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Fully variational resonance-valence-bond results on two-, three- and four-legged spin-1/2 Heisenberg ladders

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Abstract. A fully variational resonance-valence-bond (RVB) study is performed for two-, three- and four-legged isotropic spin-1/2 Heisenberg ladders under periodic boundary conditions along the rungs and periodic/twisted boundary conditions along the chains. By using the full lattice groups of these systems very accurate results have been obtained for the ground-state bulk limit properties in the case of the dimer RVB basis. The results reported here represent the best one can do within the dimer RVB *ansatz* and should be useful when comparing with results obtained from an *a priori* fixed structure of the wavefunction. Further results are presented for some finite-sized systems, extending beyond the dimer RVB approach by including RVB functions containing spin pairings between non-nearest-neighbour sites. These results indicate that the three-legged ladder with periodic boundary conditions along the rungs falls into the same universality class of ladders possessing short-range RVB wavefunctions as the two- and four-legged ladders.

1. Introduction

Over the last few years there has been an increasing theoretical and experimental interest in spin-1/2 ladders [1], due to recent discoveries of ladder-type superconducting-doped ceramic materials like $Sr_{0.4}Ca_{13.6}Cu_{24}O_{41}$ [2]. Spin-1/2 ladder structures are further present in compounds like $(VO)_2P_2O_7$ [3] and other cuprates such as $Sr_{n-1}Cu_{n+1}O_{2n}$ [4], the former one containing V^{4+} ions arranged in a two-legged ladder structure while the latter ones possess an $(1/2)(n+1)$-legged ladder configuration of Cu^{2+} ions. In all these materials, single electrons are donated from the transition metal ions, forming thus an arrangement or lattice structure of spin-1/2 particles interacting with each other. Ladders represent quasi-1D transition structures between the 1D chain and the 2D square-planar lattice, and their study is hence very important in understanding differences when moving from 1D to 2D systems arising due to quantum effects [1]. In this context it was found by extensive numerical calculations that ladders with odd numbers of legs are gapless in contrast to even-legged ones [5], i.e. they require no energy to be excited to the first excited level. All even-legged ladders possess in principle a finite energy gap Δ, starting from an apparently exact value of $\Delta = 1/2$ for the two-legged ladder [6] and decreasing exponentially in value the more closely the ladders approach the zero-gapped 2D square-planar limit.

When trying to obtain properties of infinite quantum systems, which unlike the 1D chain cannot be solved analytically, the usual way is to perform extrapolations on some finite-system results, which are obtainable either exactly by exact diagonalizations for small lattices or approximately by Monte Carlo approaches for bigger lattices. These methods, however,

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apart from inducing often significant extrapolation errors, provide almost no insight into the structure of the final wavefunction for the infinite system, making it very difficult to identify the importance of its different locally well defined substructures, which would then give us in turn directly a measure of the importance of long-range correlations. Recently, a method was developed for quasi-1D systems which became known as the density matrix renormalization group (DMRG) approach [7] and which, due to its stepwise build-up of the overall wavefunction from well defined lattice subunits (or blocks in the language of the DMRG), works best for those system states not involving significant long-range correlations. The DMRG approach avoids the necessity of extrapolation by giving directly results of very high accuracy for finite-size lattices big enough to be considered infinite. Generally, an expansion of the wavefunction in terms of some well defined substructures would be extremely helpful and would give us a much better understanding of its nature once the expansion coefficients are known to reasonable accuracy. In this context the resonance-valence-bond (RVB) class of functions were proposed in solid-state physics by Anderson [8] in connection with the recently discovered phenomenon of high-T_c superconductivity [9] and expansions of the wavefunctions of spin-1/2 Heisenberg lattices in terms of RVB-type functions [10] have received an increased amount of attention during recent years. The RVB concept however is much older, having its origins in the formulation of the classical VB model developed by Pauling and Wheland for π-electronic systems back in the early 1930s [11]. The Hamiltonian emerging from the classical VB model is entirely equivalent to the spin-1/2 Heisenberg model and it can be shown that the two resulting spectra are identical apart from a trivial level shifting [12]. The introduction of RVB functions was motivated from the chemist's idea of bond formation between two sites by allowing a singlet spin pairing between the corresponding two electrons, as was done in the early days of quantum mechanics when trying to explain the formation of the H_2 molecule [13].

While it is in principle possible, due to the Lieb–Mattis theorem [14], to describe exactly the ground-state wavefunctions of alternate spin-1/2 Heisenberg systems with equal numbers of the two types of site in a bipartite lattice in terms of all possible types of RVB function, it was hoped that only a tiny fraction of them would be responsible for the essential features of the ground-state properties, avoiding thus the combinatorial explosion of the basis, which soon puts a limit on the size of exactly treatable systems. In this context, the nearest-neighbour or dimer RVB states were found to give most of the contribution to the ground-state energies, and the classical RVB *ansatz* for the wavefunction involves an expansion in terms of dimer RVB states with equally weighted coefficients. Obvious improvements are subsequently possible, either by relaxing the constraint of equal coefficients in the dimer RVB *ansatz* or by including RVB functions of longer range, in which spin pairing is allowed to take place between non-nearest-neighbour sites. The former improvement does not change dimensions of the RVB basis and is appropriate for obtaining bulk limit properties due to the size-extensive property of the basis. The inclusion of long-range RVB functions (i.e. long-bond excitations with respect to the dimer RVB states) however has to be performed in an exponential type of coupled-cluster expansion of the wavefunction, including all possible higher excitations of the same type in a multiplicative way, if one is interested in bulk properties. Limiting the RVB basis to contain just up to n-tuply excited RVB functions with n fixed would give no improvement in the $N \rightarrow \infty$ limit. However, for fixed and not too big values of N, this last type of basis can give valuable information concerning the relative importance of the different types of long-range RVB function.

