First-order quantum phase transition in the orthogonal-dimer spin chain

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We investigate the low-energy properties of the orthogonal-dimer spin chain characterized by a frustrated dimer-plaquette structure. When the competing antiferromagnetic couplings are varied, the first-order quantum phase transition occurs between the dimer and the plaquette phases, which is accompanied by nontrivial features due to frustration: besides the discontinuity in the lowest excitation gap at the transition point, a sharp level crossing occurs for the spectrum in the plaquette phase. We further reveal that the plateau in the magnetization curve at 1/4 of the full moment dramatically changes its character in the vicinity of the critical point. It is argued that the first-order phase transition in this system captures some essential properties found in the two-dimensional orthogonal-dimer model proposed for $SrCu_2(BO_3)_2$.

I. INTRODUCTION

Recent extensive experimental and theoretical investigations on low-dimensional spin systems with frustration have been providing a variety of interesting topics. Among others, the spin-gap compound found recently by Kageyama *et al.*[1], $SrCu_2(BO_3)_2$, exhibits a number of nontrivial properties due to strong frustration.[1–5] In particular, the discovery of the magnetization plateaus at 1/3, 1/4, and 1/8 of the full moment[1,2] has stimulated further intensive studies.[6–8] The remarkable point claimed by Miyahara and Ueda[6] is that this frustrated system is a prototypical example of the two-dimensional (2D) version of *the orthogonal-dimer model*,[9] whose unique structure gives rise to various unusual properties.[10–13] This 2D model is known to be equivalent to the frustrated Shastry-Sutherland model on a square lattice with some diagonal bonds.[14] It has been recently shown that there exists a first-order quantum phase transition between the dimer and plaquette phases in the 2D orthogonal-dimer model, which is accompanied by jump and cusp singularities in the spin gap near the transition point.[15] What is most interesting is that the compound $SrCu_2(BO_3)_2$ may be located in the vicinity of the first-order transition point.[1,6]

As seen from the above studies, characteristic properties in the 2D system around the first-order transition point are certainly caused by the strong frustration common to this class of the orthogonal-dimer spin systems. Therefore, to clarify the essential properties of the system, further systematic studies of the quantum phase transition are highly desired. The 1D version of the orthogonal-dimer model[16–19] may be the most appropriate system for this purpose, because it is the simplest model which possesses the frustration effect due to the dimer-plaquette structure.

Motivated by these hot topics, in this paper, we study the first-order transition in the 1D orthogonal-dimer spin chain in detail.[16–19] The Hamiltonian we shall deal with is

$$\mathcal{H}=J\sum_{(i,j)}\mathbf{S}_i\cdot\mathbf{S}_j+J'\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S}_j-H\sum_i S_i^z, \qquad (1)$$

where $\mathbf{S}_i$ is the $s=1/2$ spin operator at the ith site, and the indices (i,j) and $\langle i,j\rangle$ represent the summation over intra- and inter-dimer pairs, respectively (see Fig. 1). The magnetic field is denoted as H for which we set $g\mu_B=1$ for convenience. Both of the exchange couplings J and J' are assumed to be antiferromagnetic. We shall use the normalized parameters $j=J'/J$ and $h=H/J$ in the following discussions.

It was previously shown by Ivanov and Richter[17,18] that the above orthogonal-dimer chain undergoes a first-order quantum phase transition between the dimer phase (for small j) and the plaquette phase (for large j) as the coupling ratio j is varied. However, the unique properties inherent in this frustrated system have not been discussed in detail yet, especially around the critical point. In what follows, by means of exact diagonalization (ED) and series expansion methods, we demonstrate that the excitation gap in this system exhibits nontrivial behaviors such as the jump and cusp singularities around the transition point, reflecting the dimer-plaquette dual properties characteristic of this frustrated system. We also point out that such properties are not specific to the 1D system but also common to the 2D orthogonal-dimer system. By computing the magnetization curve by means of the ED together with the density-matrix renormalization group (DMRG),[20,21] we further reveal that the formation of the magnetic plateaus is dramatically affected by the dimer-plaquette dual properties, and the resulting magnetic phase diagram has a rich structure.

