Frustration-induced phase transitions in the spin-S orthogonal-dimer chain

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We investigate quantum phase transitions in a frustrated orthogonal-dimer chain with an arbitrary spin $S \geqslant 1/2$. When the ratio of the competing exchange couplings is varied, first-order phase transitions occur $2S$ times among distinct spin-gap phases. The introduction of single-ion anisotropy further enriches the phase diagram. The phase transitions described by the present model possess most of the essential properties inherent in frustrated quantum spin systems.

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I. INTRODUCTION

Geometrical frustration in strongly correlated electron systems has attracted much current interest. A remarkable example of such a material is the two-dimensional spin-gap compound $SrCu_2(BO_3)_2$,[1] in which the characteristic orthogonal-dimer structure of the Cu^{2+} ions stabilizes the spin-singlet ground state.[2–4] Remarkably, strong frustration induces an anomalous first-order phase transition in addition to plateau-formation in the magnetization process.[5] More recently, another orthogonal-dimer compound Nd_2BaZnO_5 (Ref. 6) was synthesized, where higher-spin and orbital moments ($J=9/2$) show the antiferromagnetic order at the critical temperature $T_N=2.4$ K, making such higher-spin systems more interesting.

One of the most prototypical phenomena inherent in the frustrated systems is the *first*-order transition, discussed in some frustrated systems such as ladders[7,8] and orthogonal-dimer systems.[2–4,9–11] Although a systematic treatment of a system with an arbitrary spin S is not easy, it is highly desirable to clarify how such first-order transitions are induced by frustration in order to understand the essential properties common to such frustrated quantum spin systems.

In this paper, we investigate a remarkable one-dimensional (1D) spin-S orthogonal-dimer model, which possesses most of the essential properties of first-order phase transitions in this class of frustrated spin systems.[9–11] By exploiting the nonlinear sigma model (NLσM) approach as well as the exact diagonalization and the series expansion, we find distinct $(2S+1)$ spin-gap phases in the spin-S chain, which are separated by first-order quantum phase transitions. We also discuss the effect of single-ion anisotropy, which plays an important role in higher-spin systems.[12] We demonstrate that a higher-spin generalization of the model results in the remarkably rich phase diagram, which realizes the idea of valence-bond solid (VBS) (Ref. 13) in *a sequence of first-order phase transitions*.

II. MODEL HAMILTONIAN

Let us consider the 1D quantum spin system with the orthogonal-dimer structure[10,11] shown schematically in Fig. 1. The corresponding Hamiltonian reads

$$H = J\sum_j \left(\mathbf{S}_{2,j}\cdot\mathbf{S}_{4,j}+\mathbf{S}_{3,j}\cdot\mathbf{S}_{1,j+1}\right) + J'\sum_j \left(\mathbf{S}_{2,j}+\mathbf{S}_{4,j}\right)\cdot\left(\mathbf{S}_{1,j}+\mathbf{S}_{3,j}\right), \tag{1}$$

where $\mathbf{S}_{i,j}$ is the ith spin operator in the jth plaquette, and $J(J')$ is the antiferromagnetic exchange coupling. The $S=1/2$ model has been studied well.[10,11] Below, we focus on a higher-spin generalization of the system.

We first exploit the NLσM technique to clarify the topological nature of the system.[14–18] Note that the Hamiltonian has the remarkable relation $[H,\mathbf{S}_{2,j}+\mathbf{S}_{4,j}]=0$. Therefore, by introducing the composite spin T_j defined as $\mathbf{T}_j=\mathbf{S}_{2,j}+\mathbf{S}_{4,j}$, we obtain the effective mixed-spin Hamiltonian as

$$H=\sum_j \left[J'\mathbf{S}_{1j}\cdot\mathbf{T}_j+J'\mathbf{T}_j\cdot\mathbf{S}_{3j}+J\mathbf{S}_{3j}\cdot\mathbf{S}_{1j+1}\right] + \frac{J}{2}\sum_j T_j(T_j+1)-\frac{1}{4}JS(S+1)N, \tag{2}$$