In this paper I present results on the ground state of the isotropic two-, three- and four-legged spin-1/2 ladders using a fully variational RVB *ansatz*. Two types of study were performed. For a finite-sized system of each type of ladder the inclusions of long-range RVB functions of increasing excitation level were investigated and the results were compared

to those obtained from exact diagonalizations. The second type of study involved calculations on finite-sized systems using a dimer RVB function expansion followed by an extrapolation of the results to the bulk limit. I found that twisted rather than periodic boundary conditions along the chains gave much faster convergence to the bulk properties. In both types of calculation I made full use of the lattice groups, obtaining as a by-product also the ground-state lattice group states of the respective ladders.

2. Theory

The isotropic spin-1/2 Heisenberg Hamiltonian, as used in this paper, reads

$$\hat{H} = J \sum_{i \leftrightarrow j} \boldsymbol{S}_i \cdot \boldsymbol{S}_j \tag{1}$$

in which $i \leftrightarrow j$ indicates nearest-neighbour pairs of spins in the lattice and J denotes the overall positive antiferromagnetic exchange constant. Though this Hamiltonian is simple in appearance, characterizing the structure of its exact ground state represents a very difficult task for large connected lattices, due to the overall correlation of the spins involved. The combinatorial explosion of the spin basis severely limits the size of tractable systems, and exact calculations can at present be performed only for systems containing up to $N \approx 30$ spin-1/2 sites, using sophisticated methods which make full use of either symmetric [15] or unitary group [16] properties of the entire spin basis.

Next, the concept of the RVB basis in connection with the above Hamiltonian is introduced for an N-site lattice with even N. Indexing the sites of the lattice with the positive natural numbers from 1 to N, one can partition the resulting set of positive numbers $\{P\}$ into $N/2$ disjoint two-numbered sets P_{kl}. Each such partition is then associated with a spin product function ϕ, in which each two-numbered set P_{kl} of the partition gives rise to a singlet spin pairing between the sites k and l:

$$\{P\} = \sum_{k,l}^{N/2} \bigoplus P_{kl} \longrightarrow \phi = \prod_{k,l}^{N/2} \frac{1}{\sqrt{2}} (\alpha_k \beta_l - \beta_k \alpha_l). \tag{2}$$

Each of these resulting spin product functions is called a RVB function, the name reflecting the idea of constructions of bonds between the different sites of the lattice. From the above method of construction, one can deduce some important properties of the complete overall set $\{\phi\}$ of RVB functions. Since each ϕ is made up only of products of singlets, it is itself a singlet, i.e. it belongs to the $S = 0$ subspace of the entire spin basis. The number of RVB functions which can be constructed is equal to the number of different two-numbered set partitions of $\{P\}$ and hence is equal to $(N-1)!!$. On the other hand the dimension of $\{\phi\}$ is known to be equal to the dimension of the $S = 0$ subspace of the entire spin basis, which is equal to $N!/(N/2+1)!(N/2)!$. This last fact follows from the observation that $\{\phi\}$ always contains a Rumer set of RVB functions, which are known to form a basis for the above-mentioned $S = 0$ subspace and which are formally constructed by placing the numbers of $\{P\}$ on a circle and allowing only those spin pairings which give non-crossing lines inside the circle [17]. Hence one sees that $\{\phi\}$ represents a linearly dependent set of RVB functions.

A very important fact about RVB functions is their easy transformation under the symmetry operations of the lattice groups. If G denotes the lattice group of a particular system, then each of its elements $g \in G$ can be written as a permutation defined on the set $\{P\}$. From equation (2) it then follows immediately that

$$g\phi = \pm\phi^g \tag{3}$$

where ϕ^g stands for the RVB function obtained from ϕ in equation (2) by permuting and rearranging to their original order all the k, l index pairs according to g, the + or − sign holding if an even or odd number of index pair rearrangements has to be performed, respectively. G-adaptation of the RVB basis by means of projection operators is thus relatively easy, particularly for one-dimensional irreps of G, which are of interest in this paper. In this case one can deduce from equation (3) that the original set of RVB functions $\{\phi\}$ decomposes into *non-overlapping* subsets of symmetry-adapted RVB functions $\{\chi\}$ in the sense that no two χs can possess a common ϕ. Hence the usual linear dependency problems associated with the use of projection operator methods are not present in these cases and even very large RVB function sets can be G-adapted with no major computational difficulties.