This paper is organized as follows. In Sec. II, we investigate how the characteristic properties inherent in the orthogonal-dimer chain emerge in the low-energy excitations around the first-order transition point by means of the ED, the DMRG,[20,21] and the series expansion method.[22] In Sec. III, by calculating the magnetization curve by the ED and the DMRG, we show that the plateau formation in the magnetization changes its character around the transition point, re-

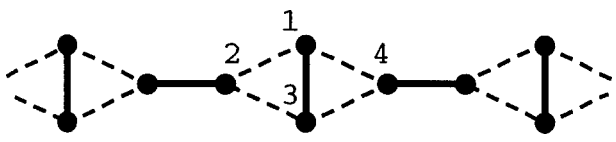

FIG. 1. The orthogonal-dimer chain. The solid and dashed lines indicate the antiferromagnetic exchange couplings J and J', respectively.

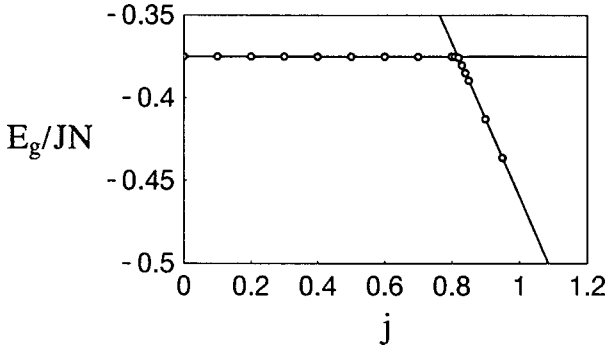

FIG. 2. The ground-state energy per site for the orthogonal-dimer chain as a function of $j(=J'/J)$. The flat line ($E_g/JN=-3/8$) indicates the exact ground-state energy of the dimer phase. The solid line is the result obtained by the plaquette expansion. We also show the results for the finite chain of $N=24$ as open circles.

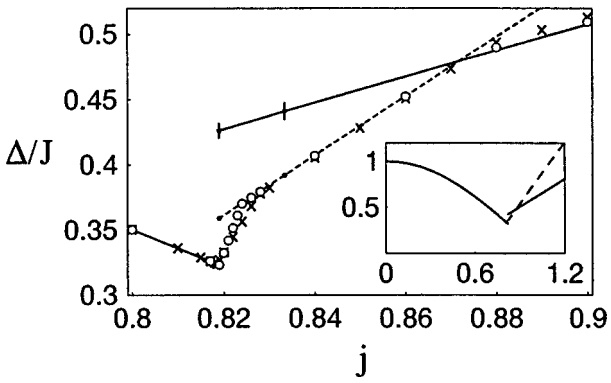

FIG. 3. The spin gap Δ/J for the orthogonal-dimer chain as a function of $j(=J'/J)$. The crosses and open circles indicate the results obtained by the ED method for the finite chain of $N=16$ and 24, respectively. See the text about the solid and dashed lines.

flecting the strong frustration effects. The last section is devoted to summary and discussions.

II. ZERO-FIELD PROPERTIES

In this section, we investigate the quantum phase transition in the orthogonal-dimer chain in the absence of the magnetic field,[17,18] by exploiting the ED and the series expansion methods.[22]

A. Ground state

We start with the ground-state properties in the dimer phase. When $j=0$, the system is reduced to an assembly of the decoupled dimers denoted by the solid line in Fig. 1, for which the product of independent dimer-singlets gives the ground state. The remarkable point for the orthogonal-dimer chain[6,17,18] is that this simple dimer-singlet state is always an exact eigenstate of the Hamiltonian (1) with the energy $E_g/JN=-3/8$ in the entire range of j (see Fig. 2), where N is the number of total sites. Accordingly, the dimer-singlet state should be the exact ground state up to a certain critical value of j.

