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Symmetry adapted finite-cluster solver for quantum Heisenberg model in two dimensions: a real-space renormalization approach

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Abstract

We present a quantum cluster solver for the spin- S Heisenberg model on a two-dimensional lattice. The formalism is based on the real-space renormalization procedure and uses the lattice point group-theoretical analysis and non-Abelian $SU(2)$ spin symmetry technique. The exact diagonalization procedure is used twice at each renormalization group step. The method is applied to the spin-half antiferromagnet on a square lattice, and a calculation of local observables is demonstrated. A symmetry-based truncation procedure is suggested and verified numerically.

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1. Introduction

Low-dimensional magnetic systems are currently a subject of intensive experimental and theoretical work. Cluster methods, which approximate the physics of the infinite system by solving the problem for a corresponding finite cluster, are the most frequently used theoretical approaches, as they account short-range correlations on the scale of the cluster size. Numerical standard methods in the field, such as quantum Monte Carlo (QMC), exact diagonalization (ED) [1], and density matrix renormalization group (DMRG) [2, 3], are able to give essentially exact results on limited size systems and form a versatile methodological triad in simulations of model Hamiltonians.

Even though these techniques have had spectacular successes in calculating ground state energies and many other properties of one-dimensional (1D) and two-dimensional (2D) quantum spin systems [4–7] there is a problem with utilizing symmetries and good quantum numbers of the Hamiltonian, which may be exploited to thin out the Hilbert space by decomposing it into a sum of sectors. Common symmetries and conservation laws encountered in spin systems are (i) Ising or XY symmetry (magnetization conservation $S_{\text{tot}}^z = \text{const}$), (ii) point-group symmetry (parity, angular momentum conserved) and (iii) full $SU(2)$ symmetry (S_{tot}^2 conserved). Among these symmetries only the first is usually exploited in numerical calculations. The full $SU(2)$ spin symmetry is rather hard to implement, since

it requires efforts similar to the diagonalization of the actual Hamiltonian to construct the eigenstates of S_{tot}^2 . An implementation of non-Abelian $SU(2)$ spin symmetry based on Clebsch–Gordan transformations and elimination of quantum numbers via the Wigner–Eckart theorem was performed for the interaction round a face (IRF) models in the framework of the IRF-DMRG method [8]. This technique has been successfully applied to the spin-1/2 Heisenberg chain and, later, to the spin-1 and spin-2 Heisenberg chains [9]. The performant DMRG method conserving a total spin quantum number has been suggested by McCulloch and Gulasci [10, 11]. An application of $SU(2)$ symmetries for the matrix product method (MPM) closely related to the DMRG [12, 13] gives a rotationally invariant formulation valid for spin chains and ladders [13, 14].

As for the lattice point symmetry, despite its importance in characterizing energy states of a spin system, there appears to have been little previous work on the subject. Even though an implementing this symmetry does not lead to a drastic reduction of a dimension of the Hilbert space sector to be diagonalized, we can resolve properties as a function of additional quantum numbers (irreducible representations of the point group). This circumstance might be crucial for efficient truncation of the Hilbert space in algorithms based on the real-space renormalization group (RSRG) procedure. This generates a motivation for the present paper, namely, we present a finite cluster solver based on the RSRG scheme which allows us to exploit both the continuous non-Abelian $SU(2)$ symmetry and discrete symmetry of the lattice point group in application to isotropic two-dimensional *spin-S* systems. As an example of illustrating features of our method we consider the spin-1/2 Heisenberg antiferromagnet (AFH) on a square lattice. This choice is motivated by two reasons. First, the physical properties of the $S = 1/2$ AFH model on the infinite square lattice at $T = 0$ have been much studied and calculated by various methods by many physicists [15, 16]. The focus has generally been on the ground-state energy and staggered magnetization, although some other quantities have also been computed (see [17], for example). Second, the underlying idea of our approach was first developed by Lin and Campbell in the study of this model system [18, 24]. Before moving on to the details, we discuss important aspects of finite-lattice simulations using the ED method regarding the cluster geometry.