where N is the total number of sites. Then the Hilbert space of Hamiltonian (1) can be classified into each subspace specified by $[S;\{T_j\}]$. The singlet ground state as well as relevant low-energy excitations are in the space with uniform $T_j(=T)$. In particular, for a given $T\neq 0$ we can describe low-energy properties by the NLσM ($T=0$ gives a trivial system with decoupled dimers). Introducing three kinds of the fluctuation fields,[16–18] we obtain the Euclidean Lagrangian $\mathcal{L}$, with the effective field $\boldsymbol{\phi}$ as

$$\mathcal{L}=\frac{1}{2g}\left(v_s\boldsymbol{\phi}'^2+\frac{1}{v_s}\dot{\boldsymbol{\phi}}^2\right)-\frac{i\theta}{4\pi}\boldsymbol{\phi}\cdot(\boldsymbol{\phi}'\times\dot{\boldsymbol{\phi}}), \tag{3}$$

where

$$\theta=2\pi T, \tag{4}$$

$$g=2AB/T, \tag{5}$$

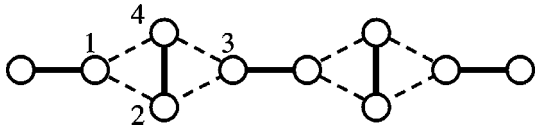

FIG. 1. Orthogonal-dimer spin chain: the dimer bonds shown by solid lines have the characteristic orthogonal structure.

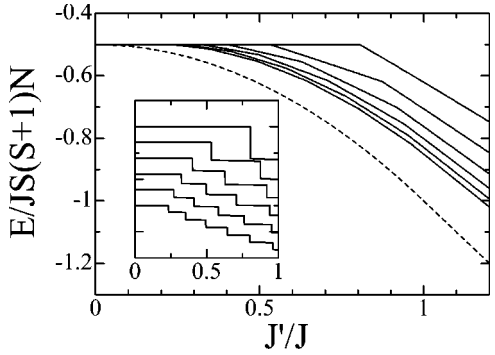

FIG. 2. Ground-state energy as a function of J'/J obtained by exact diagonalization for eight sites. From up to down, $S=1/2$, 1, 3/2, 2, 5/2, and 3. The broken line is the energy for the classical limit $S\rightarrow\infty$. The inset shows the derivative of the ground-state energy, in which the origin for each curve is shifted for convenience.

$$v_s = 6J'StB/A \tag{6}$$

with $A=(2t+j_0)^{1/2}$, $B=[4j_0t^2+2(1-2j_0)t+j_0]^{-1/2}$, $j_0=J'/J$, and $t=S/T$. Note that the topological angle θ is zero (mod 2π) for any choice of T, and the system is always gapped. Therefore, in case the phase transition between the distinct subspaces $[S;\{T_j\}]$ occurs, it should be accompanied by the discontinuity in the parameters g and v_s. This suggests that the possible quantum phase transition should be of *first* order.

III. FIRST-ORDER TRANSITIONS

To clarify this point, we numerically diagonalize Hamiltonian (1) for a small cluster with periodic boundary conditions. The ground-state energy is shown in Fig. 2. It is found that the cusps appear $2S$ times in the energy diagram for the spin-S case, implying that the *first*-order quantum phase transitions indeed occur $2S$ times. Also shown is the energy in the classical limit $S\rightarrow\infty$, which is given as $E/JS^2N=-\frac{1}{2}[1+(J'/J)^2]$, and $-J'/J$ for the helical phase ($0<J'/J<1$) and the antiferromagnetically ordered phase ($1<J'/J$). As S increases, the profile of the energy diagram

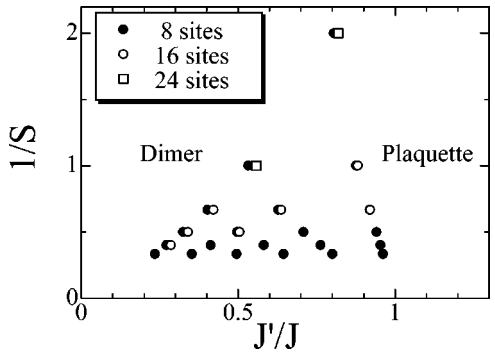

FIG. 3. Ground-state phase diagram for the orthogonal-dimer spin chain with a generic spin S. The closed circles, open circles, and open squares indicate the phase boundaries determined by the spin chain ($N=8$,16, and 24) with periodic boundary conditions.

