Rigorous results on a first-order phase transition in antiferromagnetic spin-1/2 coupled chains

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Some rigorous results are presented for a first-order quantum phase transition between the dimerized state and Haldane-type state (i.e., a state similar to the ground state of the one-dimensional spin-1 Heisenberg chain) in the spin-1/2 coupled chains with nearest-neighbor and next-nearest-neighbor Heisenberg interactions. Also presented is a class of exact excited states in both phases. A partial phase diagram of the general spin-1/2 coupled chains is discussed.

Spin-1/2 coupled chains have attracted attention for the last few years[1-3] partly because of their relevance to such materials as $(VO)_2P_2O_7$ (Ref. 4) and $Sr_2Cu_4O_6$,[5] and partly because of their intrinsic theoretical interest. For the isotropic coupled chains with only nearest-neighbor couplings, current consensus is that the system has a nonzero energy gap separating the ground and low-lying excited states, and the spin-spin correlation length of the ground state is finite.[1-3] No long-range order has been reported.

It is well known that, for one-dimensional spin chain systems, the spontaneous dimerized phase (the perfect dimer state being defined by every two adjacent atoms forming a spin-singlet pair in the spin-1/2 or spin-1 chains)[6] and Haldane-type phase (spin-1 chain)[7] are both possible. It is also well known that these two phases both exhibit nonzero gap and finite correlation length. The main difference between the two phases lies in the fact that the Haldane-type state is nondegenerate and translationally invariant with a hidden string order,[8,9] while the spontaneous dimerized state of a single chain is doubly degenerate with translational symmetry broken. For the system of spin-1/2 coupled chains, it is therefore natural to consider the possible existence of these two phases in despite of the dimerized phase being nonspontaneous and to study corresponding phase transitions if the two phases do exist. In fact, spin-1/2 coupled chains as a subject of research have a longer history.[10,11] The main interest there was to construct effective one-dimensional chains with larger spin moments from spin-1/2 coupled chains. Many interesting results were produced.[10,11] In particular, it was found that spin-1/2 coupled chains can have the same ground state as that of the one-dimensional spin-1 Heisenberg chain.

Because of their relevance to the experiments, the general spin-1/2 coupled chains are important models in their own right. In this article, we study spin-1/2 coupled chains which clearly exhibit both the dimerized phase and Haldane-type phase when the coupling parameters vary. Specifically, we consider spin-1/2 coupled chains with nearest-neighbor and next-nearest-neighbor Heisenberg interactions. The spin system consists of N pairs of $S=1/2$ atoms, with interactions described by the following Hamiltonian:

$$H = \sum_{r=1}^{N} [JS_1^r \cdot S_2^r + J'(S_1^r \cdot S_1^{r+1} + S_2^r \cdot S_2^{r+1})$$
$$+ J''(S_1^r \cdot S_2^{r+1} + S_2^r \cdot S_1^{r+1})], \qquad (1)$$

where the rung index r denotes each pair of spins with the spins on the top chain denoted as S_1^r and the spins on the bottom chain denoted as S_2^r; J is the coupling constant across each rung, J' is the nearest-neighbor coupling constant along each chain, and J'' is the next-nearest-neighbor (diagonal) coupling constant. We mainly discuss the antiferromagnetic region $(J, J', J'' \geq 0)$. Periodic boundary conditions are taken in this article. The Hamiltonian of Eq. (1) at $J=0$ is the so-called composite spin model.[11]

