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Matrix formalism for the VBS-type models and hidden order

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Abstract. We give a simple method to reduce the VBS-type states, usually expressed by the valence bonds, to the matrix-product representation. In this representation, peculiar properties of the VBS-type states are quite manifest. Using it, we investigate hidden order in the spin-S VBS model. The possibility that the Haldane phase for higher spin S is characterized by string order parameters is discussed. A picture of an approximate excitation is also presented.

1. Introduction

One-dimensional quantum-spin systems have a long history and have provided us with many examples of exactly soluble many-body systems. Since the well known exact solution of the $S = \frac{1}{2}$ Heisenberg chain due to Bethe, much work has been devoted to the study of soluble Heisenberg-like chains [1–4] and from the knowledge of these solutions it was believed for a long time that the Heisenberg chain was massless (or critical) for all spin S.

However, in 1983, on the basis of the semi-classical argument, Haldane predicted [5, 6] that the Heisenberg chains are massive for integer-S cases. Although this is quite surprising in view of the ordinary spin-wave-like argument, his prediction was verified by numerical calculations for lower S [7–9].

In 1988, Affleck, and co-workers [10] argued that a special model of bilinear–biquadratic chains

$$\mathcal{H} = \sum_j \left[(\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1}) + \frac{1}{3} (\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1})^2 \right]$$

has an exact massive ground state called the valence-bond-solid (VBS) state and indeed it possesses expected properties for Haldane systems (see [11] for a review). At least for the $S = 1$ case, the qualitative validity of the VBS picture has been verified by several authors [12–14].

In the following, we mainly treat the spin-S VBS model, whose Hamiltonian essentially consists of the projection operators of su(2) (see section 2 for the definition of the model). The ground state of the Hamiltonian is given by the so-called (spin-S) VBS state [15] (its definition of is given in the next section. see (3)). Remarkably, in this ground state we can calculate several correlation functions *exactly* [10, 15, 16] and verify that the ordinary correlation functions decay exponentially. Hence we expect these models have massive excitations in agreement with the Haldane conjecture.

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On the other hand, it was argued by den Nijs and Rommelse [17] and later by Tasaki [18] that the ground states of these models have a special type of long-range order (LRO) called *the string order*. It can be measured by the string order parameter

$$\mathcal{O}^{\alpha}_{\text{string}} = \lim_{|i-j|\nearrow\infty} \left\langle S^{\alpha}_{i} \prod_{k=i}^{j-1} \exp\left(\mathrm{i}\pi S^{\alpha}_{k}\right) S^{\alpha}_{j} \right\rangle$$

whose relevance is numerically supported [19, 20]. Furthermore, Kennedy and Tasaki successfully related the Haldane phase of $S = 1$ chains to the breakdown of the discrete $Z_2 \times Z_2$ symmetry [20, 21].

After their paper, Oshikawa tried to extend their picture to higher-S cases to obtain the fact that this symmetry is *unbroken* for even-S ($S \geqslant 2$) cases [22]. This implies that the Kennedy–Tasaki picture for the $S = 1$ case is not sufficient to describe the hidden symmetry in higher-S systems. What kind of hidden order exists in higher-S chains is still unclear. In the present paper, we attempt to attack this problem. For two reasons, we use the VBS model instead of the original Heisenberg (i.e. consisting of pure bilinear interaction) model. One reason is that the analytic calculation is possible for the VBS models while the numerical simulation becomes harder for higher values of S. The second is that the nine-fold (near) degeneracy recently observed in the numerical calculation [23] implies that the $S = 2$ VBS model can be used as the first approximation to the original Heisenberg model.

This paper is organized as follows. We give the definition of the model and its exact VBS ground state in section 2. Our main results are also summarized there. The reader who is not interested in the technical details will skip sections 3, 4, and 5. In section 3 we generalize the matrix-product representation [24, 25] to higher-S VBS states and clarify its connection to the Schwinger boson representation of the VBS states [15, 16]. In this representation, the peculiar structure of the VBS states is manifest. Calculation of the generalized string order is performed in section 4 using the matrix formalism. Section 5 is devoted to the discussion of the low-lying excitation of the VBS models. We show that an approximate elementary excitation has a topological property and that it is responsible for the rapid decrease of the long-range string order at finite temperatures recently found in the Monte Carlo calculation [26]. A remarkable equivalence between two pictures of the low-lying excitation is also proved. The matrices necessary for our calculation are summarized in appendix A. In appendix B, the difference in the structure of the low-lying excitations between integer S and half-odd-integer S is briefly discussed in the context of the Lieb–Schultz–Mattis theorem.

2. The model and the main results

The VBS model which we discuss in the following sections is defined as a collection of a kind of su(2) projection operators (strictly speaking, they are not projection operators for $S \geqslant 2$):

$$\mathcal{H}_{\text{VBS}} = \sum_{i=1}^{L} P^{2S}_{S+1}(\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1}) \tag{1}$$

where $\boldsymbol{S}_i$ is a spin operator with spin S and

$$P^{2S}_{S+1}(\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1}) \equiv \frac{1}{\mathcal{N}} \prod_{J=0}^{S} \left\{ \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1} - \frac{1}{2}\left[J(J+1) - 2S(S+1)\right] \right\} \tag{2}$$

denotes such an $(S+1)$-th order polynomial of the exchange interaction $\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1}$ as projects out the subspaces with $J = S+1, \ldots, 2S$ ($J(J+1) = (\boldsymbol{S}_i + \boldsymbol{S}_{i+1})^2$). The normalization

factor $\mathcal{N}$ has been introduced so that the linear coefficient in (2) may be unity. We list $\mathcal{H}_{\rm VBS}$ for first few values of S:

(i)

$$\mathcal{H}_{S=1} = \sum_i \left\{ \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1} + \frac{1}{3} (\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1})^2 + \frac{2}{3} \right\}$$

(ii)

$$\mathcal{H}_{S=2} = \sum_i \left\{ \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1} + \frac{2}{9} (\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1})^2 + \frac{1}{63} (\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1})^3 + \frac{10}{7} \right\}$$

(iii)

$$\mathcal{H}_{S=3} = \sum_i \left\{ \boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1} + \frac{59}{358} (\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1})^2 + \frac{19}{1611} (\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1})^3 + \frac{1}{3222} (\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1})^4 + \frac{396}{179} \right\}.$$

Since the local Hamiltonian $P^{2S}_{S+1}(\boldsymbol{S}_i \cdot \boldsymbol{S}_{i+1})$ expresses a kind of projection operator, the exact ground state is easily found to be a product of the valence-bond operators [15]

$$|\mathrm{VBS};\, p, q\rangle = Q_{\rm left}(a_1^\dagger, b_1^\dagger;\, p) \prod_{k=1}^{L-1} (a_k^\dagger b_{k+1}^\dagger - b_k^\dagger a_{k+1}^\dagger)^S Q_{\rm right}(a_L^\dagger, b_L^\dagger;\, q)|0\rangle\!\rangle. \tag{3}$$

In the above expression, $a_i^\dagger$ and $b_i^\dagger$ denote the ordinary Schwinger bosons at site i and the polynomials $Q_{\rm left}$ and $Q_{\rm right}$ represent the left and right edge states, respectively. We are free to choose them, provided that they are homogeneous polynomials in $a_1^\dagger$ and $b_1^\dagger$ ($a_L^\dagger$ and $b_L^\dagger$) of degree S. Thus, the ground state for an open chain is $(S+1) \times (S+1)$-fold degenerate. We construct an alternative expression of this state, using $(S+1) \times (S+1)$ matrices (see (15) and (16))

$$g_j(p, q) = (-1)^{S-p+1} \sqrt{{}_S\mathrm{C}_{p-1}\, {}_S\mathrm{C}_{q-1}} \sqrt{(S-p+q)!(S+p-q)!}\, |S;\, q-p\rangle_j \tag{4}$$

where ${}_m\mathrm{C}_n$ denotes the number of ways we pick up n identical objects out of m and is also expressed as $\binom{m}{n}$.

To investigate hidden strucures in the VBS model, we calculate two types of non-local string order parameter

$$\mathcal{O}^z_{\rm string}(\theta) = \lim_{|i-j|\nearrow\infty} \left\langle S_i^z \prod_{k=i}^{j-1} \exp\left(\mathrm{i}\theta S_k^z\right) S_j^z \right\rangle \tag{5}$$

$$\mathcal{O}^z_{\rm mod}(\theta) = \lim_{|i-j|\nearrow\infty} \left\langle \mathcal{P}(S_i^z) \prod_{k=i}^{j-1} \exp\left(\mathrm{i}\theta S_k^z\right) \mathcal{P}(S_j^z) \right\rangle. \tag{6}$$

The operator $\mathcal{P}(S^z)$ is defined to satisfy the condition $\mathcal{P}(\pm S) = \pm S$ and $\mathcal{P} = 0$ otherwise. A physical motivation to consider such a modified order parameter is that we can realize simple ferromagnetic correlations between $\pm S$ just as in the $S = 1$ case. We have to stress here that we consider these order parameters not to reveal the mechanism of the gap generation but to detect peculiar structures in the VBS models. The above two order parameters are shown to be given by the following simple formulae:

$$\mathcal{O}^z_{\rm string}(\theta) = \left[\frac{1}{S+2} \left(\frac{\sin((S/2)\theta) - S\sin(\theta/2)\cos([(S+1)/2]\theta)}{\sin^2\theta/2} \right) \right]^2 \tag{7}$$

$$\mathcal{O}^z_{\rm mod}(\theta) = \left(\frac{2S}{2S+1} \right)^2 \sin^2\left(\frac{S\theta}{2} \right) \tag{8}$$

respectively, as derived in section 4. The order parameter $\mathcal{O}^z_{\text{string}}(\theta)$ has two large peaks around (not exactly at) $\theta = \pm\pi/S$, whose height increases as $S^2(2/\pi)^2$ with S (for large S). This is consistent with the numerical result that $\mathcal{O}^z_{\text{string}}(\pi/2)$ is a good order parameter for the $S = 2$ Heisenberg chain [27, 23]. The S^2 behaviour may remind us of the classical Néel ordered state. However, the facts that $\mathcal{O}^z_{\text{mod}}(\pi/S)$ behaves like unity for large S and that all S^z values occur with equal probability in the VBS state (see section 4) imply that this S^2 behaviour does not come from the classical Néel-like property but that it appears as a consequence of non-trivial optimization of the correlation. It is important to note that the precise position of the peaks can vary with the change of the state (namely, the modification of the matrix elements of g). Nevertheless, a simple symmetry argument (section 4) tells us that the peak at $\theta = \pi$ has a kind of topological stability. We also point out that the θ dependence of $\mathcal{O}^z_{\text{string}}(\theta)$ in the spin-$2S$ VBS state is the same as in the spin-S dimer state (see (34) and (36)). This may suggest the long-range dimer nature of the VBS state.

We can show for our special VBS case that the slow twist used by Affleck and Lieb does not create an 'excited' state orthogonal to the ground state (see appendix B). This may give some hints to consider the low-lying excitation of the integer-S chains. Instead of such a spin-wave-like excitation, we propose the crackion excitation first proposed by Knabe [28] for the $S = 1$ case as an elementary excitation in the spin-S VBS model. Concerning this excitation, we show the following.