One of the major drawbacks of the RVB basis is its completely non-orthogonal nature, i.e. $\langle\phi_p|\phi_q\rangle \neq 0$ for all p, q. Formulae for evaluation of the overlaps in terms of some diagrammatic superposition structures between ϕ_p and ϕ_q were derived a long time ago by Pauling for the $S = 0$ and $S = 1/2$ cases [11] and subsequently extended to a more general type of RVB basis (i.e. $S \neq 0, 1/2$) by Cooper and McWeeny and by Sutcliffe [18]. Once an overlap or metric matrix element M_{pq} has been calculated, the corresponding matrix element H_{pq} for the Hamiltonian in equation (1) is obtained at only slightly additional cost. Below, the results in connection with an $S = 0$ RVB basis are presented:

$$M_{pq} = \langle\phi_p|\phi_q\rangle = (-1)^r 2^{n-N/2} \tag{4}$$

$$H_{pq} = \langle\phi_p|\hat{H}|\phi_q\rangle = \frac{3}{4} M_{pq} J \sum_{i\leftrightarrow j} \delta_{ij,L}. \tag{5}$$

In order to explain these equations we need the concept of a Rumer diagram associated with each ϕ in equation (2) [17]. To construct this diagram we place the numbers of $\{P\}$ on a circle and draw arrows between all singlet spin pairings k, l such that each arrow points from k (tail) to l (head). We can then construct a superposition pattern between ϕ_p and ϕ_q by superimposing their associated Rumer diagrams. Then, in equations (4) and (5), r is the number of arrow reversals needed to bring all arrows in the superposition pattern between ϕ_p and ϕ_q head to head and tail to tail, n denotes the number of closed loops (or islands) in the superposition pattern and $\delta_{ij,L}$ is non-zero only if i and j belong to the same loop L, in which case its value is +1 or −1 if the distance between i and j in L is even or odd, respectively. Using the G-adapted RVB basis $\{\chi\}$ we can rewrite equations (4) and (5) in terms of this basis in the following way. Denoting for example by $\hat{O}$ the projection operator for the totally symmetric irreducible representation of G, one can write for an element of $\chi_s \in \{\chi\}$, using equation (3),

$$\chi_s = \hat{O}\phi_p = \frac{1}{|G|}\sum_g g\phi_p = \frac{1}{|G|}\sum_g \pm\phi_p^g \tag{6}$$

and from the Hermitian and idempotent nature of the projection operator, i.e. $\hat{O}^\dagger = \hat{O}^2 = \hat{O}$, it follows that

$$M_{st} = \langle\chi_s|\chi_t\rangle = \langle\hat{O}\phi_p|\phi_q\rangle = \frac{1}{|G|}\sum_g \pm\langle\phi_p^g|\phi_q\rangle \tag{7}$$

$$H_{st} = \langle\chi_s|\hat{H}|\chi_t\rangle = \langle\hat{O}\phi_p|\hat{H}|\phi_q\rangle = \frac{1}{|G|}\sum_g \pm\langle\phi_p^g|\hat{H}|\phi_q\rangle \tag{8}$$

where the elements $\langle\phi_p^g|\phi_q\rangle$ and $\langle\phi_p^g|\hat{H}|\phi_q\rangle$ are evaluated according to the rules in equations (4) and (5). Hence one sees that, as a rule of thumb, dimensions of $\boldsymbol{M}$ and $\boldsymbol{H}$ are reduced by a factor of roughly the order $|G|$ of the lattice group.

A particularly useful subset of the entire original RVB set $\{\phi\}$ is the set $\{\phi^K\}$ composed of all dimer RVB functions; the reason for choosing the suffix K will become clear soon. These functions, are defined as having only spin pairings between nearest neighbours in the lattice and their number, denoted in the following by K, is usually much smaller than the order and even the dimension of $\{\phi\}$. In the VB theory of planar π molecular systems, these dimer RVB functions are in one-to-one correspondence with the so-called Kekulé functions and their importance is reflected in the derivation of several qualitative rules dealing with the stabilization of π-systems, particularly for benzenoid systems containing fused six-membered rings [11,19]. Although $K \ll \mathrm{Dim}\{\phi\}$, RVB calculations based on $\{\phi^K\}$ alone usually recover around ≈90% of the ground-state energy (see, for example, table I in [20]), even for the most unfavourable case of the linear isotropic chain lattice. This property makes $\{\phi^K\}$ an ideal candidate for finding the lattice group symmetry of the ground state, especially for big lattices containing frustration (such as those of fullerenes), for which no general rules are known. The dimer RVB functions are thus of invaluable help in obtaining qualitative information about the lattices. Note that as the coordination number of the lattice increases, K also increases, so the set $\{\phi^K\}$ approaches completeness, giving exact results for infinitely coordinated lattices, where the set $\{\phi^K\}$ becomes identical in nature to $\{\phi\}$.

To get a more systematic description of the set $\{\phi\}$, one can classify each $\phi \in \{\phi\}$ according to its degree of excitation e depending on how many singlet pairs in ϕ are between non-nearest-neighbour sites in the lattice. Furthermore, within each set $\{\phi^e\}$ of equally excited RVB functions, one can label each $\phi^e \in \{\phi^e\}$ by the intersite distance $d_1, d_2, \ldots, d_e$ associated with the first, second, $\ldots$, eth excited singlet pair in ϕ^e. An intersite distance between two sites is defined as the *minimum* number of nearest-neighbour site pairs between those sites (in the case of a 2D square lattice this distance is identical to the so-called Manhattan distance [10]). Hence we can formally decompose the RVB set in the following way:

$$\{\phi\} = \{\phi^K\} \oplus \bigoplus_{e=1}^{e^{max}} \bigoplus_{d=e\times 2}^{e\times d^{max}} \{\phi_d^e\} \tag{9}$$

where $e \times 2$ and $e \times d^{max}$ denote e excited singlet pairs of distance 2 and d^{max}, respectively. Note that the second sum in equation (9) runs over all distance value combinations such that $d_1 \leqslant d_2 \leqslant \cdots \leqslant d_e$. One can proceed even further by noting that each symmetry operation $g \in G$ of the lattice group preserves excitation and distance properties on each ϕ^K and ϕ_d^e, i.e. $g\phi^K \in \{\phi^K\}$ and $g\phi_d^e \in \{\phi_d^e\}$. Hence both sets $\{\phi^K\}$ and $\{\phi_d^e\}$ can be G-decomposed and equation (9) can be rewritten as