In the opposite limit of large j, the ground state is given by the disordered singlet state which is adiabatically connected to the isolated plaquette singlets denoted by the dashed line in Fig. 1. Therefore, starting from the isolated plaquette singlets for $j=\infty$, we can evaluate the ground-state energy E_g for the plaquette phase with finite j by means of a series expansion. Performing the plaquette expansion up to the 11th order in j^{-1} combined with the first-order inhomogeneous differential method,[23] we have obtained the ground-state energy E_g rather precisely for the plaquette phase. The result is shown as the solid line in Fig. 2. It is seen that the ground state energy of the plaquette state coincides with that of the dimer state at a certain value of j, at which the first-order quantum phase transition occurs. The critical value is estimated as $j_c=0.819\,00$, which is further confirmed to be accurate up to the above figure by the DMRG calculation for the infinite chain. We note here that the present results for the ground state are consistent with those obtained by Ivanov and Richter.[17,18]

B. Spin excitations

Let us move to the spin excitation spectrum. In the dimer phase ($0<j<j_c$), we can construct a low-energy triplet excitation by substituting a local triplet for one of singlet dimers forming the ground state. The remarkable point is that the hopping of this local triplet across the singlet dimer such as the 1-3 dimer in Fig. 1 is forbidden by its characteristic crystal structure.[6] This implies that an excited triplet state is completely localized inside the finite strip ($N=6$), and has no dispersion for its spectrum. Thus we can exactly estimate the spin gap from the ED calculation for the cluster of $N=6$, which has been already evaluated by Richter *et al.*[18] In Fig. 3, the ED results are shown as the solid line in the dimer phase.

In the plaquette phase ($j>j_c$), it is naively expected that the low-energy excitation is described in terms of a triplet excitation on the isolated plaquette. However, the frustrating diagonal interaction in each plaquette should considerably affect the excitation spectrum. To see this, it is instructive to examine the energy-level structure of the isolated plaquette with the diagonal bond (see Fig. 1), whose Hamiltonian reads

$$\mathcal{H}_{\text{plaquette}}=J\mathbf{s}_1\cdot\mathbf{s}_3+J'(\mathbf{s}_1+\mathbf{s}_3)\cdot(\mathbf{s}_2+\mathbf{s}_4). \tag{2}$$

We list the energy eigenvalues of the plaquette in Table I,

TABLE I. Eigenstates of an isolated plaquette.

	D sector		P sector			
S_{13}	0	0	1	1		
S_{24}	0	1	0	1		
S	0	1	1	0	1	2
E/J	$-\frac{3}{4}$	$-\frac{3}{4}$	$\frac{1}{4}$	$-2j+\frac{1}{4}$	$-j+\frac{1}{4}$	$j+\frac{1}{4}$

where S_{13} (S_{24}) is a quantum number of the spin $\mathbf{s}_1+\mathbf{s}_3$ ($\mathbf{s}_2+\mathbf{s}_4$) and S is that for the total spin.

It is seen in this table that the eigenstates of the isolated plaquette are classified into two sectors: the D sector for $S_{13}=0$ and the P sector for $S_{13}=1$. For $j>1/2$, the ground state is always singlet in the P sector, whereas both of two separated sectors appear for the excitations when the coupling ratio j is varied. When $1/2<j<1$, the lowest excitation is given by the S_{24} singlet and S_{24} triplet in the D sector, which are fourfold degenerate with the excitation energy $\Delta E/J=2j-1$. In the regime $j>1$, the first excitation is a P-sector triplet with the excitation energy $\Delta E/J=j$. Note that as far as the singlet-triplet excitations in the P sector are concerned, there are no contributions from the D sector. This simple fact allows us to replace S_{13} in the plaquette with the $s=1$ spin, which will be used below to perform the mixed-spin cluster expansion for the P sector.