We first consider a single rung with a pair of spin-1/2 atoms only. The rung has four states: the singlet state with zero total spin, namely $|0\rangle = (1/\sqrt{2})(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$ in the usual spin-1/2 notation; and the triplet states with total spin equal to 1, namely $|1\rangle = |\uparrow\uparrow\rangle$, $|2\rangle = (1/\sqrt{2})(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$, and $|3\rangle = |\downarrow\downarrow\rangle$, respectively. In a matrix representation,[12,13] these four states are denoted by four column matrices, respectively, and the single spin operators (S_μ^z and $S_\mu^\pm$ with $\mu = 1,2$) can then be written as 4×4 matrices. We then introduce the so-called composite operator A_{nm} $(n,m = 0,1,2,3)$, which is a 4×4 matrix with a single nonzero entry, namely $\langle n'|A_{nm}|m'\rangle = \delta_{nn'}\delta_{mm'}$. The single spin operators can then be written as linear summations of composite operators A_{nm}. The meaning and bosonization of these composite operators have been discussed in detail in Ref. 13. Briefly, A_{00} $(= 1/4 - S_1 \cdot S_2)$ is the singlet projection operator, A_{0n} and A_{n0} $(n = 1,2,3)$ make transitions between the singlet and triplet states, and A_{nm} $(n,m = 1,2,3)$ make transitions among the triplet states. A_{nm} obey pseudospin algebra, $[A_{nm}, A_{kl}] = A_{nl}\delta_{mk} - A_{km}\delta_{ln}$.

In terms of these composite operators, A_{nm}^r, with the index r denoting each rung in the coupled chains, the Hamiltonian of Eq. (1) can be rewritten as

$$H = \sum_{r=1}^{N} \left[J\left(\frac{1}{4} - A_{00}^r\right) + \frac{1}{2}(J' - J'')H_{r,r+1} \right.$$
$$\left. + \frac{1}{2}(J' + J'')H_{r,r+1}' \right], \qquad (2)$$

where

$$H_{r,r+1} \equiv (A_{02}^r + A_{20}^r)(A_{20}^{r+1} + A_{02}^{r+1})$$

$$+ (A_{01}^r - A_{30}^r)(A_{10}^{r+1} - A_{03}^{r+1})$$

$$+ (A_{10}^r - A_{03}^r)(A_{01}^{r+1} - A_{30}^{r+1})$$

and

$$H'_{r,r+1} \equiv (A_{11}^r - A_{33}^r)(A_{11}^{r+1} - A_{33}^{r+1})$$

$$+ (A_{21}^r + A_{32}^r)(A_{12}^{r+1} + A_{23}^{r+1})$$

$$+ (A_{12}^r + A_{23}^r)(A_{21}^{r+1} + A_{32}^{r+1}).$$

It is interesting to compare this Hamiltonian with that of the one-dimensional spin-1/2 frustrated Heisenberg models [i.e., Eqs. (3.4)–(3.5) of Ref. 13]. As we can see, by using the composite operators, the algebra of the spin-1/2 coupled chains becomes quite transparent. It is trivial to note that when $J' = 0$ and $J'' = 0$, the ground state of the system is simply the perfect dimer state $|D\rangle$ which is given by the product of singlet states of all N rungs, namely $|D\rangle = \Pi_{r=1}^N |0\rangle_r$. More significantly, we notice that the Hamiltonian of Eq. (2) consists of three parts, the first part consisting only of the operators A_{00}^r which is nonzero only when acting on the singlet state of the rth rung, the second part consisting only of the operators $H_{r,r+1}$ which makes transition between the singlet and triplet states, and finally the third part consisting only of the operators $H'_{r,r+1}$ which is nonzero only when acting on a state in which both the rth and $(r+1)$th rungs are in the triplet states.

In fact, it is easy to prove that $H'_{r,r+1}$ is similar to the usual spin-1 Heisenberg interaction,[11] namely

$$H'_{r,r+1} = \mathbf{P}_r \cdot \mathbf{P}_{r+1}, \qquad |\mathbf{P}| = 1, \qquad (3)$$

where we define operator $\mathbf{P}_r$ by

$$P_r^z = A_{11}^r - A_{33}^r, \qquad P_r^+ = \sqrt{2}(A_{12}^r + A_{23}^r),$$

$$P_r^- = \sqrt{2}(A_{21}^r + A_{32}^r), \qquad (4)$$

with the usual SU(2) algebra, $[P^+, P^-] = 2P^z$ and $[P^z, P^\pm] = \pm P^\pm$.