$$\{\phi\} = \left(\bigoplus_{\gamma=1}^{\gamma^{max}} \{\phi^K(\gamma)\}\right) \oplus \left(\bigoplus_{e=1}^{e^{max}} \bigoplus_{d=e\times 2}^{e\times d^{max}} \bigoplus_{\gamma=1}^{\gamma^{max}} \{\phi_d^e(\gamma)\}\right) \tag{10}$$

where γ stands for an index counting different G-adapted sets and depends on e as well as on d. Equation (10) represents the basis upon which the computer program for the present calculations was written and holds for any lattice whatsoever.

3. Results and discussion

In this section I present results obtained for L-legged ladders in the range $2 \leqslant L \leqslant 4$ with periodic boundary conditions along the rungs for $L = 3$ and $L = 4$. The sites of each rung were numbered consecutively, whereas the sites of the chains were numbered in such a way that the absolute difference between any two nearest-neighbour sites is L. Figure 1 shows a pictorial view of the ladders together with the numbering scheme used.

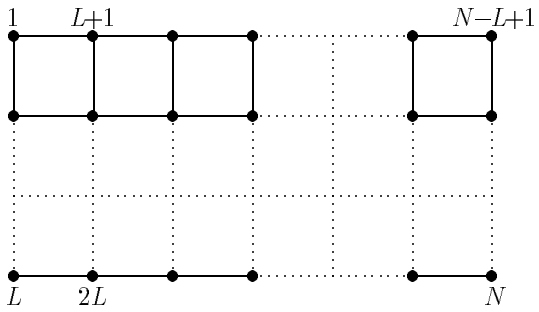

Figure 1. The labelling scheme used for the L-legged ladders with periodic boundary conditions $kL+1 \leftrightarrow (k+1)L$; $k = 0, 1, \ldots, R-1$ along the rungs for $L > 2$.

The following boundary conditions were used along the chains:

periodic (PBC):	$k \leftrightarrow N - L + k$	$k = 1, 2, \ldots, L$
twisted (TBC):	$k \leftrightarrow N - L + k + 1$	$k = 1, 2, \ldots, L-1$
	$L \leftrightarrow N - L + 1.$	

Note that the PBC/TBC give rise to frustration between the first and last rungs of the lattice depending on the parity of the number $R = N/L$ of rungs.

3.1. Lattice groups G and number K of dimer RVB functions

In order to establish the lattice groups of the ladders, the following lattice operations are introduced in the form of permutational operators, based on the numbering scheme used in figure 1:

$$\hat{C}_N = \begin{pmatrix} 1 & \to & N-L & N-L+1 & N-L+2 & \to & N \\ L+1 & \to & N & L & 1 & \to & L-1 \end{pmatrix} \tag{11}$$

$$\hat{C}_R = \begin{pmatrix} 1 & \to & N-L & N-L+1 & \to & N \\ L+1 & \to & N & 1 & \to & L \end{pmatrix} \tag{12}$$

$$\hat{C}_2 = \begin{pmatrix} 1 & \to & N \\ N & \leftarrow & 1 \end{pmatrix} \tag{13}$$

$$\hat{C}_L = \prod_{k=0}^{k=R-1} \begin{pmatrix} 1+kL & \to & L-1+kL & L+kL \\ 2+kL & \to & L+kL & 1+kL \end{pmatrix} \tag{14}$$

$$\hat{C}_s = \prod_{k=0}^{k=R-1} \begin{pmatrix} 1+kL & \to & L+kL \\ L+kL & \leftarrow & 1+kL \end{pmatrix}. \tag{15}$$

In these expressions, the symbols $\to$ and $\leftarrow$ indicate increase and decrease in units of 1. Note that $\hat{C}_N$ and $\hat{C}_R$ operate on sites along the chains, while $\hat{C}_L$ and $\hat{C}_s$ interchange sites along the rungs only. Furthermore, the operations $\hat{C}_L$ and $\hat{C}_s$ become identical for $L = 2$. Using each of the above operations in equations (11)–(15) as generators, one can construct the five cyclic groups $\mathcal{C}_N, \mathcal{C}_R, \mathcal{C}_2, \mathcal{C}_L$ and $\mathcal{C}_s$ of orders N, R, 2, L and 2, respectively. From these cyclic groups one can form two product groups, namely $D_N = \mathcal{C}_N\mathcal{C}_2$ and $D_{Rh}\mathcal{C}_L = \mathcal{C}_R\mathcal{C}_2\mathcal{C}_s\mathcal{C}_L$, which are the lattice groups for the TBC and PBC L-legged ladders, respectively. Table 1 gives an overview of the orders of these groups for the ladders considered in this paper, together with the characters of the generators for their respective ground states.

Table 1. Lattice groups G and characters of the generators for the ground states of the PBC and TBC L-legged ladders with $L = 2, 3, 4$.