(i) It takes on the form of a 'travelling matrix wave'. This 'plane-wave' state is known to reproduce the numerically observed spectrum of the VBS model for $S = 1$ [29, 30].

(ii) It has a kind of topological property. Contrary to the $S = 1$ case discussed by Fáth and Sólyom, the topological property of the crackion is not manifest in the S^z configuration itself. Nevertheless, if we observe the string-like operator V^z_j, its kink nature is visible. This is consistent with the recent observation that the Haldane gap is equal to the energy of the domain-wall formation [31, 32].

(iii) The decay of the (long-range) string order at finite temperatures [26] is brought about by the crackion condensation.

(iv) It has an elementary nature and hence the single-mode-approximation (SMA) (for the details of SMA, see [8, 34]) picture and the crackion picture are equivalent. The SMA estimate of the Haldane gap of the VBS model seems to be consistent with the well known formula (with a modified coupling constant).

3. Matrix formalism

In this section, we develop a method of representing the valence-bond-type states in terms of matrix products and describe how to compute various quantities using them.

The key ingredient is quite simple. First recall that the VBS states for open chains have left- and right-edge degrees of freedom. For the case of the spin-S VBS state

$$|\text{VBS};\, p, q\rangle = Q_{\text{left}}(a_1^\dagger, b_1^\dagger;\, p) \prod_{k=1}^{L-1} (a_k^\dagger b_{k+1}^\dagger - b_k^\dagger a_{k+1}^\dagger)^S Q_{\text{right}}(a_L^\dagger, b_L^\dagger;\, q)|0\rangle\rangle \quad (9)$$

($a_i^\dagger$ and $b_i^\dagger$ denote the Schwinger bosons) we have $(S+1)\times(S+1)$ edge states represented by two polynomials Q_{left} and Q_{right} of degree S, correspondingly. For later convenience, we choose

$$Q_{\text{left}}(a_1^\dagger, b_1^\dagger;\, p) = \sqrt{{}_S\text{C}_{p-1}}\,(a_1^\dagger)^{S-p+1}(b_1^\dagger)^{p-1} \quad (p = 1, \ldots, S+1) \quad \text{for the left edge}$$

$$Q_{\text{right}}(a_L^\dagger, b_L^\dagger;\, q) = \sqrt{{}_S\text{C}_{q-1}}\,(a_L^\dagger)^{q-1}(b_L^\dagger)^{S-q+1} \quad (q = 1, \ldots, S+1) \quad \text{for the right edge}$$

as the edge states Q_{left} and Q_{right}, respectively.

Let us express the spin-S VBS state as an $(S+1)\times(S+1)$ matrix whose entries are *states*. For example, we write the $S=1$ VBS state on a finite (length-L) open chain as

$$\begin{pmatrix} |\text{VBS}; L; \uparrow, \downarrow\rangle & |\text{VBS}; L; \uparrow, \uparrow\rangle \\ |\text{VBS}; L; \downarrow, \downarrow\rangle & |\text{VBS}; L; \downarrow, \uparrow\rangle \end{pmatrix}. \tag{10}$$

Then, we can make a new length-$(L+1)$ VBS state (written in terms of an $(S+1)\times(S+1)$ matrix) by multiplying a matrix of the same size from the right [34]

$$|\text{VBS}; L+1\rangle = |\text{VBS}; L\rangle \otimes g_{L+1}. \tag{11}$$

The $(S+1)\times(S+1)$ matrix g is determined so that the valence-bond operator $(a_L^\dagger b_{L+1}^\dagger - b_L^\dagger a_{L+1}^\dagger)^S$ is created when we contract the matrix indices. For the $S=1$ VBS state, we have obtained

$$g_{L+1} = \begin{pmatrix} -|0\rangle_{L+1} & -\sqrt{2}|1\rangle_{L+1} \\ \sqrt{2}|-1\rangle_{L+1} & |0\rangle_{L+1} \end{pmatrix}. \tag{12}$$

Replacing $\uparrow$ or $\downarrow$ in the above equation by the corresponding edge (both left and right) states, it can be easily generalized to the higher-S cases. The requirement described above determines g matrices:

$$g_j(p,q) = (-1)^{S-p+1}\sqrt{{}_S\text{C}_{p-1}\,{}_S\text{C}_{q-1}}\,(a_j^\dagger)^{S-p+q}(b_j^\dagger)^{S+p-q}|0\rangle\rangle_j$$

$$= (-1)^{S-p+1}\sqrt{{}_S\text{C}_{p-1}\,{}_S\text{C}_{q-1}}\times\sqrt{(S-p+q)!(S+p-q)!}\,|S; q-p\rangle_j \tag{13}$$

$$g^{\text{start}}(p,q) = \sqrt{{}_S\text{C}_{p-1}\,{}_S\text{C}_{q-1}}\times\sqrt{(S-p+q)!(S+p-q)!}\,|S; q-p\rangle_1. \tag{14}$$

Using g_i and g^{start}, the VBS state for a finite open chain is given by a simple product of $(S+1)\times(S+1)$ matrices:

$$|\text{VBS}; p, q\rangle = \left[g^{\text{start}}\otimes g_2\otimes\ldots\otimes g_{L-1}\otimes g_L\right]_{(p,q)}. \tag{15}$$

For the case of a periodic chain, (15) is replaced by

$$|\text{VBS}\rangle_{\text{PBC}} = \text{Tr}\left[g_1\otimes g_2\otimes\ldots\otimes g_{L-1}\otimes g_L\right]. \tag{16}$$

This kind of representation was first discussed by Fannes *et al* [35, 24] in an abstract manner, and later by Klümper *et al* [36, 25] for the $S=1$ deformed VBS model. Our method of construction is quite simple and clarifies the connection between the valence-bond representation and the matrix-product one.

Below we shall list a few examples of g-matrices.

(i) For the $S=2$ VBS state:

$$g_i = \begin{pmatrix} 2|0\rangle_i & 2\sqrt{3}|1\rangle_i & 2\sqrt{6}|2\rangle_i \\ -2\sqrt{3}|-1\rangle_i & -4|0\rangle_i & -2\sqrt{3}|1\rangle_i \\ 2\sqrt{6}|-2\rangle_i & 2\sqrt{3}|-1\rangle_i & 2|0\rangle_i \end{pmatrix}.$$

(ii) For the $S=3$ VBS state:

$$g_i = \begin{pmatrix} -6|0\rangle_i & -12|1\rangle_i & -6\sqrt{10}|2\rangle_i & -\sqrt{6!}|3\rangle_i \\ 12|-1\rangle_i & 18|0\rangle_i & 12\sqrt{3}|1\rangle_i & 6\sqrt{10}|2\rangle_i \\ -6\sqrt{10}|-2\rangle_i & -12\sqrt{3}|-1\rangle_i & -18|0\rangle_i & -12|1\rangle_i \\ \sqrt{6!}|-3\rangle_i & 6\sqrt{10}|-2\rangle_i & 12|-1\rangle_i & 6|0\rangle_i \end{pmatrix}.$$

The greatest merit of this expression is that g consists only of states of a *single* site. Therefore we have only to treat *local* states $g_i(p,q)$ instead of non-local valence-bond operators.

Furthermore, the matrix formalism clarifies the special structure of the VBS states. The $(S+1)\times(S+1)$-fold degeneracy and the edge degrees of freedom are quite naturally incorporated in it. To see the step structure of the VBS states [17] in our approach, we first note that the product of an arbitrary number of the g-matrices can be written like

$$\left[g_i \otimes g_{i+1} \otimes \ldots \otimes g_{i+k}\right]_{(p,q)} = \left|\mathrm{VBS};\ \sum_{j=i}^{i+k} S_j^z = q-p\right\rangle. \tag{17}$$

The sum of S_j^z over an *arbitrary* interval of the spin-S VBS state is restricted between $-S$ and S (and hence $|\mathrm{VBS}\rangle$ can be expressed by a finite-dimensional matrix!) and this is nothing but the 'disordered flat' structure of the VBS states first pointed out by den Nijs and Rommelse [17]. The diluted antiferromagnetic order in the $S=1$ VBS state is peculiar to the spin-1 case, where this rule allows only configurations like $(\ldots,-1,0,0,1,0,-1,1,0,\ldots)$. If we depict an S^z configuration by steps whose height differences represent S^z values at each site, for the spin-S case we have step diagrams going upward and downward within height S (see figure 1). Namely, the sum of S^z values sitting to the left of a given arbitrary site (i.e. height of a step) does not exceed a certain restricted value. We can regard this property as a kind of 'quasi-charge neutrality'. Moreover, as is clear in the matrix representation, the S^z value of the newly added site is determined *only* by this S^z_{tot} (step height). This is reminiscent of the Markov process. The relation between such a Markov-like property and the short-ranged correlations was first stressed by Fannes *et al* [35, 24]. We stress here that the size of the g matrices is determined not by the spin S but by the number of valence bonds between two adjacent sites. Therefore it is possible to construct the matrix-product ground states of spin-S chains using smaller (than $(S+1)\times(S+1)$) matrices (such possibilities are briefly discussed in the appendix of [34]).

Next, we generalize the method proposed by Klümper *et al* [25] of calculating expectation values with respect to the VBS-type states to higher-S cases.

As we have seen, the VBS-type states are simply expressed using the g matrices

$$|\mathrm{VBS}\rangle = g^{\mathrm{start}} \otimes g_2 \otimes \ldots \otimes g_L.$$

In the above expression, it is important that g is written in terms of local states (usually those of a single site). We are interested in expectation values of the following type:

$$\langle \mathcal{A}_i \rangle = \langle \mathrm{VBS}|\mathcal{A}_i|\mathrm{VBS}\rangle. \tag{18}$$

Writing the matrix indices explicitly, we can express $\langle \mathcal{A}_i\rangle$ as

$$\begin{aligned}\langle \mathcal{A}_i\rangle_{\alpha,\beta} = \sum_{\{m_j,n_j\}} & g^{\dagger}_{\mathrm{start}}(\alpha,m_1)\otimes g_2^{\dagger}(m_1,m_2)\otimes\ldots\otimes g_L^{\dagger}(m_{L-1},\beta)\\ & \times \mathcal{A}_i\, g_{\mathrm{start}}(\alpha,n_1)\otimes g_2(n_1,n_2)\otimes\ldots\otimes g_L(n_{L-1},\beta)\end{aligned} \tag{19}$$