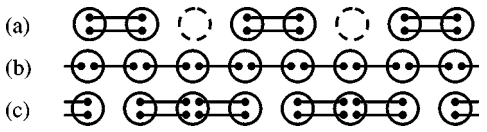

FIG. 4. VBS picture of the phases for the $S=1$ model.

gradually approaches the classical one, although the $2S$ transitions should exist for any finite S. By examining several systems with different cluster sizes, we determine the phase diagram rather precisely, as shown in Fig. 3. In this way, the present model is the remarkable spin-S microscopic model, which clearly determines a sequence of the phase transitions triggered by frustration. Moreover, the *first*-order transitions found here are contrasted to the *second*-order ones known for the ordinary spin-S Heisenberg chain with bond alternation.[15]

We now clarify the nature of each spin-gap phase by taking the $S=1$ model, which contains three distinct spin-gap phases, as an example. For this purpose, a VBS description[13] of the ground state is useful, where the topological nature is specified by a combination of singlet bonds between the decomposed $S=1/2$ spins, as shown in Fig. 4. Recall that in the small (large) J'/J region, the composite spin $T=0(2)$ on each diagonal bond is realized. Hence the dimer (plaquette) phase characterized by Fig. 4(a) [4(c)] is stabilized there. Note that in the dimer phase there is no correlation among singlet dimer bonds due to the orthogonal-dimer structure, in contrast to the plaquette phase in which a weak correlation still exists among singlet plaquettes. On the other hand, in the intermediate phase, strong frustration induces the spin $T=1$ on each diagonal bond, resulting in the singlet phase characterized by Fig. 4(b). This may be regarded as the *frustration-induced Haldane phase*.

To confirm the above predictions based on a VBS analysis, we perform a numerical diagonalization of small clusters in the corresponding subspaces. In Fig. 5(a), the flat line is the energy for the exact dimer state. The energy for the Haldane phase ($N=8$,16, and 24) obtained by the exact diagonalization in the subspace $[\{T_j=1\}]$ is shown by the solid line with closed circles, open circles, and open squares, respectively. We also show the ground-state energy of the original orthogonal-dimer spin chain ($N=16$) by the bold line with closed triangles. As is clearly seen, the increase of J'/J triggers a first-order quantum phase transition from the dimer phase ($T_j=0$) to the Haldane phase ($T_j=1$) at the critical value $(J'/J)_c\sim 0.56$. Shown in Fig. 5(b) is the energy

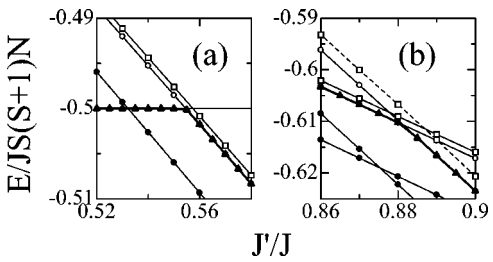

FIG. 5. The energy for the $S=1$ orthogonal-dimer spin chain in the restricted subspace.

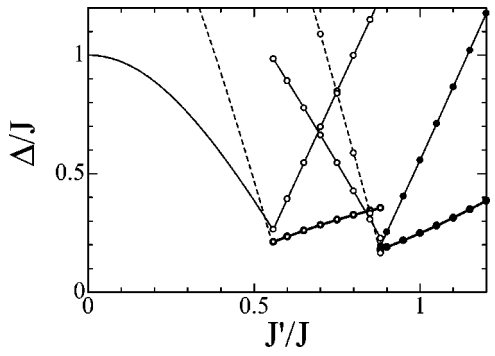

FIG. 6. Various excitations in the $S=1$ orthogonal-dimer spin chain. The bold lines indicate a dispersive magnetic excitation, while the solid (broken) lines indicate dispersionless magnetic (nonmagnetic) excitations.

around the other first-order critical point between the Haldane phase ($T_j=1$) and the plaquette phase ($T_j=2$). In the plaquette phase, we have used the series expansion[19] by choosing an isolated plaquette as the unperturbed system, and regarded the interaction between plaquettes as a perturbation. The ground-state energy, calculated up to the ninth order, is shown in Fig. 5 by the broken line from which we find the critical point $(J'/J)_c \sim 0.88$ between the Haldane and plaquette phases. It is thus concluded that first-order quantum phase transitions occur among three singlet phases specified by the distinct subspaces $[S;\{T_j\}](T_j=0,1,$ and $2)$, as predicted by the NLσM approach.