Keeping the above properties in mind, we now turn back to the orthogonal-dimer chain in the thermodynamic limit, by taking into account the interaction among plaquettes. Note that excitations in the D sector are still completely separated from those in the P sector, according to the constraint specific to the orthogonal-dimer structure. This remarkable fact enables us to study the excitation spectrum for the D sector and the P sector separately. Namely, we perform the plaquette expansion up to the 11th order in j^{-1} for the D sector, whereas for the P sector we exploit the mixed-spin cluster expansion[24] up to the seventh order in j^{-1} with a starting spin configuration $1/2\circ 1\circ 1/2$.[24] Applying the first-order inhomogeneous differential method[23] to the obtained series, we then deduce the lowest excitations both for the D sector and the P sector, as shown in Fig. 3. In this figure, the dashed line in the plaquette phase represents the energy for fourfold degenerate excitations in the D sector whereas the solid line is for a triplet excitation in the P sector. It is seen that the energy of two kinds of excitations intersects each other at $j_c'=0.872$, giving rise to a cusp singularity in the spin gap as a function of j. In the region $j>j_c'$, both of the ground state and the lowest excited state belong to the P sector on each plaquette, as is the case for the simple plaquette chain.[16] On the other hand, when $j_c<j<j_c'$, the lowest excitation is described by fourfold-degenerate level in the D sector although the ground state still belongs to the plaquette phase (P sector). The present plaquette expansion further uncovers that the wave function of the localized D-sector excitation with no dispersion is spatially extended over a number of sites, which shows sharp contrast to the dimer phase where the wave function of the triplet excitation is completely localized at a given site. This may imply that the fourfold-degenerate excitations characterized by the D sector possess the intermediate properties between those typical for the dimer phase and the plaquette phase. As j is further decreased, we encounter the first-order quantum phase transition, at which the ground-state energy of the two phases coincide with each other, while the first excited states have still different energies. Therefore, the spin gap at the transition point jumps from $\Delta^+(j_c)=0.3590(2)$ for the plaquette phase to $\Delta^-(j_c)=0.323\,09$ for the dimer phase.

In order to complement the series-expansion results, we also compute the lowest triplet excitation by the ED method for the finite chain ($N=16,24$) with the periodic boundary condition. The obtained spin gap Δ is shown as the crosses and the open circles in Fig. 3. It is seen that they are in fairly good agreement with the results obtained by the series expansion method, except for the vicinity of the first-order transition point j_c, where the finite-size effect in the ED still remains. Here it should be noticed that the present results provide much more detailed information for spin excitations than those obtained by Ivanov and Richter, where the spin gap as a function of j has only a cusplike singularity at the critical point.[17,18] We also note that the ladder system with a similar orthogonal-dimer structure has the jump and the cusp singularities in the spin gap.[25]

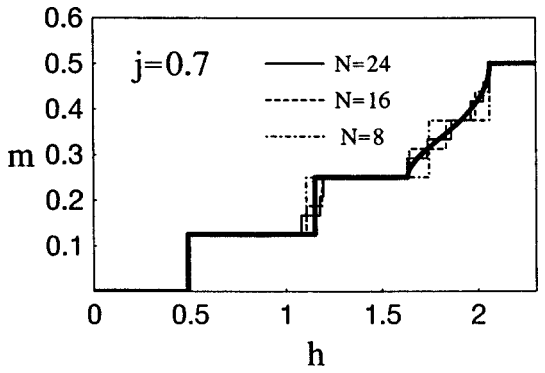

FIG. 4. The magnetization m ($=M/N$) as a function of h in the dimer phase ($j=0.7$). Three thin lines represent the magnetization curves for the finite chain of $N=8$, 16, and 24. The bold line is the result obtained by the DMRG method for the infinite chain.

III. PLATEAUS IN THE MAGNETIZATION CURVE

In this section, we study the magnetic properties of the orthogonal-dimer chain. By using the ED and the DMRG (Refs. 20 and 21) methods, we calculate the magnetization curve as a function of the magnetic field h ($=H/J$), and show that their characteristic behavior stems from the dimer-plaquette dual properties found for the excitations in the previous section. To clearly understand the magnetization curve in the low-field region, we shall use the description based on hard-core bosons, in which low-energy triplet excitations are regarded as hard-core bosons.

A. Dimer phase

Let us start with the dimer phase. In Fig. 4, we show the magnetization curve for $j=0.7(<j_c)$ obtained by the ED method for the finite chain of $N=8$, 16, and 24. We also show the results obtained by the DMRG method. In this figure, the plateaus appear in the magnetization curve at 1/4 and 1/2 of the full moment ($m=1/2$) clearly.