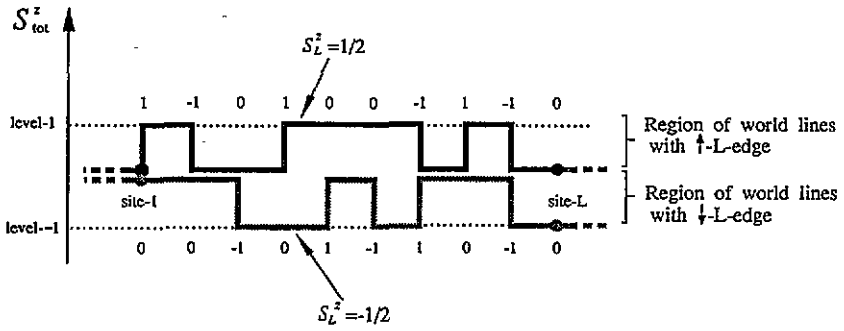
with equations (13) and (14), where α and β denote the left and right edge states respectively and $g^{\dagger}$ is a matrix which is obtained by replacing the 'ket' vectors in g by 'bra' vectors without transposing it. Using the local property of g, it can be rewritten as follows:

$$\begin{aligned}\langle \mathcal{A}_i\rangle_{\alpha,\beta} = \sum_{\{m_j,n_j\}} & g^{\dagger}_{\mathrm{start}}(\alpha,m_1) g_{\mathrm{start}}(\alpha,n_1)\times g_2^{\dagger}(m_1,m_2) g_2(n_1,n_2)\times\ldots\\ & \times g_i^{\dagger}(m_{i-1},m_i)\mathcal{A}_i\, g_i(n_{i-1},n_i)\times\ldots\times g_L^{\dagger}(m_{L-1},\beta) g_L(n_{L-1},\beta).\end{aligned} \tag{20}$$

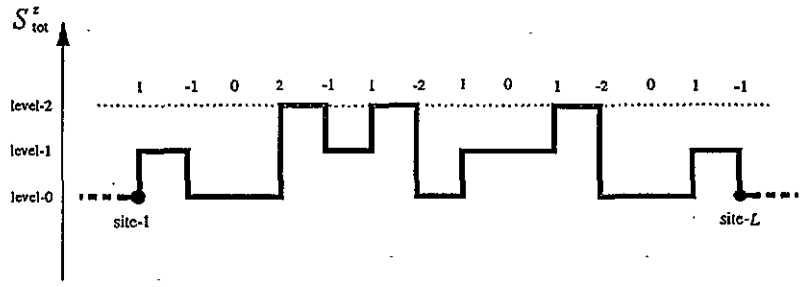
Then we introduce the G matrices whose entries are c numbers (see appendix A for concrete forms of G and $G^{\mathcal{A}}$)

$$G_{(m_{j-1},n_{j-1};m_j,n_j)} \equiv g_j^{\dagger}(m_{j-1},m_j) g_j(n_{j-1},n_j) \tag{21}$$

$$G^{\mathcal{A}}_{(m_{i-1},n_{i-1};m_i,n_i)} \equiv g_i^{\dagger}(m_{i-1},m_i)\mathcal{A}_i g_i(n_{i-1},n_i). \tag{22}$$



(a)



(b)

Figure 1. The step diagrams of a typical S^z configuration of the VBS state. (a) $S = 1$ VBS state. (b) $S = 2$ VBS state. The diluted AF order is obscured in the $S = 2$ case.

For our spin-S case, G is given by

$$G_{(\alpha,\beta;\gamma,\delta)} = (-1)^{\alpha+\beta}\sqrt{{}_S\mathrm{C}_{\alpha-1}\,{}_S\mathrm{C}_{\beta-1}\,{}_S\mathrm{C}_{\gamma-1}\,{}_S\mathrm{C}_{\delta-1}}$$
$$\times\sqrt{(S+\gamma-\alpha)!(S-\gamma+\alpha)!(S+\delta-\beta)!(S-\delta+\beta)!}\,\delta_{\alpha-\beta,\gamma-\delta}. \tag{23}$$

If we adopt the lexicographical ordering for the double (tensorial) indices of G to regard it as an ordinary matrix, then the RHS of (20) becomes

$$\langle\mathcal{A}_i\rangle_{\alpha,\beta} = (M_{\mathcal{A}})_{(\alpha,\alpha;\beta,\beta)} = \left[G^{\mathrm{start}}(G)^{i-2}G^{\mathcal{A}}(G)^{L-i}\right]_{(\alpha,\alpha;\beta,\beta)}. \tag{24}$$

For example, $\langle\mathcal{A}_i\rangle$ can be obtained as the following elements:

$$\langle\mathcal{A}_i\rangle_{\uparrow\downarrow} = (M_{\mathcal{A}})_{1,1} \qquad \langle\mathcal{A}_i\rangle_{\uparrow\uparrow} = (M_{\mathcal{A}})_{1,4} \qquad \langle\mathcal{A}_i\rangle_{\downarrow\downarrow} = (M_{\mathcal{A}})_{4,1} \qquad \langle\mathcal{A}_i\rangle_{\downarrow\uparrow} = (M_{\mathcal{A}})_{4,4}$$

for the $S = 1$ case.

For the case of a periodic boundary condition, (24) is replaced by

$$\langle\mathcal{A}_i\rangle = \sum_{\alpha,\beta}\sum_{\{m_j,n_j\}} g_1^{\dagger}(\alpha,m_1)\otimes\ldots\otimes g_L^{\dagger}(m_{L-1},\alpha)\mathcal{A}_i\, g_1(\beta,n_1)\otimes\ldots\otimes g_L(n_{L-1},\beta)$$
$$= \mathrm{Tr}(G)^{L-1}G^{\mathcal{A}}. \tag{25}$$

Equations (24) and (25) are the fundamental formulae for our calculation. If we regard G as the transfer matrix of a one-dimensional (not (1+1)-dim!) *classical* statistical system, equations (24) and (25) are nothing but the expressions of the expectation values. In [15],

Arovas *et al* argued that using the coherent-state method an expectation value $\langle \mathcal{A} \rangle$ can be written as that of a classical spin system. Our expressions ((24) and (25)) are the transfer-matrix version of it.

In general, G becomes huge as S increases ($((S+1)^2 \times (S+1)^2$ matrix for spin-S VBS state!). However, there is a nice property which simplifies the calculation—the 'asymptotic orthogonality'. As can be easily verified, the VBS states with different edge states are *not orthogonal and actually have exponentially small overlaps. For example, we have [10]*

$$\langle S=1;\uparrow,\downarrow | S=1;\downarrow,\uparrow\rangle = -2\left(\frac{-1}{3}\right)^L .$$

Therefore, the VBS states are only asymptotically orthogonal. This implies that in considering the thermodynamic limit we have only to keep the elements $G(i,i;j,j)$, which correspond to pairs with the same edge states. Namely, we are left with much smaller ($(S+1)\times(S+1)$ matrix!) matrices than the original ones. Fortunately, we can obtain all the $(S+1)^2$ eigenvalues of G for the spin-S VBS state from the results of the coherent-state method [16]. They are

$$\lambda(l) = (-1)^l \frac{(2S+1)!}{S+1} \frac{{}_S\mathrm{C}_l}{{}_{S+l+1}\mathrm{C}_l} \qquad (l=0,1,\ldots,S).$$

The degeneracy of $\lambda(l)$ is $2l+1$, and hence the maximum eigenvalue $\lambda(0) = (2S+1)!/(S+1)$ is unique. Combining this with the fact that $\lambda(0)$ belongs to the reduced $(S+1)\times(S+1)$ matrix, we can prove the above-mentioned 'asymptotic orthogonality' in the matrix formalism.

As is clear from the above argument, it is convenient to define the *reduced* matrices

$$G_{\mathrm{Red}}(p,q) \equiv G(p,p;q,q) = {}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}(S+p-q)!(S-p+q) \qquad (>0). \tag{26}$$

They are much smaller than the original ones and hence easier to handle. Some G_{Red} matrices relevant for our purpose are listed in appendix A. There is an interesting interpretation of the G_{Red} matrix. If we properly renormalize G_{Red} (we call the renormalized one $\widetilde{G}$), $\widetilde{G}$ can be viewed as a stochastic matrix (i.e. $\sum_j \widetilde{G}_{i,j} = 1$). In this picture, the edge states correspond to states of the Markov process and $\widetilde{G}$ gives transition probabilities between left and right edge states. This fact is useful in evaluating the asymptotic form of $G^L_{\mathrm{Red}}(L\to\infty)$.

4. The generalized string order

In the last section, we developed a method of computing various quantities using the matrix-products. In this section, we apply it to the calculation of string correlation functions and others and we attempt to uncover the physics of the VBS models.

The first attempt at calculating the string order parameters for the higher-S VBS models was done by Oshikawa [22]. He obtained the result that the ordinary string order parameter is *zero* for even S, while it is non-zero for odd S. This implies that the hidden $\mathrm{Z}_2\times\mathrm{Z}_2$-symmetry-breaking picture is not successful in explaining the Haldane phase for higher-S cases. For the $S=1$ case, we can relate the breakdown of this discrete symmetry to the appearance of the Haldane phase. Roughly speaking, this is because the Kennedy–Tasaki transformation succeeds in converting the VBS state into the 'diluted' ferromagnetic state. A brief calculation tells us that it does not work for $S \geqslant 2$, since the topological AF order is obscured there.

To detect the hidden order in higher-S cases, some generalized string order parameters have been proposed. Among them, the most popular one is [22, 27]

$$\mathcal{O}^z_{\text{string}}(i, j; \theta) = \left\langle S^z_i \prod_{k=i}^{j-1} \exp\left(\mathrm{i}\theta S^z_k\right) S^z_j \right\rangle. \tag{27}$$

In the singlet ground state (owing to the Marshall–Lieb–Mattis theorem, the Heisenberg model on a finite chain with an even number of sites satisfies this condition), it has the following property:

$$\mathcal{O}^z_{\text{string}}(i, j; \theta) = \mathcal{O}^z_{\text{string}}(i, j; \theta + 2\pi) = \mathcal{O}^z_{\text{string}}(i, j; -\theta). \tag{28}$$

Especially, it follows that $\mathcal{O}^z_{\text{string}}(i, j; \theta)$ is real and symmetric with respect to $\theta = \pi$ (and so is the string order parameter $\mathcal{O}^z_{\text{string}}(\theta) = \lim_{|i-j|\nearrow\infty} \mathcal{O}^z_{\text{string}}(i, j; \theta)$). Hatsugai [27] numerically investigated $\mathcal{O}^z_{\text{string}}(\theta = \frac{\pi}{2})$ for the $S = 2$ Heisenberg chain and concluded that it would be a good order parameter.

From this viewpoint, it is interesting to calculate the generalized string order parameter $\mathcal{O}^z_{\text{string}}(\theta)$ for the spin-S VBS state. We expect that the result may give us important information about the hidden order of the true spin-S Heisenberg model.

Before calculating $\mathcal{O}^z_{\text{string}}(\theta)$, we investigate the probability distribution of S^z values. To do this, we calculate the following average:

$$\frac{1}{\mathcal{N}} \langle \text{VBS} | P(S^z = m) | \text{VBS} \rangle$$

where $P(S^z = m)$ means a projection operator onto an $S^z = m$ state. It can be readily evaluated using the matrix formalism, since $P(m)$ is easily realized in terms of the G matrix (where necessary matrices are summarized in appendix A). Therefore we get

$$\text{Prob}(S^z_j = m) = \frac{1}{2S+1}. \tag{29}$$

This implies that each S^z value occurs with *equal* probability in the S VBS state. This is the reason for the vanishing one-point function (for the deformed VBS states [25, 37]: $\text{Prob}(S^z = m)$ values are unequal, though they still preserve the symmetry $\text{Prob}(S^z = m) = \text{Prob}(S^z = -m)$.