At $J' = J''$, the second part of H disappears. The Hamiltonian is then reduced to

$$H_0 = \sum_{r=1}^N (-A_{00}^r + j' \mathbf{P}_r \cdot \mathbf{P}_{r+1}), \qquad J'' = J', \qquad (5)$$

where, for convenience, we have set $J'/J = j'$ and $J = 1$, and where we have ignored the constant term, $N/4$. We notice that operators A_{00}^r and $\mathbf{P}_r \cdot \mathbf{P}_{r+1}$ commute with one another. We have therefore arrived at a Hamiltonian consisting of two decoupled parts. The first part, $-\Sigma_r A_{00}^r$, is trivial, with no interaction between different rungs and with a gap value of 1 between the singlet and triplet states. The second part, $\Sigma_r \mathbf{P}_r \cdot \mathbf{P}_{r+1}$, is similar to the spin-1 Heisenberg chain with each rung in the coupled chains corresponding to each site in the spin-1 chain (the only difference is appearance of a new type of excitation to be discussed later). The spin-1 Heisenberg model has been the focus of intensive investigations in the last decade and its physical properties are now well un-

derstood. The state-of-the-art calculations were recently performed by the density-matrix renormalization-group technique[9] and extremely accurate results have been obtained. In particular, the long-range string order, $g(\infty) = 0.374\,325\,096(2)$, is obtained by calculation of the string correlation function $g(r)$ which is defined as

$$g(r) = \left\langle P_0^z \left(\prod_{r'=1}^{r-1} e^{i\pi P_{r'}^z} \right) P_r^z \right\rangle. \qquad (6)$$

Since the two parts of H_0 are decoupled and since all excited states of both parts have nonzero energy gaps (to be discussed later), its ground state is either that of the first part, or that of the second part, depending on the value of j'. The exact ground-state energy of H_0 is therefore given by

$$\frac{E_g}{N} = \begin{cases} -1 & \text{for } j' < j_t, \\ -\dfrac{j'}{j_t} & \text{for } j' > j_t, \end{cases} \qquad (7)$$

where j_t is the transition coupling constant, defined by $j_t \equiv 1/e_0 = 0.713\,529\,353\,310(1)$ with e_0 the ground-state energy per site of the spin-1 Heisenberg chain.[9] Clearly, the phase transition at $j' = j_t$ is a first-order one because the first-order energy derivative with respect to j' is discontinuous at the transition point.

We next discuss excitation states in two phases. For the dimerized phase ($j' < j_t$), the excitations states can be easily obtained. For convenience, we define n singlet-to-triplet spin-flip (STSF) state as a configuration in which n rungs are in the triplet states, while the remaining $(N-n)$ rungs are in the singlet state as in the dimer ground state. If each flip is separated from each another in a n-STSF configuration, the energy gap (i.e., total excitation energy minus total ground-state energy) of the n-STSF state is simply $\Delta E_d(n) = n$ with 3^n-fold degeneracy. However, if n flips form a contiguous cluster, the excitation gap can be much lower, with gap values given by $\Delta E_d(n) = n + E(n)j'$, where $E(n)$ are the eigenvalues of the open-ended n spin-1 Heisenberg chain. As well known, the ground state of the open-ended spin-1 chain is singlet when n is even and triplet when n is odd; the energy difference between the singlet (triplet) ground state and next triplet (singlet) excited state for a given n decreases exponentially to zero in the large-n limit and the exact ground-state energy was obtained for up to $n = 14$ in Ref. 14. For the one-flip state, $E(1) = 0$, therefore, $\Delta E_d(1) = 1$ with triplet degeneracy. For the contiguous 2-STSF cluster, $\Delta E_d(2) = 2(1-j')$, $2-j'$, and $2+j'$ for the singlet, triplet, and quintuplet states, respectively, with a minimum gap of about 0.573 when $j' \to j_t$. (For 3- and 4-STSF clusters, the minimum gaps as $j' \to j_t$ are about 0.895 and 0.685, respectively). If we consider a single STSF state as a (soliton) particle with spin momentum equal to one, it is obvious these particles attract to one another at low values of their total spin quantum numbers. For large n ($n > 14$), the lowest $E(n) \equiv [-e_0(n-1) + e']$, where e' represents the residual boundary effect [its magnitude is expected to be much smaller than e_0 (Ref. 14], the minimum gap of the n-STSF cluster state is then given by

$$\Delta E_d(n) = n + [-e_0(n-1) + e']j', \qquad n \text{ large.} \qquad (8)$$

When $j' \to j_t$, $\Delta E_d(n) \to 1 + e' j_t$, a number independent of n. Since $|e'| \ll 1$, $\Delta E_d(n) \approx 1$ for large n. When n approaches N, it is easy to see that the dimer ground state is unstable against the large n-STSF cluster state when $j' > j_t$, as expected.