L	BC	G	Ord$\{G\}$		$\hat{C}_N$	$\hat{C}_R$	$\hat{C}_L$	$\hat{C}_2$	$\hat{C}_s$
2	PBC	$D_{R\mathrm{h}}$	$2N$	$R =$ even		$+1$		$+1$	$+1$
				odd		$+1$		-1	-1
	TBC	D_N	$2N$	even	-1			$+1$	
				odd	-1			-1	
3	PBC	$D_{R\mathrm{h}}C_3$	$4N$	$R/2 =$ even		$+1$	$+1$	$+1$	$+1$
				odd		-1	$+1$	-1	$+1$
	TBC	D_N	$2N$	even	-1			$+1$	
				odd	$+1$			-1	
4	PBC	$D_{R\mathrm{h}}C_4$	$4N$			$+1$	$+1$	$+1$	$+1$
	TBC	D_N	$2N$		$+1$			$+1$	

Next some recursion relations are presented for evaluating the number of dimer RVB functions. Denoting by P_R and Q_R the numbers K of dimer RVB functions for open and periodic or twisted boundary conditions for L-legged ladders consisting of R periodic rungs, respectively, these relations are given by

$$L = 2: \qquad P_R = P_{R-1} + P_{R-2} \qquad P_0, P_1 = 1, 1 \tag{16}$$

$$L = 3: \qquad P_R = 5P_{R-2} - P_{R-4} \qquad P_0, P_2 = 1, 4 \tag{17}$$

$$L = 4: \qquad P_R = 3P_{R-1} + 3P_{R-2} - P_{R-3} \qquad P_0, P_1, P_2 = 1, 2, 9. \tag{18}$$

Relations (16)–(18) are easily proved by adding one periodic rung at a time and counting the resulting extra dimer RVB functions. Note that relation (16) reproduces the Fibonacci sequence, and hence for the two-legged ladder P_R is equal to one of the Fibonacci numbers [21]. Having the P_R-numbers, the equations for the Q_R-numbers are given by

$$L = 2: \qquad Q_R = P_R + P_{R-2} + 2(\delta_{per,R=\mathrm{even}} + \delta_{tw,R=\mathrm{odd}}) \tag{19}$$

$$L = 3: \qquad Q_R = 2P_R + 6\delta_{per} + 6\sum_{i=2}^{R/2}(i-1)P_{R-2i} \tag{20}$$

$$L = 4: \qquad Q_R = 6P_{R-1} + 2\delta_{R=\mathrm{even}} - 4\delta_{R=\mathrm{odd}} + W_R(\delta_{per,R=\mathrm{even}} + \delta_{tw,R=\mathrm{odd}}) \tag{21}$$

with

$$\begin{aligned} &W_R = 2W_{R-1} + W_{R-2} - 4(1 + 4\delta_{R=\mathrm{odd}}) \\ &W_2, W_3 = 22, 30 \end{aligned} \tag{22}$$

where $\delta_{x,y,\ldots}$ is equal to 1, if simultaneously the conditions $x, y, \ldots$ are fulfilled, 0 otherwise and per and tw are shorthand notation for periodic and twisted boundary conditions along the chains. As an example of application of the above formulae, the number Q_7 of dimer RVB functions for a 4×7 TBC ladder is evaluated. First one must evaluate P_6 using equation (18) and W_7 using the recurrence relation (22). One obtains $P_6 = 1681$ and $W_7 = 958$. For a 4×7 TBC ladder, one has $\delta_{tw,R=\mathrm{odd}} = \delta_{R=\mathrm{odd}} = 1$ and equation (21) gives thus $Q_7 = 6P_6 - 4 + W_7 = 11\,040$. An interesting quantity is further the number of dimer RVB functions per site in the limit $N \to \infty$, expressed as $K^{1/N}$. One obtains for the

above ladders, using equations (16)–(18),

$$L = 2: \qquad \lim_{R\to\infty} \sqrt[2R]{P_R} = \left(\frac{1+\sqrt{5}}{2}\right)^{1/2} \approx 1.2720 \tag{23}$$

$$L = 3: \qquad \lim_{R\to\infty} \sqrt[3R]{P_R} = \left(\frac{5+\sqrt{21}}{2}\right)^{1/6} \approx 1.2984 \tag{24}$$

$$L = 4: \qquad \lim_{R\to\infty} \sqrt[4R]{P_R} = (2+\sqrt{3})^{1/4} \approx 1.3899. \tag{25}$$

Comparing the values obtained for the $L = 2$ and $L = 4$ ladders with the value obtained for the infinite 2D square lattice (i.e. $L = \infty$) of $\lim_{N\to\infty} K^{1/N} = e^{C/\pi} \approx 1.3385$, where $C = \sum_{n=1}^{\infty} (-1)^n n^{-2}$ is Catalan's constant [22], one can deduce that the exact energy per spin values for the ground state will be ordered in the sequence $L = 2$, $L = \infty$ and $L = 4$ as is indeed the case, the exact values being -0.578 [5, 23], -0.669 [24] and -0.683 [25] in units of J, respectively. Note that including the value of $L = 3$ in the above comparison makes no sense, due to frustration present along the rungs.

3.2. Variational dimer RVB calculations

This subsection presents variational dimer RVB results on the ground state for some finite-sized two-, three- and four-legged PBC and TBC ladders, together with their extrapolated bulk values. Full use was made of the lattice groups introduced in the last subsection, in order to perform calculations on some reasonably sized systems. For the extraction of the lowest eigenstates a modified Davidson algorithm was used to allow for an overlap matrix different from unity (for details of the necessary modifications see [20]). Tables 2, 3 and 4 give the results obtained for the energies per spin. The most striking observation from the data in these tables is the much faster convergence to the bulk energies for the chain-frustrated $L = 2$ and $L = 4$ ladders. For the 2×20 ladder, the chain-frustrated TBC ladder already gave an energy per spin value to an accuracy of within 10^{-10}, whereas the corresponding chain-alternate PBC one had its energy value converged to within only 10^{-6}. The same pattern was observed for the

Table 2. Ground-state energy per spin (E_0/spin) results in units of J for $L = 2$ PBC [TBC] ladders with alternation (*alt*) and frustration (*frus*) along the chains, together with orders of the original $\{\phi^K\}$ and G-adapted $\{\chi^K\}$ dimer RVB spaces.