Keeping this in mind, we now consider excitations in the $S=1$ orthogonal-dimer spin chain. In the dimer phase ($J'/J<0.56$), the ground state is given by the product of isolated dimers $[\{T_j=0\}]$, which allows us to estimate the excitation gap exactly from the finite-size calculation. The lowest magnetic excitation is described by a defect in the uniform spin alignment, i.e., it is given by the lowest triplet state in the space of $[1, 0, 0, 0, \ldots]$. It is also found that, in the vicinity of the critical point ($J'/J \sim 0.56$), a nonmagnetic excitation belonging to the space $[1,1, 0, 0, 0, \ldots]$ can be the lowest one, as shown by the broken line. In the Haldane phase ($0.56<J'/J<0.88$), we estimate several kinds of spin gaps by an exact diagonalization for 24 sites. The Haldane gap, expected naively, is the lowest away from the critical points, shown by the bold line in Fig. 6. There are other dispersionless excitations, which can be described by a defect in the spin alignment such as $[0,1,1,1, \ldots]$, and $[2,1,1,1, \ldots]$. These excitations are bound into another nonmagnetic state, which is the lowest excitation in the Haldane phase close to the plaquette phase ($J'/J \sim 0.88$). In the plaquette phase ($0.88<J'/J$), the series expansion is more efficient to obtain dispersive and dispersionless excitations. The results, computed up to the seventh and ninth orders are shown by the closed circles in Fig. 6. In this way, several different excitations become almost degenerate around the first-order phase transition points, reflecting strong frustration.

IV. EFFECT OF SINGLE-ION ANISOTROPY

Finally we discuss the effect of single-ion anisotropy[12] with the Hamiltonian $H_D=D\Sigma(S^z_{i,j})^2$. The composite spin

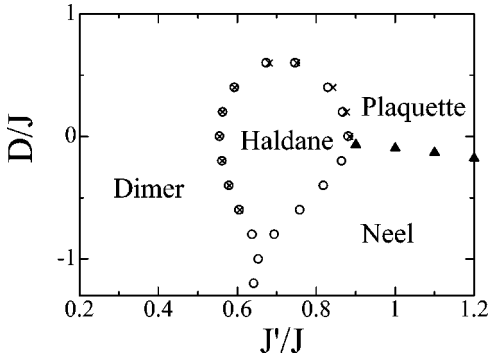

FIG. 7. Phase diagram for the anisotropic $S=1$ chain with the orthogonal-dimer structure.

T_j is not a good quantum number in the presence of the D term. The frustration-induced intermediate phases, however, are stable against the introduction of small anisotropy. In fact, cusps still exist in the energy diagram for the spin $S=1$ chain ($N=16$), from which we determine the phase boundaries shown by the open circles in Fig. 7. The Haldane phase gradually shrinks with the increase of $D(>0)$, and finally disappears. We note that around $D \gtrsim 0.8$ the dimer and plaquette phases indeed merge into a single phase. In order to clearly distinguish the Haldane phase and the dimer (or plaquette) phase, we make use of the symmetry of the space inversion P and the spin reversal τ. Under twisted boundary conditions[20] $(S^x_{1,N/4+1}=-S^x_{1,1}, S^y_{1,N/4+1}=-S^y_{1,1}, S^z_{1,N/4+1}=S^z_{1,1})$, the dimer or plaquette state has the eigenvalue $P=\tau=1$, while the Haldane state has the value $P=\tau=-1$. Therefore, we can distinguish these phases by diagonalizing the Hamiltonian with twisted boundary in the restricted space specified by $P=\tau=\pm 1$. The results for $D=0.4$ are shown in Fig. 8. It is found that the two lowest-energy levels intersect each other twice when the ratio of the exchange coupling J'/J is varied. This implies that the quantum phase transitions between phases with distinct symmetry ($P=\tau=\pm 1$) occur twice, being consistent with the above predictions based on the VBS analysis. By scaling the critical values as $(J'/J)_c(N)=(J'/J)_c(\infty)+aN^{-2}$, we estimate the

