 1)$. Eq. (4.14) then reduces to

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

with the physical solution

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

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

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

where the constants $k_1$ and $k_2$ are defined by

$$k_1 \equiv \frac{1}{K_2^2} (4K_1 K_2 + 8K_2^2 b_1 - 4K_1^2 X), \quad k_2 \equiv \frac{4K_1(K_1 - K_3)}{K_2^2},$$  \hspace{1cm} (4.19a)

and where $X$ is defined by

$$X \equiv \sum_{r=1}^{N/2} b_r b_{r+1}$$

$$= \frac{1}{2\pi} \int_{-\pi}^{\pi} dq \frac{1}{4K_1^2 \cos 2q}$$

$$\times \left( 2K_1 \cos 2q - K_2 + K_2 \sqrt{1 - k_1 \cos 2q + k_2 \cos^2 2q} \right)^2.$$  \hspace{1cm} (4.19b)

After obtaining $b_1$ as a function of $J$ from Eq. (4.17) or (4.18), the ground-state energy is then given by substituting $b_1$ into Eq. (4.13). These ground-state energies are shown in Fig. 2, together with the results of the spin-wave theory and of the numerical calculations [19,20] for comparison. Similar to
the spin-wave theory described in Sec. III, 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 $E_g/N = -0.4268$, slightly higher 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 Eq. (4.18). The corresponding energy values are $-0.5172$ and $-0.6977$ respectively. The CCM SUB2 terminating points have been argued to correspond to the phase transition critical points in the past [14,15]. It seems reasonable to consider this possibility again here. From Fig. 2, one sees that the extremely simple SUB2 scheme gives much better results for a wide range of the coupling constant $J$ than the spin-wave theory does, at least as far as the ground-state energy is concerned.

V. Discussion

In this paper, I have studied the dimerization problem by a microscopic approach, employing the proper set of composite operators of Parkinson [7]. Two approximations schemes, namely the spin-wave theory and a CCM analysis, have been applied to the 1D frustrated spin-$1/2$ 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 a variational trial wave function of the type of Eq. (4.1), similar to the calculation of Sachdev for the spin-$1/2$ Heisenberg model [23].

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-$1/2$ 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].

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Appendix

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

Notice that in the similarity transformation of Eq. (4.3), any quadratic term in the Hamiltonian of Eqs. (3.4)-(3.5) can be transformed as

$$e^{-S} A_{ij}^r A_{kl}^{r+1} e^S = (e^{-S} A_{ij}^r e^S)(e^{-S} A_{kl}^{r+1} e^S),$$  \hspace{1cm} (A.1)

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

$$e^{-S} A_{ij}^r e^S = A_{ij}^r + [A_{ij}^r, S] + \frac{1}{2!}[[A_{ij}^r, S], S] + \cdots.$$  \hspace{1cm} (A.2)

Since the correlation operator $S$ consists of only the creation operators $A_{10}, A_{30}$ and $A_{20}$, the expansion series of Eq. (A.2) terminates at most at the second-order by the pseudo-spin algebra of Eq. (2.4). Therefore the similarity-transformed Hamiltonian of Eq. (4.3) terminates at the fourth-order.
In the SUB1 scheme, one replaces $S \rightarrow S_1$, where $S_1$ is given by Eq. (4.5a). From Eq. (A.2), it is straightforward to derive the following SUB1 similarity transformations

\[
\begin{align*}
\tilde{A}_{r0}^n &= A_{r0}^n, \\
\tilde{A}_{nm}^r &= A_{nm}^r, \\
\tilde{A}_{r0}^n &= A_{r0}^n - S_r A_{20}, \\
\tilde{A}_{n2}^r &= A_{n2}^r + S_r A_{n0}, \\
\tilde{A}_{r2}^r &= A_{r2}^r + S_r (A_{00} - A_{22}) - S_{2r} A_{20},
\end{align*}
\]

(3.3)

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

\[
\tilde{A}_{ij}^r \equiv e^{-S_1 A_{ij}^r} e^{S_1},
\]

is used.