R	Ord$\{\phi^K\}$	Ord$\{\chi^K\}$	E_0/spin(*alt*)	E_0/spin(*frus*)
8	49 [47]	9 [8]	−0.560204	[−0.558228]
9	76 [78]	9 [10]	[−0.559204]	−0.558247
10	125 [123]	15 [14]	−0.558716	[−0.558254]
11	199 [201]	16 [17]	[−0.558480]	−0.558255
12	324 [322]	27 [26]	−0.558365	[−0.558256]
13	521 [523]	31 [32]	[−0.558309]	−0.558256
14	845 [843]	50 [49]	−0.558281	[−0.558256]
15	1364 [1366]	64 [65]	[−0.558268]	−0.558256
16	2209 [2207]	100 [99]	−0.558262	[−0.558256]
17	3571 [3573]	133 [134]	[−0.558259]	−0.558256
18	5780 [5778]	210 [209]	−0.558257	[−0.558256]
19	9349 [9351]	291 [292]	[−0.558256]	−0.558256
20	15129 [15127]	456 [455]	−0.558256	[−0.558256]
∞				−0.5582557513

Table 3. Ground-state energy per spin (E_0/spin) results in units of J for $L = 3$ PBC [TBC] ladders with alternation (*alt*) and frustration (*frus*) along the chains, together with orders of the original $\{\phi^K\}$ and G-adapted $\{\chi^K\}$ dimer RVB spaces.

R	Ord$\{\phi^K\}$	Ord$\{\chi^K\}$	E_0/spin(*alt*)	E_0/spin(*frus*)
6	224 [218]	12 [12]	−0.567597	[−0.550835]
8	1058 [1052]	30 [36]	−0.550525	[−0.543217]
10	5054 [5048]	77 [112]	−0.542091	[−0.538884]
12	24200 [24194]	252 [400]	−0.538159	[−0.536823]
14	115934 [115928]	852 [1511]	−0.536474	[−0.535936]
16	555458 [555452]	3278 [6089]	−0.535789	[−0.535573]
∞				−0.5351

Table 4. Ground-state energy per spin (E_0/spin) results in units of J for $L = 4$ PBC [TBC] ladders with alternation (*alt*) and frustration (*frus*) along the chains, together with orders of the original $\{\phi^K\}$ and G-adapted $\{\chi^K\}$ dimer RVB spaces.

R	Ord$\{\phi^K\}$	Ord$\{\chi^K\}$	E_0/spin(*alt*)	E_0/spin(*frus*)
4	272 [194]	24 [13]	−0.676683	[−0.637516]
5	722 [888]	30 [37]	[−0.663023]	−0.644667
6	3108 [2702]	101 [82]	−0.656242	[−0.646946]
7	10082 [11040]	200 [244]	[−0.652424]	−0.647571
8	39952 [37634]	642 [683]	−0.650336	[−0.647757]
9	140450 [146024]	1662 [2196]	[−0.649188]	−0.647809
10	537636 [524174]	5351 [6923]	−0.648564	[−0.647824]
∞				−0.64785

$L = 4$ ladders, enabling one to extrapolate the energy of the bulk system to a fairly accurate value believed to be accurate within 10^{-5}. For the $L = 3$ ladders almost no difference was observed in the convergence pattern, the energy for the chain-alternate systems for R rungs being always almost equal to the energy of the chain-frustrated ones with $R - 2$ rungs with modest convergence to the limit. Consequently, the $L = 3$ extrapolated energy value is the least accurate of the three. A comparison with the values of −0.556 029 [21] and −0.641 489 [26] for the $L = 2$ and $L = 4$ ladders with an equally weighted dimer RVB function *ansatz* shows that there is only slight improvement when allowing for a full variational interaction between the dimer RVB functions, this improvement being more pronounced for the $L = 4$ ladder. This shows that within the dimer RVB functional space for alternate lattices the equally weighted dimer RVB function *ansatz* is reasonable for the present quasi-1D systems. For odd L with periodic rungs there is no *a priori* well defined equal-weighted dimer RVB function *ansatz* due to frustration along the rungs, which prevents a phase assignment to all dimer RVB functions such that all the entries of the overlap matrix M_{pq} in equation (4) are positive. However, one can define an equal-weighted dimer RVB function *ansatz after* a variational dimer RVB calculation has been carried out, taking as the phase factors the corresponding signs of the resulting variational wavefunction, i.e. letting the variational calculation choose the sign.