It is instructive to calculate the same quantity for the transformed VBS state defined by $|\widetilde{\text{VBS}}\rangle = U|\text{VBS}\rangle$, where U is the Kennedy–Tasaki unitary transformation compactly written down as [22]

$$U = \prod_{i=1}^{L} \exp\left(\mathrm{i}\pi \sum_{k=1}^{i-1} S^x_i S^z_k\right).$$

This gives a hint to understand why the hidden $Z_2 \times Z_2$-symmetry breaking does not occur for even-S models. Since the probability distribution for the transformed state $\widetilde{\text{Prob}}(S^z_j)$ is given by

$$\widetilde{\text{Prob}}(S^z_j = m) = \frac{1}{\mathcal{N}} \langle \text{VBS} | U^{-1} P_S(S^z_j = m)\, U | \text{VBS} \rangle$$

we have to compute the expectation value of $U^{-1} P_S U$. Noting that

$$\begin{aligned}
\langle \widetilde{\text{VBS}} | \left(S^z_j\right)^n | \widetilde{\text{VBS}} \rangle &= \left\langle \prod_{k=1}^{j-1} \exp\left(\mathrm{i}\pi S^z_k\right) \left(S^z_j\right)^n \right\rangle \\
&= \frac{(-1)^\alpha}{(2S+1)!} \sum_{p,q=1}^{S+1} {}_S\mathrm{C}_{p-1}\, {}_S\mathrm{C}_{q-1} (q-p)^n (S+p-q)!(S-p+q)! \\
&\quad \times (-1)^p \qquad \text{for } n \in 2\mathbb{Z}+1
\end{aligned}$$

and

$$\langle\widetilde{\mathrm{VBS}}|\left(S_j^z\right)^n|\widetilde{\mathrm{VBS}}\rangle = \frac{1}{(2S+1)!}\sum_{p,q=1}^{S+1} {}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}(q-p)^n(S+p-q)!(S-p+q)!$$

for $n \in 2\mathbb{Z}$

we finally obtain the following results:

(i) For the S=1 case:

$$(\widetilde{\mathrm{Prob}}(S^z=1), \widetilde{\mathrm{Prob}}(S^z=0), \widetilde{\mathrm{Prob}}(S^z=-1)) = \begin{cases} \left(\frac{2}{3}, \frac{1}{3}, 0\right) & \text{for left} = \uparrow \\ \left(0, \frac{1}{3}, \frac{2}{3}\right) & \text{for left} = \downarrow \end{cases} \tag{30}$$

(i.e. $U|\mathrm{VBS}\rangle = |\widetilde{\mathrm{VBS}}\rangle$ is ferromagnetic).

(ii) For $S \in 2\mathbb{Z}$:

$$\widetilde{\mathrm{Prob}}(S^z=m) = \frac{1}{2S+1} \quad (=\mathrm{Prob}(S_j^z=m))$$

(it depends on neither the value of S^z nor the edge states).

They suggest that the Kennedy–Tasaki unitary transformation has no effect on the probability distribution of S_j^z for even S, that is, it transforms the (Z_2-symmetric) VBS state to a *Z_2-symmetric* state. Hence the string order parameter vanishes. For odd-S ($S \geqslant 3$) cases, it converts the VBS state into a Z_2-asymmetric state according to the left edge state, though $U|\mathrm{VBS}\rangle$ is no longer ferromagnetic. This is why the string order parameter takes a non-zero value when S = odd.

The generalized string order parameter

$$\mathcal{O}_{\mathrm{string}}(\theta) \equiv \lim_{L\nearrow\infty}\lim_{|i-j|\nearrow\infty}\frac{1}{\mathcal{N}}\left\langle S_i^z\prod_{k=i}^{j-1}\exp\left(\mathrm{i}\theta S_k^z\right)S_j^z\right\rangle \tag{31}$$

is easily calculated using the asymptotic orthogonality of the VBS state described in section 2. Note that the following relation:

$$\left\langle S_i^z\prod_{k=i}^{j-1}\exp\left(\mathrm{i}\theta S_k^z\right)S_j^z\right\rangle = \langle V_i^{z\dagger}V_j^z\rangle$$

holds for $V_j^z = \prod_{k=1}^{j-1}\exp(\mathrm{i}\theta S_k^z)S_j^z$. By definition (5), the string order parameter is given by the square of $\langle V_j^z\rangle$. Then, all we have to compute here is $\langle V_j^z\rangle$. Using matrices summarized in appendix A, we obtain

$$\lim_{j.L\nearrow\infty}\langle V_j^z\rangle = \frac{1}{(2S+1)!}\mathrm{e}^{-\mathrm{i}\alpha\theta}\sum_{p=1}^{S+1}\mathrm{e}^{\mathrm{i}p\theta}\sum_{q=1}^{S+1}(q-p)\,{}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}(S+p-q)!(S-p+q)!. \tag{32}$$

The following identities (which can be proved by elementary combinatorics)

$$\sum_{q=1}^{S+1} {}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}(S+p-q)!(S-p+q)! = \frac{(2S+1)!}{S+1}$$

$$\sum_{q=1}^{S+1}(q-1)\,{}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}(S+p-q)!(S-p+q)! = (2S+1)!\frac{S(S+2-p)}{(S+1)(S+2)}$$

enable us to simplify the right-hand side further. Thus, we finally arrive at the following expression:

$$\lim_{j,L\nearrow\infty}\langle V_j^z\rangle = \begin{cases} \dfrac{2\mathrm{i}}{S+2}\mathrm{e}^{-\mathrm{i}\alpha\theta}\mathrm{e}^{\mathrm{i}[(S+2)/2]\theta}\displaystyle\sum_{k=0}^{(S-1)/2}(2k+1)\sin\left[\left(k+\frac{1}{2}\right)\theta\right] & \text{for } S\in 2\mathbb{Z}+1 \\ \dfrac{2\mathrm{i}}{S+2}\mathrm{e}^{-\mathrm{i}\alpha\theta}\mathrm{e}^{\mathrm{i}[(S+2)/2]\theta}\displaystyle\sum_{k=0}^{(S/2)}2k\sin(k\theta) & \text{for } S\in 2\mathbb{Z} \end{cases} \tag{33}$$

where the symbol α $(=1,\ldots,S+1)$ labels the left edge states. Note that the value of $\lim_{j\nearrow\infty}\langle V_j^z\rangle$ depends on the left edge states through the phase factor $\mathrm{e}^{-\mathrm{i}\alpha\theta}$. The string order parameter is given by the square of its modulus as follows:

$$\mathcal{O}^z_{\mathrm{string}}(\theta) = \begin{cases} \left\{\dfrac{4}{S+2}\displaystyle\sum_{k=0}^{(S-1)/2}\left(k+\frac{1}{2}\right)\sin\left[\left(k+\frac{1}{2}\right)\theta\right]\right\}^2 & \text{for } S\in 2\mathbb{Z}+1 \\ \left\{\dfrac{4}{S+2}\displaystyle\sum_{k=1}^{S/2}k\sin(k\theta)\right\}^2 & \text{for } S\in 2\mathbb{Z}\,. \end{cases} \tag{34}$$

After an elementary calculation, we obtain the aforementioned result (7). It always vanishes at $\theta=0$ corresponding to the fact that the ordinary correlators are exponentially decaying. Another node $\theta=\pi$ appears for even S. This reflects the above mentioned fact that the Kennedy–Tasaki transformation does not change the probability distribution of S^z values for even S. Because of the symmetry property of $\mathcal{O}^\alpha_{\mathrm{string}}(\theta)$ (equation (28)), the node at $\theta=\pi$ always exists if the number of nodes is even. Namely, the nodes $\theta=0,\pi$ are topologically stable while the location of the others may be variable. For several values of S, we give the profiles of $\mathcal{O}^z_{\mathrm{string}}(\theta)$ (figure 2). We can clearly see that they have a large peak near π/S (for $S=1$ and 2, the largest peak is exactly at π/S). The reason for this can be understood as follows. If we focus only on $S^z=S$ and $-S$ ('particles') and regard others as a *background*, an interval between two adjacent particles has a 'background charge' $-S$ (when they are both S) or zero (when they are S and $-S$). Therefore, insertion of the string operator $\prod_{k=i}^{j-1}\exp(\mathrm{i}(\pi/S)S_k^z)$ always keeps the correlation between 'particles' ferromagnetic. This is the picture of the string order in $S=1$ chains. For higher values of S, we should take into account correlations between other values of S^z, which are not so simple as S and $-S$ and are smaller in their modulus. Thus we have the largest peak around π/S. Since the left edge state appears in $\langle V_j^z\rangle$ as the phase factor $\mathrm{e}^{-\mathrm{i}\alpha\theta}$ $(\alpha=1,\ldots,S+1)$, we cannot distinguish the $(S+1)\times(S+1)$-fold degenerate ground states by the value of $\langle V_j^z(\theta=\pi)\rangle$. On the other hand, the choice $\theta=\pi/S$ assigns different factors $\mathrm{e}^{-\mathrm{i}\pi/S},\ldots,-\mathrm{e}^{-\mathrm{i}\pi/S}$ to $\langle V_j^z\rangle$ for the $(S+1)$ edge states.

On the basis of the above results, we may take $\mathcal{O}^\alpha_{\mathrm{string}}(\pi/S)$ as a good order parameter to detect the hidden order in higher-S chains, though we do not know a unitary transformation that transforms $S_i^z\prod_{k=i}^{j-1}\exp(\mathrm{i}(\pi/S)S_k^z)S_j^z$ to the ordinary ferromagnetic correlation. However, it should be stressed here that if we modify the weights of the g matrix the location of the largest peak can be changed. The detailed analysis of the correlations between several values of S^z tells us that the situation becomes quite simple if we consider only S and $-S$. The correlation between them is always maximized by choosing θ to satisfy the relation $\cos S\theta=-1$, namely $\theta=\pi/S$.