The excitations in the Haldane-type phase have been discussed for the Hamiltonian of Eq. (1) at $J = 0$ in Ref. 11. Here, we take advantage of the recent accurate results for the spin-1 Heisenberg chain,[9] and focus on the excitations of Eq. (5) for $j' > j_t$. As we know, the low-lying excited state of the spin-1 chain is given by the one-magnon state with momentum $q = \pi$, and with the well-known Haldane gap $\Delta = 0.41050(2)$.[9] Multimagnon states have larger gaps.[9,15] We consider here a new type of excitations where the spin-1 chain is embedded with singlet rungs.[11] Again, for convenience, we define n triplet-to-singlet spin-flip (TSSF) configuration as a state in which there are n singlet rungs and the remaining $(N-n)$ rungs are in their Haldane-ground-state configuration. We consider a single TSSF configuration, which corresponds to the ground state of the $(N-1)$ spin-1 Heisenberg chain with two open ends enclosing the singlet rung. Its total energy can be written as $E_h(1) = E(N-1)j' - 1$, where, as before, $E(N-1) = -e_0(N-2) + e'$. Hence, $\Delta E_h(1) = (2e_0 + e')j' - 1$. Since $|e'| \ll 1$, we obtain $\Delta E_h(1) \approx 1$ as $j' \to j_t$, a value larger than $0.4105 j_t \approx 0.293$ of the one-magnon Haldane gap. Furthermore, from the numerical calculations of the open-ended spin-1 chains,[9] we note that each of the two ends enclosing the singlet rung in the 1-TSSF state has an effective $S = 1/2$ spin with exponential decay of the local spin moment away from the ends, and with decay length $\xi \approx 6.03(1)$ (which is also the usual spin-spin correlation length). Therefore, the single TSSF configuration can be considered as particlelike (soliton) state with a size of about 14 lattice spacings (the core occupying two lattice spacings). We also notice that the degeneracy of the single TSSF state is four, while the one-magnon state of the spin-1 chain is triplet. Similar to the n-STSF cluster state in the dimer phase, the n-TSSF state has lowest energy when n flips are contiguous, with the minimum gap

$$\Delta E_h(n) = [(n+1)e_0 + e']j' - n. \tag{9}$$

We notice that when $j' \to j_t$, all clusters have the same minimum gap of $(1 + e' j_t) \approx 1$, a number identical to that of the (large) n-STSF cluster state of the dimer phase when approaching the transition point. When $n \to N$, the Haldane-type phase is unstable against formation of the large n-TSSF cluster state when $j' < j_t$, as expected.

The above analysis for the Hamiltonian of Eq. (1) at $J' = J''$ represents our main results in this article. Before we discuss the implication of these rigorous results to the general phase diagram, we should point out that the Hamiltonian at $J' = J''$ is an interesting model on its own right; it represents, among other things, the Heisenberg model on a tetrahedronic chain (i.e., every two nearest-neighbor pairs of atoms form a tetrahedron) with only nearest-neighbor couplings.