It would be interesting to see what happens to the variational energy gain in the dimer RVB *ansatz* as one moves to higher-legged $L > 4$; $L =$ even ladders and in particular what the situation is for the 2D square-planar lattice. Will the energy gain be less or greater than for the $L = 4$ ladder studied in this paper? Variational dimer RVB calculations were performed on fragments of the 2D square-planar lattice by Johnson and Subbaswamy [27], producing on extrapolation an energy per spin value of $-0.600J$ for the infinite lattice, which can be

compared with results from the equal-coefficient dimer RVB *ansatz*, namely $-0.604J$ [10] obtained from a Monte Carlo study on 2D lattice fragments and -0.599 [28] obtained from extrapolation on results of finite-legged ladders. These values suggest no energy difference between the two approaches. However, the variational RVB calculations performed in [27] made no use of the lattice group symmetry and were therefore limited by calculations performed on rather small $N \leqslant 20$ 2D lattice fragments, inducing thus a possibly great extrapolation error on their bulk limit result. Fully variational RVB calculations are difficult to perform for the 2D square-planar lattice, because of the need for reasonably large lattice sizes for extrapolation and the consequent intractability of the large resulting RVB spaces even when using the full lattice groups.

For the absolute values of the spin–spin correlation functions of distance 1 along the rungs and distances 1, 2, 3 and 4 along the chains, the following values presented in table 5 were obtained either directly ($L = 2$) or upon extrapolation ($L = 3$ and $L = 4$). A notable feature of this table is the observation that frustration along the rungs lowers considerably its correlation values, a fact which is also observed when performing exact calculations. Also the rung correlation value of the $L = 4$ case is lower than the corresponding one of the $L = 2$ ladder. A very naive explanation of this trend can be given by looking only at the rung part of the ladders. For the $L = 2$ rung there is only one way to place a spin pairing. For the $L = 4$ rung, because of periodic boundary conditions, there are two possible ways, whereas for the $L = 3$ one, the spin pairing can be placed in three different ways, leaving one site of the rung non-paired. Hence the ratios for the rung correlation values should be approximately 1:1/3:1/2 for the respective two-, three- and four-legged ladders, which by looking at table 5 can be seen to be roughly fulfilled. Again one can compare the absolute distance-1 correlation values obtained for $L = 2$ (rungs $= 0.505\,991$ and chain $= 0.305\,260$) and $L = 4$ (rungs $= 0.377\,35$ and chain $= 0.270\,50$) with the ones obtained from the equally weighted dimer RVB function *ansatz* [26]: $L = 2$ (rungs $= 0.539\,779$ and chain $= 0.286\,139$) and $L = 4$ (rungs $= 0.395\,096$ and chain $= 0.246\,393$), showing again that the variational dimer RVB calculations alleviate only slightly the main defect of all dimer RVB calculations in putting too much correlation on the rung part and too little along the chain part of the ladders.

Table 5. Absolute values of spin–spin correlation functions for distance 1 along the rungs ($rung$) and distances 1, 2, 3 and 4 along the chains ($chain1$, $chain2$, $chain3$ and $chain4$) for the infinite two-, three- and four-legged ladders in the variational dimer RVB space.

L	$rung$	$chain1$	$chain2$	$chain3$	$chain4$
2	0.505991	0.305260	0.051365	0.011751	0.002686
3	0.1974	0.3378	0.0493	0.0173	0.0039
4	0.37735	0.27050	0.05115	0.01331	$< 0.00334^{\dagger}$

† This value corresponds to the one obtained for the 4×10 TBC ladder, since too few values were obtained for extrapolation.

3.3. Variational excited RVB calculations on some finite systems

This subsection is devoted to some finite-sized-ladder calculations involving excitations beyond the simple dimer RVB *ansatz*. The choice of the finite-sized two-, three- and four-legged ladders was governed by the possibility of performing exact calculations on one hand and some fairly excited RVB calculations on the other hand. Hence the 24-sited 2×12, 3×8 and 4×6 PBC ladders were chosen and the results are presented in table 6. Several observations can be made from these results. First, excited RVB functions with only $d = 3$ functions give a

Table 6. Ground-state energy (E_0) results in units of J and distance-1 correlation values along the rung ($rung$) and the chain ($chain$) part for the 2×12, 3×8 and 4×6 PBC ladders obtained using excited RVB functions as presented in equation (10) on top of the dimer RVB functional space $\{\phi^K\}$. The designations $\gamma = ccc, ccr$ and crr denote the G-invariant distance-3 types of excitation starting from a specific site and moving to the next sites along chain–chain–chain, chain–chain–rung and chain–rung–rung paths, respectively. Omitted d- and γ-designations imply the full range of respective values.

e, d, γ in $\{\phi_d^e(\gamma)\}$	Ord$\{\phi_d^e(\gamma)\}$	E_0	$rung$	$chain$
2×12 ladder				
Dimer RVB	324	−13.400752	−0.505092	−0.305818
$e = 1, d = 3, \gamma = ccc$	1188	−13.656022	−0.473714	−0.332144
$e = 1, d = 3, \gamma = ccr$	1668	−13.750182	−0.476831	−0.334509
$e = 1, d = 3$	2532	−13.795453	−0.468176	−0.340723
$e = 1$	3732	−13.801258	−0.466853	−0.341626
$e = 2, d = 3, 3$	11028	−13.853735	−0.459208	−0.347635
$e = 2$	99708	−13.875178	−0.454350	−0.350957
Exact†	208012	−13.880932	−0.452918	−0.351913
3×8 ladder				
Dimer RVB	1058	−13.212602	−0.187473	−0.363052
$e = 1, d = 3, \gamma = ccc$	6602	−13.675768	−0.185942	−0.383882
$e = 1, d = 3, \gamma = ccr$	9218	−13.687031	−0.187051	−0.383242
$e = 1, d = 3$	14762	−13.800578	−0.186172	−0.388852
$e = 1$	41114	−13.803729	−0.186198	−0.388958
$e = 2, d = 3, 3$	95894	−13.899884	−0.185945	−0.393216
Exact†	208012	−13.911234	−0.185681	−0.393954
4×6 ladder				
Dimer RVB	3108	−15.749810	−0.359582	−0.296661
$e = 1, d = 3, \gamma = ccc$	8532	−15.991749	−0.355161	−0.311162
$e = 1, d = 3, \gamma = crr$	16260	−16.153227	−0.366205	−0.306846
$e = 1, d = 3, \gamma = ccr$	25668	−16.275739	−0.354072	−0.324084
$e = 1, d = 3$	44244	−16.426147	−0.358229	−0.326194
$e = 1$	49284	−16.426925	−0.358194	−0.326261
$e = 2, d = 3, 3$	326544	−16.536758	−0.358034	−0.330998
Exact†	208012	−16.552514	−0.357721	−0.331967