The mechanism of the 1/2-plateau formation is simply understood according to the unit-cell structure of the chain. As will be shown below, the 1/2 plateau appears in the entire range of j. On the other hand, the 1/4 plateau may be realized by the commensurate state in which each triplet excitation is aligned periodically at every finite strip of $N=8$, reflecting

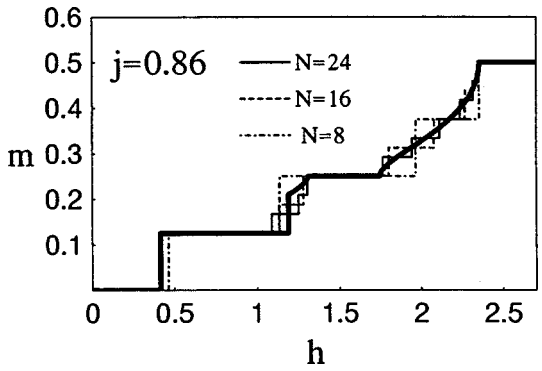

FIG. 5. The magnetization m ($=M/N$) as a function of h in the plaquette phase ($j=0.86$). Three thin lines represent the magnetization curves for the finite chain of $N=8$, 16, and 24. The bold line is the result obtained by the DMRG for the infinite chain.

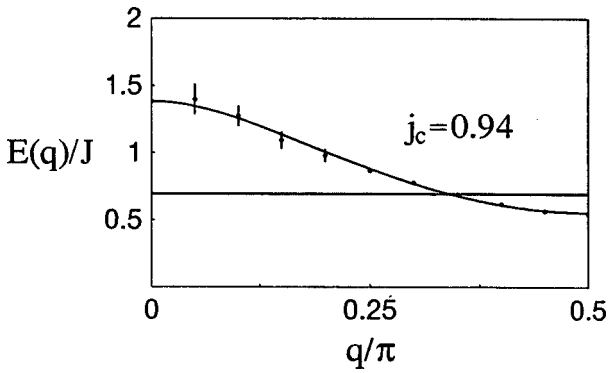

FIG. 6. The dispersion relation for $j=0.94$. The flat line and the solid curve represent the fourfold-degenerate D-triplet excitations and the P-triplet excitations, which are obtained by the mixed-spin cluster expansion and the plaquette expansion, respectively.

the completely localized nature of the triplet excitation discussed in the previous section. Since the localized triplet does not correlate with those in the neighbor strips, the macroscopic number of the triplets become degenerate at the lower critical field h_c (= the spin gap Δ), where the magnetic first-order transition occurs. In Fig. 4, we can indeed see that the magnetization curve jumps from $m=0$ to $m=1/8$ (1/4 plateau) at h_c without any finite-size correction. Increasing the magnetic field, the other first-order transition to the 1/2-plateau state occurs. It is seen in Fig. 4 that there still remains the large finite-size effect near this critical point, since the triplet excitations correlate with each other in contrast to the transition discussed above.

We here note that several different phases observed in the magnetization process are specified by the spatial arrangement of the spin quantum number, S_{13}, for the diagonal bond in each plaquette (see Table I). In the dimer phase, the D sector ($S_{13}=0$) is realized in each plaquette and in the 1/4-plateau state, the D and the P sectors are crystallized alternately. On the other hand, in the region of $m>1/4$, each diagonal bond in the plaquette forms $S_{13}=1$ (the P sector), and hence the frustration does not play any role for the magnetization process in this region. Therefore by calculating the magnetization for the unfrustrated plaquette chain, we can simply reproduce that for the orthogonal-dimer chain in this region.

B. Plaquette phase

We recall that there are two sort of excitations characterized by the P and the D sectors in the plaquette phase, which may provide a variety of the magnetization process. More precisely, the hard-core boson description suggests three possible behaviors for the magnetization curves.

(i) Case I: D-triplet excitations lie energetically lower than P-triplet excitations. The magnetization curve has a structure similar to that in the dimer phase.

(ii) Case II: D-triplet excitations lie slightly above the bottom of the P-triplet dispersion curve (see Fig. 6). Thus both of the features specific to the D-triplet and the P-triplet excitations appear in the magnetization curve.

(iii) Case III: D-triplet excitations lie sufficiently higher above the spin gap for P-triplet excitations. The magnetization curve may be characterized solely by the P-triplet excitation.

Since the above classification is simply based on the hard-core boson description, the interaction among bosons becomes more important when the number of bosons (i.e., the strength of the external field) is increased, which requires more proper discussions beyond the hard-core boson description. In what follows, based on the numerical results, we confirm that the magnetization curves are indeed classified into the above three categories.