In relation to the analogy between the dimerized state and the Haldane state [38], there is a very suggestive fact about $\langle\mathcal{O}_{\mathrm{string}}(\theta)\rangle$. We consider the spin-$S$ completely dimerized

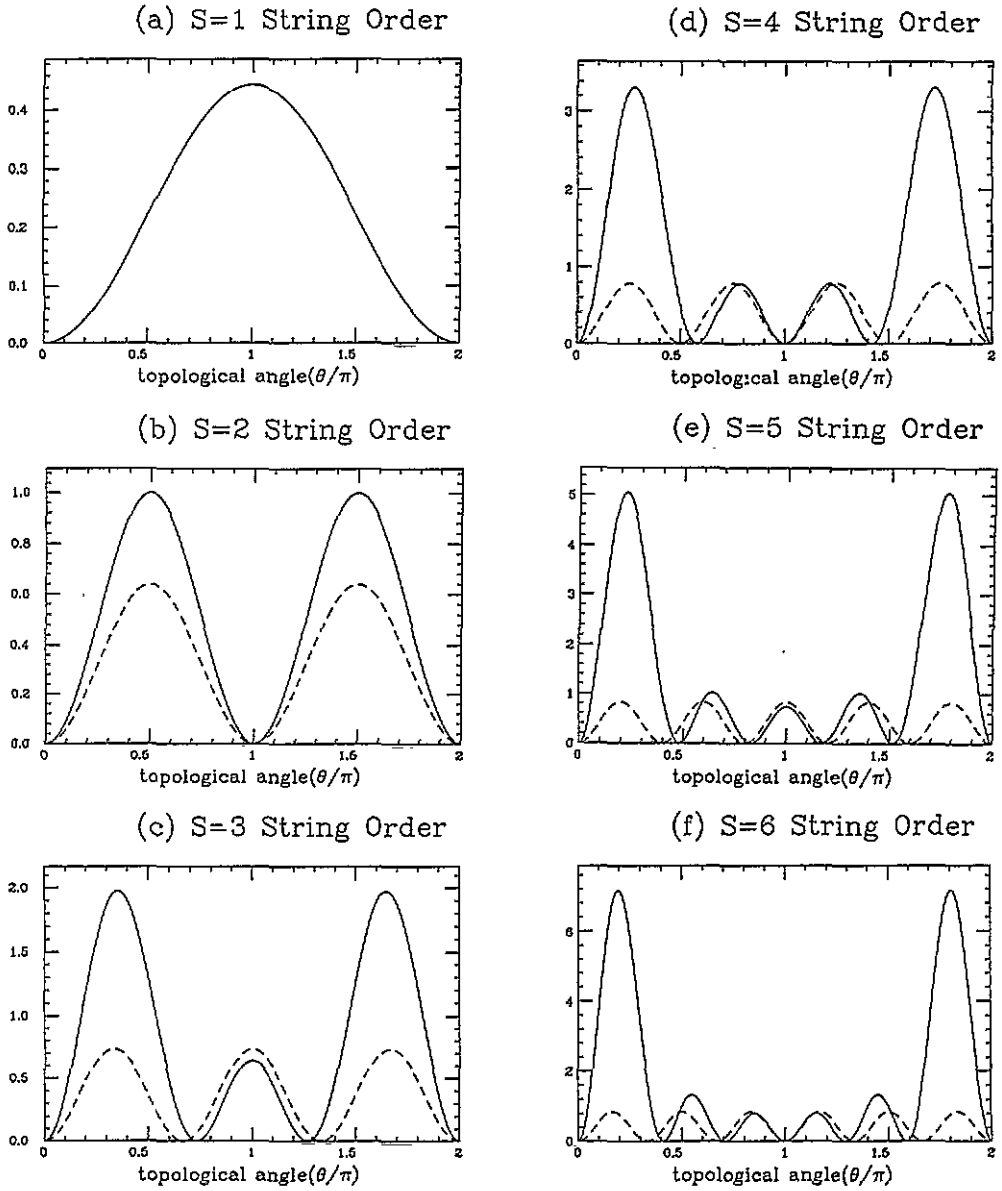


Figure 2. The profiles of a generalized string order parameter for several values of S. (a) $S = 1$. (b) $S = 2$. (c) $S = 3$. (d) $S = 4$. (e) $S = 5$. (f) $S = 6$. Here $\mathcal{O}^z_{\text{string}}(\theta)$ and $\mathcal{O}^z_{\text{mod}}(\theta)$ are plotted by solid lines and dashed ones, respectively. From these figures, we can clearly see that two large peaks exist around $\theta = \pm\pi/S$.

state:

$$|\text{dimer}\rangle = \prod_{i=1}^{N-1} (a^\dagger_{2i} b^\dagger_{2i+1} - b^\dagger_{2i} a^\dagger_{2i+1})^{2S} |0\rangle\rangle \otimes \ldots \otimes |0\rangle\rangle. \tag{35}$$

A direct calculation leads to the following result:

$$\langle (S^z_{2j-1}+S^z_{2j}) \exp\left(\mathrm{i}\theta \sum_{l=2j-1}^{2k-2} S^z_l\right)(S^z_{2k-1}+S^z_{2k})\rangle_{\text{dimer}}$$
$$= \begin{cases} \left(\dfrac{2}{2S+1}\right)^2 \left(\displaystyle\sum_{k=1}^{S} k \sin k\theta\right)^2 & \text{for } S \in \mathbb{Z} \\ \left(\dfrac{2}{2S+1}\right)^2 \left(\displaystyle\sum_{k=1}^{(2S-1)/2} \left(k+\frac{1}{2}\right) \sin\left(k+\frac{1}{2}\right)\theta\right)^2 & \text{for } S \in \mathbb{Z}+\tfrac{1}{2}. \end{cases} \tag{36}$$

Note that it has the same θ dependence as $\mathcal{O}^z_{\text{string}}(\theta)$ has for the spin-$2S$(!) VBS state (equation (34))†. Since, as mentioned above, the modification of the weights changes the θ-dependence, this result is non-trivial and may suggest the (long-range) dimer nature of the VBS state. In the language of the Markov process, the G matrix for the VBS state represents an ergodic chain and that of the dimer state corresponds to a sequence of independent events (a trivial Markov chain!). After a long interval, the former reduces to the latter. Thus, as long as we consider physics of a scale larger than the correlation length, the VBS state and the dimer state are essentially the same.

We can consider another order parameter which is parallel to the $S = 1$ string order parameter. From the preceding argument, it is obvious that we can obtain a 'ferromagnetic' correlation, if we concentrate only on $S^z = S$ or $-S$. In this viewpoint, it is natural to define a 'modified' string order parameter

$$\mathcal{O}^z_{\text{mod}}(\theta) = \lim_{|i-j|\nearrow\infty} \left\langle \mathcal{P}(S^z_i) \prod_{k=i}^{j-1} \exp\left(\mathrm{i}\theta S^z_k\right) \mathcal{P}(S^z_j) \right\rangle \tag{37}$$

where $\mathcal{P}(S^z) = S^z$ if $S^z = \pm S$ and $\mathcal{P}(S^z) = 0$ otherwise. The order parameter $\mathcal{O}^z_{\text{mod}}(\theta)$ can be written as a limiting case of the two-point function $\langle V^{z\dagger}_{\text{mod}\,i} V^z_{\text{mod}\,j}\rangle$ of the string-like operator

$$V^z_{\text{mod}\,j} = \prod_{k=1}^{j-1} \exp(\mathrm{i}\theta S^z_k)\mathcal{P}(S^z_j).$$

The same technique as before yields

$$\lim_{j\nearrow\infty} \langle V^z_{\text{mod}\,j}\rangle = -\left(\frac{2S}{2S+1}\right) \mathrm{i}\, \mathrm{e}^{-\mathrm{i}\alpha\theta} \mathrm{e}^{\mathrm{i}[(S+2)/2]\theta} \sin\left(\frac{S}{2}\theta\right) \tag{38}$$

and consequently

$$\mathcal{O}^z_{\text{mod}}(\theta) = \left(\frac{2S}{2S+1}\right)^2 \sin^2\left(\frac{S\theta}{2}\right) \qquad \text{for } S \in \mathbb{Z}. \tag{39}$$

Thus, $\mathcal{O}^z_{\text{mod}}(\theta)$ has S peaks at $\theta = (2m-1)\pi/S$ $(m = 1, \ldots, S)$ and, as is expected from the construction, it coincides with $\mathcal{O}^z_{\text{string}}(\theta)$ for $S = 1$. The value $\mathcal{O}^z_{\text{mod}}(\theta = \pi/S) = (2S/(2S+1))^2$ has a simple interpretation. Since the case $S^z = \pm S$ occurs with probability $1/(2S+1)$ (see equation (29)), $\mathcal{O}^z_{\text{mod}}(\theta = \pi/S)$ is computed also as $(\sum_{m=-S}^{S}[1/(2S+1)]|\mathcal{P}(m)|)^2 = (2S/(2S+1))^2$. We can easily evaluate $\mathcal{O}^z_{\text{mod}}(\theta = \pi/S)$ for the spin-S dimer state to obtain zero. Thus we can distinguish the VBS phase from the

† The only difference is that equation (34) is valid only for the long-distance limit, while equation (36) holds for any distances.

dimer phase. It is remarkable since $\mathcal{O}^z_{\text{string}}(\theta = \pi/S)$ does not vanish even in the spin-S dimer state owing to small shifts of the peaks.

In the large-S limit, $\mathcal{O}^z_{\text{mod}}(\pi/S)$ behaves like unity. This is to be compared with the fact that $\mathcal{O}^z_{\text{string}}(\pi/S)$ behaves like $(2/\pi)^2 S^2$. If the large-S VBS state has a Néel-like property, $\mathcal{O}^z_{\text{mod}}(\pi/S)$ is also $O(S^2)$. Thus we can see that the S^2 behaviour of $\mathcal{O}^z_{\text{string}}(\pi/S)$ is not a consequence of the classical nature‡. Note that even for higher values of S, $\mathcal{O}^z_{\text{mod}}(\pi/S)$ takes a non-zero value through the same mechanism as in the $S = 1$ case. The θ dependence $\sin^2(S\theta/2)$ in the above equation universally appears, whenever we take into account the contribution only from S and $-S$. This is the reason why $\sin^2(\theta/2)$ is unchanged even if we modify the weights of g in the $S = 1$ case, where S and $-S$ are the only non-zero S^z.

As was pointed out by [39], it is possible to construct VBS-type states which can be represented by the product of smaller matrices. For example, we can represent the $S = 2$ intermediate-D VBS state in terms of the product of the following 2×2 matrices [34]:

$$g_i = \begin{pmatrix} -2|0\rangle_i & -\sqrt{6}|1\rangle_i \\ \sqrt{6}|-1\rangle_i & 2|0\rangle_i \end{pmatrix} \tag{40}$$

where the states $|2\rangle$ and $|-2\rangle$ are excluded. Note that g_i can be obtained also by modifying the g matrix for the $S = 1$ VBS state. For this intermediate-D phase, the generalized string order parameter (31) is computed as

$$\mathcal{O}^z_{\text{string}}(\theta) = \frac{9}{25}\sin^2\frac{\theta}{2}. \tag{41}$$

Its θ dependence is the same as that of the $S = 1$ string order parameter $\mathcal{O}^z_{\text{string}}(\theta) = 4/9\sin^2(\theta/2)$. It is important to note that the generalized string order parameter for the $S = 2$ VBS state

$$\mathcal{O}^z_{\text{string}}(\theta) = \sin^2\theta$$

given by equation (34) has a node at $\theta = \pi$ and hence has a θ dependence completely different from that of equation (41). This implies that the generalized string order parameter (31) can distinguish the Haldane phase from the intermediate-D phase. Recent numerical calculations by Nishiyama *et al* [23] show that $\mathcal{O}^z_{\text{string}}(\theta = \pi) = 0$ at the $S = 2$ Heisenberg point, while $\mathcal{O}^z_{\text{string}}(\theta = \pi/2) \neq 0$ there. Hence the ground state of the $S = 2$ Heisenberg model does not belong to the intermediate-D phase. Thus the ground state is expected to be characterized by the set of the two string order parameters $\mathcal{O}^z_{\text{string}}(\pi)$ and $\mathcal{O}^z_{\text{string}}(\pi/S)$ for even S.

5. Low-lying excitations

Recently, Fáth and Sólyom [29] postulated that the elementary excitation of the $S = 1$ VBS model is a triplet 'defect' in the valence-bond solid called a 'crackion'. It has spin 1 and creates a domain wall in the string order.

In this section, we will show that this picture can be generalized to the higher-S VBS models and that it might give some keys to understanding the nature of low-lying excitations in the integer-S chains [8].