We now discuss the phase diagram of Eq. (1) on a spherical surface of unit radius with J, J', J'' being the three axes, as shown in Fig. 1(a). We firstly discuss the region near the
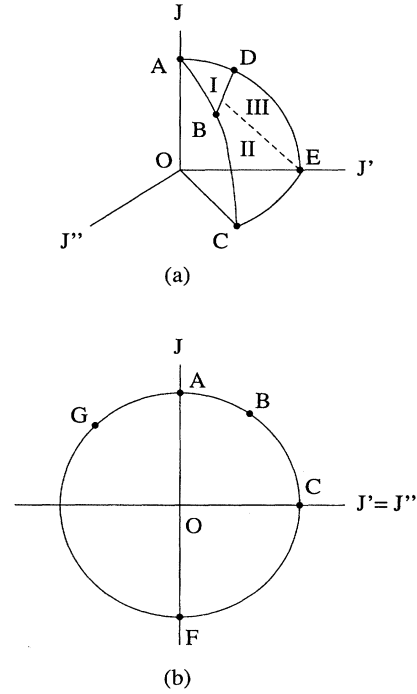

(a)

(b)

FIG. 1. A partial phase diagram for the spin-1/2 coupled chains of Eq. (1). (a) Region I (ABD) is the dimerized phase, II (BCE) is the Haldane-type phase, III (BED) may be some new spin-liquid or still the Haldane-type phase. ABC is given by $J'' = J'$ with the exact transition point at B, D is obtained by the valence-bond spin-wave theory (Ref. 16), and the dashed line is based on the exact results of BC and numerical results from Ref. 11. (b) The complete phase diagram when $J'' = J'$, with GAB denoted the dimer phase, BCF Haldane-type phase, and FG ferromagnetic phase.

north pole A. We have applied the (dimerized) valence-bond spin-wave theory developed by us[13] via bosonization for the composite operators A_{nm}^r in Eq. (2) with $J'' = 0$, starting from the perfect dimer state $|D\rangle$ which is the exact ground state at $J' = 0$.[16] Good results for the ground and spin-wave excited states are obtained for a small nonzero region of J'/J, but the theory breaks down at $J'/J = 1/2$ [denoted as D in Fig. 1(a)] and the dimerization order (similar to that of the one-dimensional spin-1/2 frustrated chain[13]) is nonzero in the region $0 \leq J'/J < 1/2$. This breakdown seems to suggest a transition from the dimerized phase to other phase(s). Combining with the exact dimer state along AB where B is the exact transition point between the dimer and Haldane-type phases, we draw a phase boundary BD enclosing dimerized region I as shown in Fig. 1(a).

Secondly, we discuss the phase diagram near the Haldane-type phase. Since the Hamiltonian of Eq. (1) at E ($J = J'' = 0$) reduces to the two decoupled spin-1/2 Heisenberg chains which is exactly known to be critical and gapless, E point provides another exact reference for the phase diagram. Combining with the rigorous results for $J'' = J'$ along BC presented above, the numerical results along CE from Ref. 11 which indicates CE is Haldane-type, and the fact that the Haldane-type phase have a *nonzero* gap, we enclose this Haldane-type phase II by a dashed line joining

from BD to E. Clearly, the boundary (dashed line) between the Haldane-type phase (II) and a third possible phase (III) is uncertain at this moment. We consider the three possible scenarios for III: (a) The valence-bond spin-wave theory is unreliable and the dimerized phase persists from I to III; (b) The valence-bond spin-wave theory is qualitatively correct and the whole region enclosed by $BCED$ (i.e., II plus III) is the Haldane-type phase, including the isotropic model with $J=J'$ and $J''=0$; (c) A new phase (e.g., a new spin liquid) appears in III which also has a nonzero gap but without any long-range order (dimerized or string order). We consider the first scenario most unlikely because the (dimerized) valence-bond spin-wave theory is expected to be at least qualitatively correct if the system is dimerized.[16] Results from the numerical calculations[1–3] seem to suggest the third scenario. However, since there is not any report on

calculations of the string order, we cannot rule out the second scenario. In any case, it is interesting to apply powerful numerical techniques such as the density-matrix renormalization-group[3] to calculate the string order particularly in region III, using Eqs. (4) and (6).

Finally, we should also point out that there are other regions in Fig. 1(a), where exact results can be obtained. For example, we show in Fig. 1(b) the complete phase diagram for the Hamiltonian of Eq. (1) at $J''=J'$, where GAB denotes the dimer phase region, BCF the Haldane-type phase region, and FG ferromagnetic region. All of the three critical points, G, B, and F, which separate the three phases, are first-order ones.

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