In the SUB2 scheme, one replaces $S \rightarrow S_1 + S_2$. One can firstly make the SUB1 similarity transform by Eq. (3.3), and then apply the following SUB2 similarity transform for each $A_{ij}^r$,

\[
\begin{align*}
\tilde{A}_{r0}^n &= A_{r0}^n, \\
\tilde{A}_{n1}^r &= A_{n1} + \sum_{r''}^r S_{r,r'} A_{n0}^r A_{50}, \\
\tilde{A}_{r2}^r &= A_{r2}^r - \sum_{r'}^r S_{r,r'} A_{r0}^r A_{20}, \\
\tilde{A}_{n1}^r &= A_{n1} + \sum_{r''}^r S_{r,r'} A_{r0}^r A_{10}, \\
\tilde{A}_{r0}^n &= A_{r0}^n - \sum_{r'}^r (S_{r,r'} A_{10} A_{30} + S_{r',r} A_{10}^r A_{30} - S_{r,r'} A_{20} A_{20}^r), \\
\tilde{A}_{01}^n &= A_{01}^n + \sum_{r''}^r (S_{r,r''} A_{10}^r A_{10} A_{30} - S_{r',r} A_{10}^r A_{10} A_{50}) \\
&\quad - \frac{1}{2} \sum_{r',r''}^r S_{r,r'} S_{r,r''} A_{10} A_{20} A_{30} - 2 S_{r,r'} S_{r',r} A_{10} A_{30} - S_{r,r'} A_{20} A_{20}^r A_{30}) \\
\tilde{A}_{02}^r &= A_{02}^r - \sum_{r'}^r (S_{r,r'} A_{20}^r A_{10} A_{10} A_{22} + S_{r',r} A_{10} A_{10} A_{12} + S_{r',r} A_{30} A_{12}) \\
&\quad - \frac{1}{2} \sum_{r',r''}^r S_{r,r'} A_{20}^r (2 S_{r',r} A_{20} A_{20}^r - S_{r',r'} A_{10} A_{30} - S_{r',r} A_{10} A_{30}) \\
&\quad + \frac{1}{2} \sum_{r',r''}^r (S_{r,r'} A_{10} A_{10} A_{30} A_{20} + S_{r',r} A_{10} A_{10} A_{20}) \\
\tilde{A}_{03}^r &= A_{03}^r + \sum_{r''}^r (S_{r,r''} A_{10}^r A_{10} A_{50} - S_{r',r} A_{10} A_{10} A_{13}) + S_{r',r} A_{20} A_{23}) \\
&\quad + \frac{1}{2} \sum_{r',r''}^r S_{r,r'} A_{10} (2 S_{r',r} A_{20} A_{20}^r - 2 S_{r',r} A_{30} A_{30} - S_{r',r'} A_{30} A_{30}) \\
&\quad - \frac{1}{2} \sum_{r',r''}^r (S_{r,r'} A_{10} A_{10} A_{10} A_{10} + S_{r',r} A_{10} A_{10} A_{10} A_{10}) \\
&\quad - S_{r',r} A_{10} A_{20} A_{20} A_{10},
\end{align*}
\]

(3.5)

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

\[
\tilde{A}_{ij}^r \equiv e^{-S_2 A_{ij}^r} e^{S_2}.
\]

(A.6)
In deriving Eqs. (A.5), I have used the fact that
\[ S_{r,r}^{(1)} = S_{r,r}^{(2)} = 0, \quad S_{r,r}^{(2)} = S_{r',r}^{(2)}. \]
(A.7)

After the similarity transformations of Eqs. (A.3) and (A.5), the CCM equations of Eqs. (4.6)-(4.9) can be derived by using the pseudo-spin algebra of Eq. (2.4) to move in each term all the creation operators \( A_{10}, A_{20}, A_{30} \) to the left, all the destruction operators \( A_{01}, A_{02}, A_{03} \) (also \( A_{nm}, n, m \neq 0 \)) to the right, and by using the fact that
\[ \langle D | A_{r}^{n_0} = A_{mn}^{n_0} | D \rangle = 0, \quad n \neq 0, \]  
(A.8)
and
\[ \langle D | A_{00}^{r} = \langle D, A_{00}^{r} | D \rangle = | D \rangle, \]  
(A.9)
for all \( r \).

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Figure Captions

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

Fig. 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 (short 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 Ref. [19,20] are also included (solid).

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