‡ The 'classical' limit $S \to \infty$ is subtle for the VBS models. For example, a naive $S \to \infty$ limit in the ordinary S^z correlation function yields $(-1)^{|i-j|}S^2/3$ (Néel-like!). From this, one may conclude that the $S \nearrow \infty$ limit of the VBS model is the classical Néel state. However, since the relation $\langle S^x_i S^x_j\rangle = \langle S^y_i S^y_j\rangle = \langle S^z_i S^z_j\rangle$ holds even in this limit, it is not classical.

First we briefly recapitulate the conjectured structure of the spectrum. According to Haldane [5, 6], the elementary excitation of the integer-S chain is a massive triplet while the generic half-odd-integer S chains have a massless *doublet* excitation. That is, the elementary excitation of the latter case is that of the $S = \frac{1}{2}$ Heisenberg model—a spin-$\frac{1}{2}$ soliton first pointed out by Faddeev and Takhtajan [40, 41]. For the half-odd-integer case, a rigorous argument [42] and the exact (Bethe *ansatz*) solution [2] suggest the naturalness of such an excitation. Since we can explicitly verify that such a picture breaks down for the VBS model (see appendix B), we need an alternative simple model of the elementary excitation for the integer-S case.

The crackion is defined by replacing a singlet valence-bond operator by a triplet operator [28]

$$(\alpha a_i^\dagger + \beta b_i^\dagger)(\alpha a_{i+1}^\dagger + \beta b_{i+1}^\dagger) \tag{42}$$

where α, β are auxiliary variables that label S^z values ($\alpha^2, \alpha\beta, \beta^2$ label $S^z = 1, 0, -1$, respectively). Apparently, the system has spin 1 in the presence of a crackion (for simplicity we consider the case of a periodic boundary condition). Knabe treated it variationally, and obtained a finite gap (of course, within the approximation). In a recent paper, Fáth and Sólyom [29] considered a 'moving' crackion and showed that its spectrum is in relatively good agreement with the numerical results. They also postulated that a crackion plays a kink-like role in the ferromagnetic state generated by the Kennedy–Tasaki transformation.

Before we generalize this to the higher-S cases, we must recall that the Kennedy–Tasaki transformation fails to convert the VBS state to a ferromagnetic state (even for the odd-S case!) as shown in the previous section. Therefore, the kink nature of a crackion is obscure in the S^z configuration itself. However, it can be observed if we consider the expectation value of the string-like operator V_j^z.

We begin with the g-matrix representation of the crackion configurations. The boson representation of the crackion is

$$|\Psi_j^a\rangle = (\ldots)(a_{j-1}^\dagger b_j^\dagger - b_{j-1}^\dagger a_j^\dagger)^S\, T^a(a_j^\dagger, b_j^\dagger a_{j+1}^\dagger, b_{j+1}^\dagger)\,(a_{j+1}^\dagger b_j^\dagger - b_j^\dagger a_{j+1}^\dagger)^{S-1}(\ldots)|0\rangle\rangle \tag{43}$$

where $T^a(a_i^\dagger, b_i^\dagger, a_{i+1}^\dagger, b_{i+1}^\dagger)$ means triplet operators

$$T^1 = a_j^\dagger a_{j+1}^\dagger \qquad T^0 = (a_j^\dagger b_{j+1}^\dagger + b_j^\dagger a_{j+1}^\dagger) \qquad T^{-1} = b_j^\dagger b_{j+1}^\dagger.$$

According to the prescription described in section 2, the g matrices for these configuration are easily found as

$$g_j^1(p,q) = (-1)^{S-p+1}\sqrt{{}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}}\sqrt{(S-p+q+1)!(S+p-q-1)!} \times \frac{S+1-q}{S}|q-p+1\rangle_j \tag{44}$$

$$g_j^0(p,q) = (-1)^{S-p+1}\sqrt{{}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}}\sqrt{(S-p+q)!(S+p-q)!} \times \frac{2(q-1)-S}{S}|q-p\rangle_j \tag{45}$$

$$g_j^{-1}(p,q) = (-1)^{S-p+1}\sqrt{{}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}}\sqrt{(S-p+q-1)!(S+p-q+1)!} \times \frac{q-1}{S}|q-p-1\rangle_j. \tag{46}$$

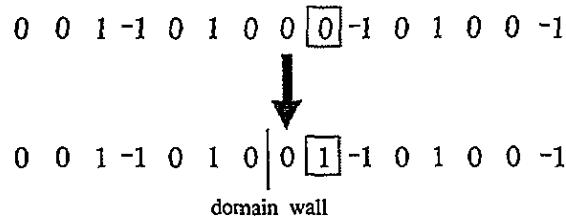
For example, the $S = 1$ crackions are expressed by the following matrices:

$$g_j^1 = \begin{pmatrix} \sqrt{2}|1\rangle_j & 0 \\ -|0\rangle_j & 0 \end{pmatrix} \quad g_j^0 = \begin{pmatrix} -|0\rangle_j & \sqrt{2}|1\rangle_j \\ \sqrt{2}|-1\rangle_j & -|0\rangle_j \end{pmatrix} \quad g_j^{-1} = \begin{pmatrix} 0 & -|0\rangle_j \\ 0 & \sqrt{2}|-1\rangle_j \end{pmatrix}.$$

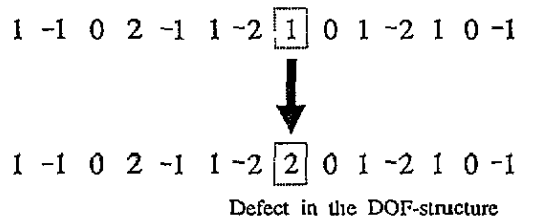
Using them, we can represent the crackion $|\Psi_j^a\rangle$ as

$$|\Psi_j^a\rangle = g^{\text{start}} \otimes g_2 \otimes \ldots \otimes g_{j-1} \otimes g_j^a \otimes g_{j+1} \otimes \ldots \otimes g_L. \tag{47}$$

For the $S = 1$ case, as is easily verified, each $+1$ or -1 crackion creates a domain wall somewhere around the site j (its precise location is obscured owing to the liquid-like nature of the VBS state) (see figure 3(a)). Since for the higher-S cases the meaning of the string order becomes ambiguous, we have to use the term 'domain wall' in a generalized sense.



(a)



(b)

Figure 3. *The effect of the $+1$ crackion. It creates a 'domain wall' in the perfect string order but its precise location is implicit.* (a) The $S = 1$ VBS case. (b) The $S = 2$ VBS case.

Using the above matrices, we can evaluate the expectation value of V^z in the presence of crackions. For the case of a single crackion, what we have to compute is

$$\langle \Psi_j^a; \alpha, \beta | V_i^z | \Psi_j^a; \alpha, \beta \rangle$$
$$= \begin{cases} \frac{1}{N} G_{\text{start}}^{e^{i\pi S}} (G^{e^{i\pi S}})^{j-2} (G_{\text{crack}}^{e^{i\pi S}}) (G^{e^{i\pi S}})^{i-j-1} G^{S^z} (G)^{L-i} & \text{for } i > j \\ \frac{1}{N} G_{\text{start}}^{e^{i\pi S}} (G^{e^{i\pi S}})^{i-2} G^{S^z} (G)^{j-i-1} G_{\text{crack}} (G)^{L-j} & \text{for } i < j. \end{cases}$$

With the help of the asymptotic orthogonality, we can evaluate it for the long-distance limit to obtain

$$\langle V_i^z \rangle_{\text{1crackion}} = \begin{cases} \langle V_i^z \rangle_{\text{VBS}} e^{-ia\theta} & \text{for } i > j \\ \langle V_i^z \rangle_{\text{VBS}} & \text{for } i < j. \end{cases} \tag{48}$$

Note that $\langle V_i^z \rangle$ is multiplied by an extra phase factor $e^{-ia\theta}$ if a crackion is located to the left of V_i^z. It is easy to generalize this result to multicrackion cases. The desired formula is

$$\langle V_i^z \rangle_{\text{multicrackion}} = \langle V_i^z \rangle_{\text{VBS}} e^{-i\theta \sum a_k} \tag{49}$$

where $\sum a_k$ counts a 'crackion charge' between site 1 and site i. If we choose $\theta = \pi$, this is a Z_2 charge and adding a $+1$ or -1 crackion flips the sign of $\langle V^z \rangle$ for odd S. Of course,

if we observe the string operator in the x direction, $\langle V_i^x \rangle$, a 0 crackion also flips the sign. Thus the three crackions form a triplet of a topological excitation.

It is straightforward to generalize this result to the higher-S cases using the higher-S version of the crackion matrices (44)–(46). The result is the same as in the $S = 1$ case and is given by equation (49). Therefore, we obtain the expectation value of the string correlator in the presence of the crackions as follows:

$$\langle V_i^{z\dagger} V_j^z \rangle_{\text{multicrackion}} = \langle \mathcal{O}^z_{\text{string}} \rangle_{\text{VBS}} \exp\left(-\mathrm{i}\theta \sum_{k=i}^{j} a_k \right). \tag{50}$$

When $\theta = \pi$, this quantity alternates its sign according to the parity of $\sum_{k=i}^{j} a_k$ (Z_2 charge).

Taking into account the multicrackion configurations, we can approximately estimate the temperature dependence of the string correlator. Following Fáth and Sólyom [29], we adopt the 'moving' crackion

$$|\Psi^a(k)\rangle = \frac{1}{\sqrt{L}} \sum_{x=1}^{L} \mathrm{e}^{-\mathrm{i}kx} |\Psi_x^a\rangle \tag{51}$$

as an elementary excitation (where the relation between a moving crackion and an elementary excitation in the SMA picture is discussed below). Since $|\Psi_j^a\rangle$ is obtained by replacing g_j in the matrix product of the VBS state by the crackion matrix g_j^a, we can regard $|\Psi^a(k)\rangle$ as the matrix version of a spin-wave state. For the moment, we restrict ourselves to the $S = 1$ case. The first step is to approximate the crackions by the gas of hard-core bosons [44–46] (or free fermions, equivalently: [46] discusses a fermionic picture of elementary excitations; see also [47] and references cited therein). For low enough temperatures, it is sufficient to consider low-energy excitations around a momentum $k = \pi$. That is, we approximate the crackions by free fermions with the following dispersion relation (namely an approximate expression of equation (61)):

$$\epsilon^a(k) = \Delta_{\text{H}} + \alpha(k - \pi)^2 \qquad (\Delta_{\text{H}} = \tfrac{20}{27}, \alpha = \tfrac{5}{9}). \tag{52}$$

Furthermore, we assume that the density of crackions is not very high and hence we can neglect the intercrackion interaction (which decays exponentially with a correlation length $\sim 1/\ln 3$). This is reminiscent of the well known dilute-instanton-gas approximation. This assumption may be justified at sufficiently low temperatures. Now we divide the calculation into two parts, that is, the computation of the expectation value of $\mathcal{O}_{\text{string}}$ in the N-crackion configurations and the summation over all configurations. Neglecting the sum over the plane-wave factors, we can calculate $\langle \{k_i\} | \mathcal{O}_{\text{string}} | \{k_i\} \rangle$ as $(1 - 2p_{\text{crack}})^{N_{+1}+N_{-1}}$ (N_a denotes number of the crackions of species a). With the help of the familiar technique in elementary statistical mechanics, we finally arrive at the following desired result† (note that among the three crackions only two contribute to the reduction of the string order):

$$\langle \mathcal{O}_{\text{string}} \rangle_\beta = \frac{4}{9} \exp\left[-\frac{2}{\sqrt{\pi\alpha}} \frac{1}{\sqrt{\beta}} p_{\text{crack}} \mathrm{e}^{-\beta\Delta_{\text{H}}} \right] \quad \left(\beta \equiv \frac{1}{k_{\text{B}} T} \right). \tag{53}$$

In the above expression, the quantity p_{crack} is related to the probability of finding a single crackion in the interval between the two end-points (i and j) of $\mathcal{O}_{\text{string}}$, and is of the order $|i - j|/L$. Its precise form can be evaluated from the expression of $\langle k | \mathcal{O}_{\text{string}} | k \rangle$. For the $p_{\text{crack}} \sim 1$ case, it decreases rapidly as the temperature increases above Δ_{H}. On the other hand, it is almost constant for a small enough value of p_{crack}. Namely, the long-range

† In this expression, we have kept only the lowest-order terms. A more complete expression including higher-order terms is written using the Appell function.

string order decays at finite temperatures, while the short-range one is robust. This is in agreement with the recent Monte Carlo result [26]. Note that the domain-wall property of the elementary excitations of the $S = 1$ Heisenberg chain recently argued by several authors [31, 32] is naturally explained by the crackion picture.