We start with the analysis of the case I. We show the magnetization process for $j=0.86$ in Fig. 5, where we can see the 1/4 and 1/2 plateaus clearly. In particular the magnetization jumps from zero to the 1/4 plateau at the critical field h_c with a small finite-size correction.

In the plaquette phase, the 1/4 plateau is generated by the commensurate state in which the plaquette singlet and triplet in the D sector are crystallized alternately. The characteristic point in the 1/4 plateau state is that D-triplet excitations interact with each other when they are condensed to form the 1/4 plateau, in contrast to those in the dimer phase where each triplet excitation is localized completely. We can indeed see this interaction effect in the ED spectrum, where the triplets gain the condensation energy, by which the critical field h_c shifts to a slightly lower field than the spin-gap value. We note here that since the mechanism of the 1/4 plateau formation (first-order transition) at h_c is essentially the same in the dimer phase and in the plaquette phase, so that the critical field h_c does not show the discontinuity at $j=j_c$ as a function of j (see Fig. 9), being contrasted to the discontinuity observed in the spin gap at $j=j_c$. Increasing the field, the magnetic first-order transition takes place to the state in which $S_{13}=1$ is formed in each plaquette. Beyond this critical field, the magnetization traces the curve which is the same as that for unfrustrated plaquette chain.

In case II, the D-triplet excitation level lies slightly above the bottom of the P-triplet dispersion curve, as shown in Fig. 6.

The coexistence of two kinds of distinct excitations influences the magnetization curve considerably, as seen in Fig. 7. As h increases beyond the critical field ($h_c=\Delta$), the mag-

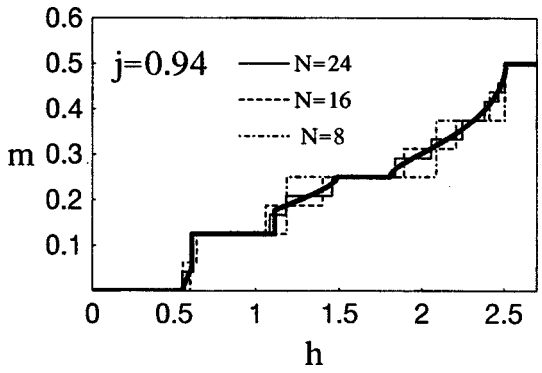

FIG. 7. The magnetization m $(=M/N)$ as a function of h in the plaquette phase $(j=0.94)$. Three thin lines represent the magnetization curves for the finite chain of $N=8$, 16, and 24. The bold line is the result obtained by the DMRG for the infinite chain.

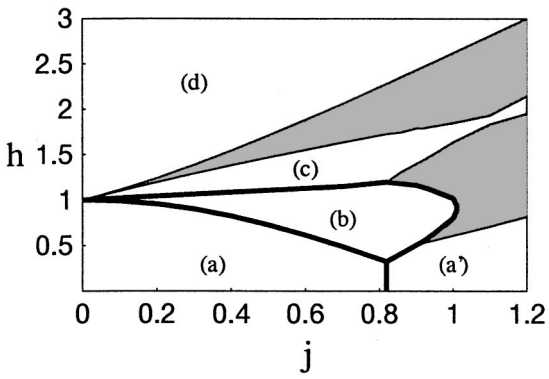

FIG. 9. The ground-state phase diagram on the j vs h plane. The phase boundaries are determined by the ED method for the finite chain of $N=24$ and the DMRG method for the infinite chain.

netization should develop with $m\sim(h-h_c)^{1/2}$, since it is dominated by the P-triplet excitation whose dispersion relation is quadratic near the bottom $(q=\pi/2)$. In fact, we can see a staircase structure below the 1/4 plateau for the ED results in Fig. 7, implying that the jump is now changed to the continuous increase in the magnetization. With slightly increasing h, however, the magnetization may stop to increase continuously and jump to the 1/4 plateau. Beyond the 1/4 plateau, we again encounter the first-order phase transition, which is accompanied by the jump in m. This jump singularity is followed by the continuous increase in m, as already discussed in case I.