† All these values were calculated in the exact $S = 0$ spin space with the SPINSGA program [29], making full use of the SU(2) symmetry of the spin basis.

far larger contribution to the energy than any other d-type, as can be seen by comparing both $e = 1, d = 3$ and $e = 1$ results for the two- and three-legged ladders. The corresponding results for the four-legged ladder are not conclusive enough, the $e = 1, d = 3$ RVB space covering almost the entire $e = 1$ RVB space. Within each $e = 1, d = 3$ set, the different types of G-adapted subset (ccc, ccr and crr) give comparable contributions to the energy when including them separately, the amount each of them contributes being roughly proportional to their respective orders. Still the entire $e = 1, d = 3$ set gives a substantial lowering as compared to each of its subsets; in other words there is no single dominant $e = 1, d = 3$ subset. Hence in excited RVB calculations the single excitations most important by far in the wavefunctions are those coming from the *entire* $e = 1, d = 3$ set, even in the presence of frustrations, as shown by the three-legged ladder results. This result is consistent with excited RVB calculations [30] performed on some fullerene lattice structures with $20 \leqslant N \leqslant 60$ containing five- and six-membered rings as opposed to the three- and four-membered rings

in the present ladders; hence this result seems to be quite universal. The inclusion of the $e = 2, d = 3, 3$ set on top of the dimer RVB set already gives $\approx$99, 9% of the true ground-state energies and results very close to the true correlation values, even for the 2×12 ladder, where this type of RVB space is of much lower order than the exact spin space. Thus the dimer RVB plus the $e = 2, d = 3, 3$ set cover the essential features of the ground state of lattices in the range of say $N \leqslant 60$. Note that this last type of RVB space is not free of problems, the main drawback being the possibility of linear dependencies within the RVB functions as is obviously the case for the 4×6 ladder, in which the order of the $e = 2, d = 3, 3$ set alone far exceeds the dimension of the exact spin space. However the bigger the lattice and the lower their coordination numbers, the less likely it is that linear dependency problems will occur.

4. Summary and conclusions

In this paper fully variational RVB calculations were performed for two-, three- and four-legged ladders with periodic boundary conditions along the rungs and periodic/twisted boundary conditions along the chains. In the dimer RVB space approximation some fairly large lattices were treated, due to consequent use of the respective lattice groups. It was found that chain-frustrated ladders rather than chain-alternate ones lead to much faster convergence for both the energy per spin and the correlation function values, this feature enabling very accurate extrapolations to the respective bulk limit properties. The observed convergence patterns for the energies were quite different between the $L = 2$ and $L = 4$ ladders and the $L = 3$ ladder. In the former ones, chain alternation and chain frustration approach the $N \rightarrow \infty$ limit from two opposite sides, namely from below (lower energy) and above (higher energy), respectively. The $L = 3$ ladder on the other hand approaches this limit only from below, i.e. both chain-alternate and chain-frustrated finite lattices are always lower in energy than the corresponding infinite lattice. The resulting energy per spin values obtained for the $L = 2$ and $L = 4$ ladders are only slightly lower than the corresponding ones obtained from an equally weighted dimer RVB function *ansatz*, the lowering being more pronounced for the $L = 4$ ladder. The results presented in this paper should be useful when comparing with results obtained in the same space but with an *a priori* fixed structure of the wavefunction. As a by-product of the present calculations, the lattice group states for the ground states were obtained and given in the form of their one-dimensional characters for the corresponding lattice group generators.

It was further shown, from excited RVB calculations on some finite ladder systems, that excited RVB functions containing long spin pairings with $d = 3$ are by far the most important ones, even for such highly frustrated systems as the cylindrical three-legged ladder considered in this paper. For the $L = 2$ and $L = 4$ ladders this result comes as no surprise, since it is known that they belong to the short-range RVB universality class with excited RVB function amplitudes in their ground-state wavefunctions decaying exponentially in d and with a finite excitation gap [5]. For the $L = 4$ ladder this result is insensitive to the boundary conditions along the rungs. For $L = 3$ the type of boundary condition used along the rungs is important. Open boundary conditions lead to power-law-decaying excited RVB function amplitudes and the system becomes gapless. As a consequence, excited RVB functions with $d > 3$ are important and not negligible in describing the ground state. On the other hand, the similarity in excited RVB results obtained in the present paper between the $L = 2$ and $L = 4$ ladders and the cylindrical $L = 3$ ladder shows that in contrast to the $L = 3$ ladder with open boundary conditions along the rungs, the cylindrical $L = 3$ ladder should fall into the short-range RVB universality class with a finite excitation gap, a picture also supported by recent DMRG studies on this system [31].

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