To conclude this section, we point out the *elementary* nature of the crackions. Fáth and Sólyom [29] showed that the action of magnon operators can be written in terms of crackions. A little calculation shows that for the spin-S case, we have

$$S_j^+|\text{VBS}\rangle = S\left(|\Psi_{j-1}^{+1}\rangle - |\Psi_j^{+1}\rangle\right) \tag{54}$$

$$S_j^z|\text{VBS}\rangle = -(S/2)\left(|\Psi_{j-1}^{0}\rangle - |\Psi_j^{0}\rangle\right) \tag{55}$$

$$S_j^-|\text{VBS}\rangle = -S\left(|\Psi_{j-1}^{-1}\rangle - |\Psi_j^{-1}\rangle\right). \tag{56}$$

From these relations, it follows that crackions have an elementary nature for all integer-S VBS models. Furthermore, it can be shown that the energy of a moving crackion is equal to $\omega_{\text{SMA}}(k)$ (the spectrum obtained by the single-mode approximation). The proof is quite simple. Within the single-mode approximation (SMA), an approximate dispersion relation is given by

$$\omega_{\text{SMA}}(k) = -\frac{1}{2}\frac{\langle\text{VBS}|\,[\,[\mathcal{H}, S^z(k)], S^z(-k)\,]\,|\text{VBS}\rangle}{\langle\text{VBS}|S^z(k)S^z(-k)|\text{VBS}\rangle}. \tag{57}$$

Using the elementary property of the crackions, the numerator can be rewritten as

$$\begin{aligned}
(\text{numerator}) &= \frac{1}{2}2\langle\text{VBS}|S^z(k)\mathcal{H}S^z(-k)|\text{VBS}\rangle \\
&= \frac{1}{L}\left(\frac{S}{2}\right)^2\sum_{x,y=1}^{L} e^{ik(x-y)}\left(\langle\Psi_{x-1}^0| - \langle\Psi_x^0|\right)\mathcal{H}\left(|\Psi_{y-1}^0\rangle - |\Psi_y^0\rangle\right) \\
&= \frac{1}{L}\left(\frac{S}{2}\right)^2\sum_{x,y=1}^{L} e^{ik(x-y)}2(1-\cos k)\,\langle\Psi_x^0|\mathcal{H}|\Psi_x^0\rangle\delta_{x,y} \\
&= (S/2)^2\,2(1-\cos k)\,\langle\Psi^0(k)|\mathcal{H}|\Psi^0(k)\rangle.
\end{aligned} \tag{58}$$

In rewriting the second line to the third one, we have changed the dummy indices.

In a similar manner, we can rewrite the denominator (the static structure factor)

$$\langle\text{VBS}|S^z(k)S^z(-k)|\text{VBS}\rangle = (S/2)^2\,2(1-\cos k)\,\langle\Psi^0(k)|\Psi^0(k)\rangle. \tag{59}$$

By the rotational symmetry, the above results hold also for $+1$ and -1 crackions. Combining equations (58) and (59), we arrive at the following equality:

$$\omega_{\text{SMA}}(k) = \epsilon_{\text{crack}}(k) \equiv \frac{\langle\Psi^a(k)|\mathcal{H}|\Psi^a(k)\rangle}{\langle\Psi^a(k)|\Psi^a(k)\rangle}. \tag{60}$$

This equality was first pointed out by Fáth and Sólyom for the $S = 1$ case [29].

It is straightforward to evaluate the RHS of (60). After some algebra using the Clebsch–Gordan coefficients, we obtain

$$\epsilon_{\text{crack}}(k) = \omega_{\text{SMA}}(k) = \frac{50 + 30\cos k}{27} \qquad \text{for } S = 1 \tag{61}$$

$$\epsilon_{\text{crack}}(k) = \omega_{\text{SMA}}(k) = \frac{5 + 4\cos k}{5} \qquad \text{for } S = 2 \tag{62}$$

$$\epsilon_{\text{crack}}(k) = \omega_{\text{SMA}}(k) = \frac{132(17 + 15\cos k)}{6265} \qquad \text{for } S = 3 \tag{63}$$

$$\epsilon_{\text{crack}}(k) = \omega_{\text{SMA}}(k) = \frac{715(13 + 12\cos k)}{90318} \qquad \text{for } S = 4 \tag{64}$$

etc. The expression ω_{SMA} for the $S = 1$ VBS model was first obtained by Arovas *et al* [15]. Since the recent numerical calculation [46, 30] shows that the SMA is a good approximation at least near $k = \pi$, we may expect that the above results also reproduce the true values fairly well. Within our approximation, the dispersion relations for $S = 2$, 3, and 4 are qualitatively the same as for the $S = 1$ case.

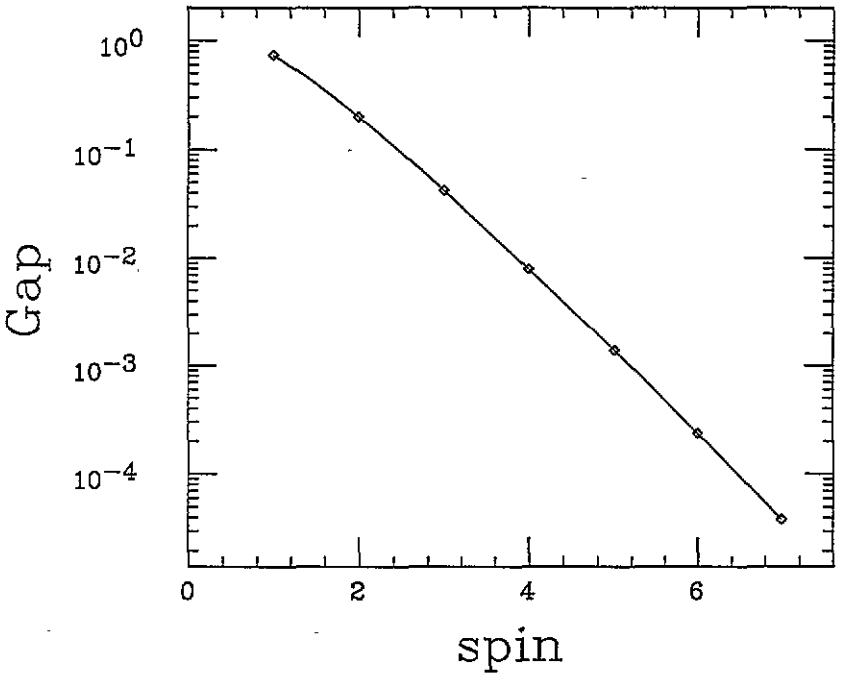


Figure 4. A logarithmic plot of the Haldane gap for several values of S. The gap is obtained by the single-mode approximation (SMA). It exhibits an evident linearity, which implies that the σ-model prediction holds also in our VBS case.

The approximate value of the Haldane gap Δ_H for the VBS model is listed in table 1 for $S = 1, \ldots, 6$. According to the general theory of the SMA, the true gap is smaller than this value. A logarithmic plot of the gaps (see figure 4) shows that the large-S behaviour of the gaps is well reproduced by the famous formula

$$\Delta = \text{constant} \times e^{-\alpha S}.$$

In our case, α seems to be 1.7–1.8 (precisely, using data for $S = 4, 5, 6, 7$, we obtain the value $\alpha = 1.78$: since there may be an S-dependent prefactor, this value must be regarded as a crude estimate), which is different from the well known σ-model prediction [5, 11] $\alpha = \pi$. A remark is in order here. Using the method of Affleck [49], we can map the spin-S VBS model to the O(3) non-linear σ model with an S-dependent prefactor. The trouble is that this prefactor can take a negative value for some S. From this reason, we can not completely rely on this mapping in our case. However, if we take into account the renormalization effect of the coupling constant (the bare one is $2/S$), this discrepancy may be resolved. Thus, we believe that the VBS model also belongs to a class of models which are effectively described by the O(3) non-linear σ model (without the Π_2 topological term). Recent numerical results [50] suggest that the only differences between the true Heisenberg model and the VBS model are a prefactor of the exponential and the renormalized coupling constant.

We expect that this crackion picture gives an important clue to understanding the elementary excitations in integer-S chains.

Table 1. The gap estimated using the SMA for several values of S. The Haldane gap decreases exponentially as the value of S increases.

Spin	Gap (SMA)	(Approximation)
1	$\frac{20}{27}$	7.41×10^{-1}
2	$\frac{1}{5}$	2.00×10^{-1}
3	$\frac{264}{6265}$	4.21×10^{-2}
4	$\frac{715}{90318}$	7.92×10^{-3}
5	$\frac{884}{634711}$	1.39×10^{-3}
6	$\frac{282625}{1203274644}$	2.35×10^{-4}

6. Discussion

In the preceding sections, we gave a simple method of expressing the valence-bond- (VB-) type states in terms of a simple matrix product. In our formalism, the edge states which naturally arise in the VB-type states correspond to the matrix indices, and the size of the g matrix is determined by the number of VBs on a given link. From this viewpoint, the matrix-product representation is a natural expression for the VB-type states. It is important that the size of the g matrices is always smaller than $2S+1$ reflecting the VB nature of the states.

A generalization of our method to other VB-type states is straightforward. For example, the partially dimerized state (where the number of VBs alternates in links) is expressed by two non-square matrices g^{A} and g^{B} as

$$g_1^{\mathrm{A}} \otimes g_2^{\mathrm{B}} \otimes g_3^{\mathrm{A}} \otimes g_4^{\mathrm{B}} \otimes \dots .$$

(see appendix A of [34] for other examples).