In case III, the excitation energy of the D triplet is pushed up in the higher-energy region of the dispersion curve, as shown in Fig. 8. Thus, before the Zeeman energy lowers the D-triplet level down to zero, a number of bosons are accommodated in the P-triplet excited levels, and then the interaction effect between bosons becomes too significant to use the hard-core boson description. In this parameter region, we find that the 1/4 plateau completely disappears and the magnetization curve becomes smooth up to the 1/2 plateau.

Consequently, we end up with the magnetic phase diagram on the $(j-h)$ plane as shown in Fig. 9.

In this figure, (a) and (a′) indicate the dimer-singlet and plaquette-singlet phases, respectively. The region (b) exhibits the plateau in the magnetization curve at 1/4 of the full moment. We can see that the 1/4 plateau phase with the dimer-singlet structure exists not only in the dimer phase $(j<j_c)$ but also in a part of the plaquette phase. In region (c), the 1/2 plateau appears, which is stable in the whole parameter region of j. All spins are polarized in (d). The bold (solid) line shows the phase boundary of the first- (second-) order transition. It is now seen that the magnetic phase diagram possesses quite a rich structure, which is indeed caused by the dimer-plaquette dual properties inherent in this strongly frustrated spin chain. Here we wish to mention that the ladder system with a similar orthogonal-dimer structure[25] has the jump and the cusp singularities in the magnetization curve.[26]

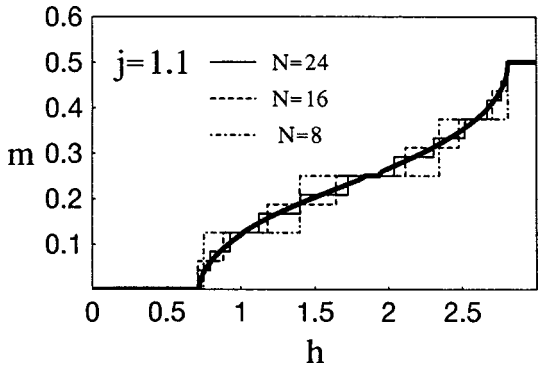

FIG. 8. The magnetization m $(=M/N)$ as a function of h in the plaquette phase $(j=1.1)$. Three thin lines represent the magnetization curves for the finite chain $N=8$, 16, and 24. The bold line is the result obtained by the DMRG for the infinite chain. Note that the 1/2-plateau state still exists near $h=1.9$.

IV. SUMMARY AND DISCUSSION

We have studied the low-energy properties of the orthogonal-dimer spin chain with a frustrated dimer-plaquette structure, by means of the exact diagonalization, the density matrix renormalization group, and the series expansion method. When the interdimer couplings are varied, the first-order quantum phase transition occurs at the critical value $j_c=0.819\,00$ between the dimer-singlet and plaquette-singlet phases. In the dimer phase, the ground state is exactly given by the decoupled dimer-singlet state, and triplet excitations over it are completely localized. In the plaquette phase, on the other hand, we have found that the excited states are characterized by the dual properties: the coexistence of plaquettelike and dimerlike excitations. As a result, both of the cusp and the jump appear in the spin gap as a function of j, which are caused by the level crossing between the excited states. We have also clarified how the above dual properties influence the magnetization process of the chain by analyzing the numerical results in terms of the hard-core boson description. In particular, we have shown that three types of the magnetization curves appear around the magnetization plateau at 1/4 of the full moment, according to how two kinds of triplet excitations change their relative positions energetically. We have thus found that the magnetic phase diagram on the j-h plane has a rich structure.

Although we have dealt with the one-dimensional chain in

this paper, we believe that the present study captures some essential properties of the first-order phase transition and the resulting rich structure in low-lying excited states even in two dimension. For the compound $SrCu_2(BO_3)_2$, it is indeed pointed out that the low-energy triplet excitation has quite small mobility reflecting the orthogonal-dimer structure.[6] Since this compound is located in the vicinity of the first-order phase transition point, the present results encourage us to interpret the plateau-formation mechanism of the 2D compound in terms of the dual properties originating from the dimer and plaquette structures. In particular, it is an interesting problem to study how the characteristic properties in 1D are changed into those in 2D by adiabatically introducing the interchain coupling, which may provide a clue to fully understand the physics in 2D orthogonal-dimer system.

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