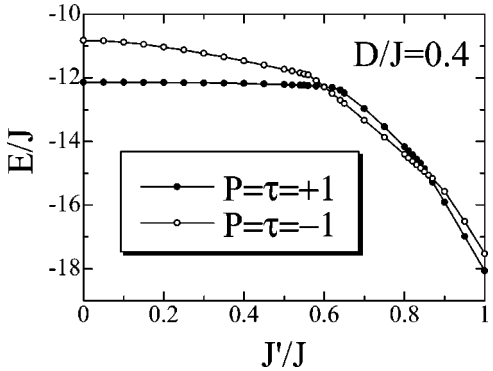

FIG. 8. Two lowest energies with twisted boundary conditions for $N=16$ and $D=0.4$. The energy of the Haldane state ($P=\tau=-1$) and the dimer or the plaquette state ($P=\tau=1$) are shown by open and closed circles.

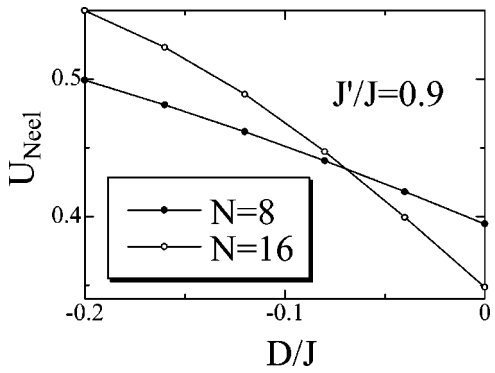

FIG. 9. The Binder parameter U_{Neel} for $J'/J=0.9$. The closed and open circles ($N=8$ and 16) are calculated with periodic boundary conditions.

phase boundaries shown by the crosses in Fig. 7, which agree well with those determined from the original plaquette chain ($N=16$) shown by open circles. In contrast, when D is negative, the antiferromagnetic correlation is enhanced, and the system is driven to the Néel-ordered phase. To characterize this transition, we check the behavior of the Binder parameter,[21]

$$U_{\mathrm{Neel}}=1-\frac{\langle O^4\rangle}{3\langle O^2\rangle^2}, \tag{7}$$

where O is the order parameter defined as

$$O=\sum_{i,j}(-1)^{i+j}S^z_{i,j}. \tag{8}$$

In Fig. 9, the Binder parameters for $N=8$ and 16 are shown as a function of D with $J'/J=0.9$. Since the Binder parameter stays invariant with the change of N at the transition point, we can determine the critical value $(D/J)_c\sim-0.07$. Consequently, we end up with the phase diagram for the spin $S=1$ chain with anisotropy, as shown in Fig. 7.

V. SUMMARY

We have studied the spin-S quantum spin chain with the orthogonal-dimer structure. The obtained phase diagram has a rich structure with various type of spin-gap phases which are separated by first-order phase transitions inherent in fully frustrated systems. Although we have given detailed accounts for the $S=1$ case, it is straightforward to generalize the discussions to an arbitrary-spin case. For example, four distinct spin-gap phases for $S=3/2$ in Fig. 3 are completely classified by the VBS states shown in Fig. 10.

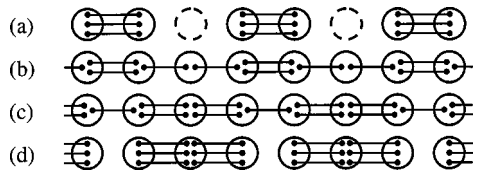

FIG. 10. VBS picture of the phases for the $S=3/2$ model.

Finally we emphasize again that the first-order phase transitions in the present model are triggered by strong frustration, which may capture most of essential properties common to this class of fully frustrated systems. To demonstrate this point clearly, it is desirable to study whether the distinct spin-gap phases obtained here for the 1D orthogonal-dimer system can persist even in the 2D system, when the interchain couplings are introduced. For this purpose, we have performed exact diagonalization studies for the 4×4 $S=1$ orthogonal-dimer system. We have found that the first-order transition points are continuously changed with the increase of the interchain couplings and thus the frustration-induced Haldane phase persists even in the 2D $S=1$ orthogonal-dimer system. Therefore, we believe that the frustration-induced spin-gap phases obtained here play a key role in clarifying the phase diagram of the 2D spin-S orthogonal-dimer model.

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