It is also possible to generalize it in another direction. Starting from the matrix-product representation of a given state, we can construct its anisotropic version (preserving the $U(1)$ symmetry) by modifying the weights of the matrix elements. However, the Hamiltonian whose exact ground state is given by this is not so simple [25, 37, 34]. Recently, Zittartz constructed such Hamiltonians for several values of S [39]. Our method may give a starting point in such an approach. We can also use such modified matrices as trial functions for the realistic models. For example, an $S=1$ modified g matrix

$$g_j(\Theta) = \begin{pmatrix} -\cos\Theta|0\rangle_j & -\sin\Theta|1\rangle_j \\ \sin\Theta|-1\rangle_j & \cos\Theta|0\rangle_j \end{pmatrix}$$

serves as a trial state for the ground state of the $S=1$ XXZ chain with the D term [31]. It interpolates between the large-D phase ($\Theta = 0$ or π) and the Néel phase ($\Theta = \pi/2$). The variational parameter θ changes the concentration of zeros.

In the preceding sections, we have investigated hidden structures in the VBS model. Our calculation was motivated by a recently observed [23] ninefold (near) degeneracy in the ground state of the $S = 2$ Heisenberg model, which implies that the $S = 2$ VBS model may also be a good approximation to the true Heisenberg model. As was shown by Kennedy and Tasaki, we can *relate* the fourfold (near) degeneracy found in the $S = 1$ Haldane systems to the breakdown of the hidden $Z_2 \times Z_2$ symmetry (or non-vanishing string order parameters $\mathcal{O}^x_{\text{string}}(\pi)$ and $\mathcal{O}^z_{\text{string}}(\pi)$). However, this picture is not successful in higher-S cases. Furthermore, there is a model which exhibits the Haldane gap without breaking the full $Z_2 \times Z_2$ symmetry. Therefore, we modestly consider the string order parameter as a probe that distinguishes the Haldane phase(s) from other more trivial massive phases. At present, we do not know whether there exists a unitary transformation that relates order parameters and a hidden symmetry for higher-S cases. Since our results imply a resemblance between the spin-S dimer state and the spin-$2S$ VBS state, the dimer picture seems to be successful for general spin-S cases.

Acknowledgments

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Appendix A. Relevant G matrices

In this appendix, we summarize some G matrices necessary for our calculation.

(i) G matrix:

$$G(a,b;c,d) = (-1)^{a+b}\sqrt{{}_S\mathrm{C}_{a-1}\,{}_S\mathrm{C}_{b-1}\,{}_S\mathrm{C}_{c-1}\,{}_S\mathrm{C}_{d-1}} \times\sqrt{(S+a-c)!(S-a+c)!(S+b-d)!(S-b+d)!}\,\delta_{a-b,c-d} \tag{A1}$$

(unreduced form)

$$(G_{\text{Red}})_{(p,q)} = G(p,p;q,q) = G^{\text{start}}(p,p;q,q) = {}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}(S+p-q)!(s-p+q)! \tag{A2}$$

(where G and G_{Red} are symmetric).

We list a few G matrices:
(a) $S = 1$ VBS state

$$G = \begin{pmatrix} 1 & 0 & 0 & 2 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 2 & 0 & 0 & 1 \end{pmatrix} \qquad G_{\text{Red}} = \begin{pmatrix} 1 & 2 \\ 2 & 1 \end{pmatrix}$$

(b) $S = 2$ VBS state

$$G = \begin{pmatrix} 4 & 0 & 0 & 0 & 12 & 0 & 0 & 0 & 24 \\ 0 & -8 & 0 & 0 & 0 & -12 & 0 & 0 & 0 \\ 0 & 0 & 4 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -8 & 0 & 0 & 0 & -12 & 0 \\ 12 & 0 & 0 & 0 & 16 & 0 & 0 & 0 & 12 \\ 0 & -12 & 0 & 0 & 0 & -8 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 4 & 0 & 0 \\ 0 & 0 & 0 & -12 & 0 & 0 & 0 & -8 & 0 \\ 24 & 0 & 0 & 0 & 12 & 0 & 0 & 0 & 4 \end{pmatrix} \qquad G_{\mathrm{Red}} = \begin{pmatrix} 4 & 12 & 24 \\ 12 & 16 & 12 \\ 24 & 12 & 4 \end{pmatrix}.$$

(ii) $G_{\mathrm{Red}}^{\mathrm{e}^{i\theta S}}$:

$$\begin{aligned}(G_{\mathrm{Red}}^{\mathrm{string}})_{(p,q)} &= g^{\dagger}(p,q)\,\mathrm{e}^{i\theta S^z}\,g(p,q) \\ &= {}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}(S+p-q)!(s-p+q)!\mathrm{e}^{\mathrm{i}(q-p)\theta}. \end{aligned} \tag{A3}$$

(iii) $G_{\mathrm{Red}}^{\mathrm{e}^{i\theta S}S^z}$:

$$\begin{aligned}(G_{\mathrm{Red}}^{\mathrm{e}^{i\theta S}S^z})_{(p,q)} &= g^{\dagger}(p,q)\,S^z\mathrm{e}^{i\theta S^z}\,g(p,q) \\ &= (q-p)_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}(S+p-q)!(s-p+q)!\mathrm{e}^{\mathrm{i}(q-p)\theta}. \end{aligned} \tag{A4}$$

(iv) $G_{\mathrm{Red}}^{P(S^z=m)}$ ($P(S^z=m)$ denotes a projection operator):

$$\begin{aligned}(G_{\mathrm{Red}}^{P(S^z=m)})_{(p,q)} &= g^{\dagger}(p,q)\,P(S^z=m)\,g(p,q) \\ &= \delta_{p+m,q}\,{}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}(S+p-q)!(S-p+q)!. \end{aligned} \tag{A5}$$

In fact, G_{Red} and $G_{\mathrm{Red}}^{\mathrm{string}}$ are related by the following unitary transformation:

$$U(\theta) = \mathrm{diag}(\mathrm{e}^{i\theta}, \mathrm{e}^{2i\theta}, \ldots, \mathrm{e}^{\mathrm{i}(S+1)\theta})$$

and hence have the eigenvalues in common.

It is not so easy to compute exactly $(G_{Red})^n$ or $(G_{\mathrm{Red}}^{\mathrm{e}^{i\theta S}})^n$ for general S. However, their asymptotic ($n \gg 1$) form is calculable because of the asymptotic orthogonality. The result is

$$(G_{\mathrm{Red}}^{\mathrm{e}^{i\theta S}})^n_{(p,q)} \to \frac{1}{S+1}\left[\frac{(2S+1)!}{S+1}\right]^n \mathrm{e}^{\mathrm{i}(q-p)\theta}. \tag{A6}$$

An easy way of obtaining this result is to use the well known result in the theory of the Markov process. Since $[(S+1)/(2S+1)!]G_{\mathrm{Red}}$ denotes a double stochastic matrix with an ergodic property, all the matrix elements are the same in its asymptotic form. Thus we obtain (A6).

Using this result, it is possible to show that we can reduce the infinite-volume expectation values to those of a *finite* volume (spin-S version of lemma 2.6 of [10]).

Of course, since $\lambda(0)/\lambda(1) \to 1$ in the $S \to \infty$ limit, the above argument is invalid and we have to consider all the eigenvalues. Accordingly, the above-obtained expressions of the string order parameters are valid only for finite S.

Appendix B. Relation to the Lieb–Schultz–Mattis theorem

In [51], Lieb, Schultz and Mattis (LSM) proved the famous Lieb–Schultz–Mattis theorem. More than twenty years later, Affleck and Lieb generalized the idea of LSM to prove the existence of a gapless excitation in generic XXZ-type spin chains with half-odd-integral S [42].

The main idea of Affleck and Lieb is as follows. First we assume that the finite-chain ground state of $\mathcal{H}_{XXZ}$ (i) is unique and (ii) satisfies $S^z_{tot} = 0$. Then a moderate twist realized by a unitary transformation

$$U = \exp\left(\mathrm{i}\frac{\pi}{l}\sum_{j=-l}^{l} j\, S^z_j\right)$$

creates a state $U|\mathrm{GS}\rangle$, which has an energy higher than that of $|\mathrm{GS}\rangle$ by an order of $O(1/l)$. For half-odd-integer S, we can show that $\langle \mathrm{GS}|U|\mathrm{GS}\rangle = 0$ and the possibility of a unique infinite-volume ground state with a gap is excluded. Furthermore, we can show that $U|\mathrm{GS}\rangle$ has a momentum π and this is consistent with the fact that the low-lying excitation of the Bethe *ansatz*-solvable models [1, 4] [4] is gapless and k linear around $k = \pi$.

However, for integer S, we can not tell whether $|\mathrm{GS}\rangle$ and $U|\mathrm{GS}\rangle$ are different or not by their method.

In this appendix, we show that the transformed state $U|\mathrm{VBS}\rangle$ approaches the original state $|\mathrm{VBS}\rangle$ in the infinite-volume limit. This suggests the difference between the low-lying excitations of integer-S chains and those of half-odd-integer-S chains.

First we compute $U|\mathrm{VBS}\rangle$ and rewrite it in terms of the matrix product. After some algebra, we obtain

$$U|\mathrm{VBS}\rangle_{\mathrm{PBC}} = (\ldots)\prod_{k=-l}^{l-1}(a^\dagger_k b^\dagger_{k+1} - \mathrm{e}^{\mathrm{i}2\pi/l} b^\dagger_k a^\dagger_{k+1})^S(\ldots)|0\rangle\rangle$$

where the ellipsis denotes the usual valence-bond operators. The same calculation as in section 2 yields g matrices

$$\tilde{g}^{\mathrm{start}} = \left(\mathrm{e}^{\mathrm{i}2\pi/L}\right)^{S-p+1}\sqrt{{}_S\mathrm{C}_{p-1}\,{}_S\mathrm{C}_{q-1}}\sqrt{(S-p+q)!(S+p-q)!}|q-p\rangle_1. \tag{B1}$$

The argument about the reduced matrices in section 2 and appendix A is unchanged in the presence of the extra phase factors and equation (A6) applies also to this case. The evaluation of the overlap $\langle \mathrm{VBS}|U|\mathrm{VBS}\rangle$ is straightforward. All we need is the following G matrix:

$$G'(p,q,;r,s) = g^\dagger(p,r)\tilde{g}(q,s). \tag{B2}$$

Using this, the desired quantity is computed as

$$\langle \mathrm{VBS}|U|\mathrm{VBS}\rangle = \mathrm{Tr}(G')^L. \tag{B3}$$

Although the precise form of this is complicated, the asymptotic form is simple,

$$\langle \mathrm{VBS}|U|\mathrm{VBS}\rangle = -1 + \mathrm{O}(1/l). \tag{B4}$$

This implies that the ground state $|\mathrm{VBS}\rangle$ and the twisted state $U|\mathrm{VBS}\rangle$ are not orthogonal even for finite l. Therefore, in our special case we can conclude that the unitary transformation employed by Affleck and Lieb, when applied to the integer-S chains, does not create a different state which is orthogonal to the ground state. On the other hand, it is obvious that the excited states created by crackions are orthogonal to the ground state since they contain different quantum numbers S^z_{tot}.

Although the above fact does not exclude the possibility of a gapless excitation in the VBS models, our result suggests that such an excitation, if it exists, is not created by a 'spin-wave-like' twist.

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