The method of exact diagonalization has been used on the best bipartite finite square lattices with up to $N = 38$ vertices [25, 27]. On each of the set of finite lattices, the Hamiltonian of the quantum spin model is diagonalized exactly to find the ground-state energy and the ground-state eigenvector. The ground-state properties can then be calculated exactly. The exact ground-state data for each physical property of the model on all finite lattices are extrapolated against an appropriate inverse power of N to obtain an estimate of the property on the infinite lattice at zero temperature [28]. Haan *et al* [30] showed that certain parallelogram clusters could produce good results in finite-size exact diagonalization calculations. Later, Betts *et al* developed a grading scheme of parallelogram tiles of the square lattice that could generate the best finite clusters [27]. From a symmetry point of view, this approach has an apparent flaw: the point symmetry of a parallelogram cluster does not match that of the infinite square lattice.

In this respect, a renormalization-group (RG) approach suggested by Lin and Campbell combining exact diagonalization results with a RG-type analysis seems to be more promising. The basic idea of their calculations for 2D AFH on an $n \times n$ cluster (n is an odd integer) is to divide this cluster into two parts: an inner $(n - 2) \times (n - 2)$ cluster and the perimeter (‘outer ring’). The AFH model is firstly solved for the inner cluster and its ground state is mapped onto a single-effective spin (all the excited states are thrown away). Thus, the problem is reduced to an effective 1D AFH model in which spins on the outer ring experience antiferromagnetic nearest-neighbour interactions and interact individually with the effective central spin. The

latter plays the role of a staggered external magnetic field. The procedure is repeated for the increasing values of n and demonstrates convincingly that the staggered magnetic long-range order exists at zero temperature. We note especially that the lattice point symmetry holds for all the clusters, and their ground state always has spin $S = 1/2$ (not a singlet) according to the Lieb–Mattis theorem [31].

We offer to change the real-space RG strategy of the approach making it closer to DMRG methodology. Our treatment begins by dividing a cluster into a central spin and its environment. In the course of real-space RG iterations the environment increases (technical details are discussed in the text) and we determine how coupling between the central spin and the environment varies. Note especially that we address to the exact diagonalization procedure twice at each RG step. The first use gives access to states of the environment and the second one does the same to those of the whole cluster that provides its spectrum and observables of interest. Within our RG framework, local results such as the energy per bond ε and the staggered magnetic moment m are measured on the central site.

We have carried out the renormalization procedure through systems of size $\sqrt{17} \times \sqrt{17}$, and, in contrast to approach in Ref. [18], we keep *not only* the ground state. For small clusters ($\sqrt{5} \times \sqrt{5}$, 3×3 , $\sqrt{13} \times \sqrt{13}$) we use all of the excited states of the environment found by exact diagonalization, whereas for the cluster of size $\sqrt{17} \times \sqrt{17}$ we apply a symmetry-based truncation procedure, retaining only the states with largest weight in the environment density matrix. For this cluster we have compared the exact diagonalization result for ε and m with those obtained via our renormalization group, and we regard the resulting better than $10^{-2}\%$ agreement as support for the reliability of our calculations.

We note that several other methods to improve the RSRG calculations have been previously formulated to study low-energy properties of spin lattice models. Among the most important and successful ones, one may cite the real-space renormalization group with effective interactions (RSRG-EI) [19], and the dressed cluster method (DCM) [20, 21]. The first method is an improvement of the RSRG method originally proposed by Wilson. By considering the blocks of lattice it extracts effective interactions between the blocks through the exact diagonalization of dimers of blocks. Knowledge of the exact spectrum of the dimers enables one to define interblock effective interactions via an effective Hamiltonian. This procedure is iteratively repeated to blocks of blocks providing at a very low cost reasonable estimate of the energy per bond for 1D and 2D spin lattices. The second method (DCM) uses a single-reference wavefunction as do the coupled cluster method (CCM) [22, 23]. This wavefunction is used as a bath in which a finite cluster is embedded and treated exactly. The effect of excitations occurring on the bonds around the cluster is taken into account through a dressing of the cluster configuration interaction (CI) matrix. This approach gives results for the cohesive energy of the same accuracy as the best QMC ones. The DCM can be seen as a convenient approximation of the CCM. However, the problem is formulated as a diagonalization of a dressed CI matrix instead of the resolution of a nonlinear system of equations.

The paper is organized as follows. The general formalism for two-dimensional spin- S systems is introduced in the following section. In section 3 we apply the method to the AFH model on a square lattice. Finally, our conclusions and an outlook are presented in section 4.

2. Two-dimensional isotropic Heisenberg spin- S system

2.1. Cluster states and observables

In the first step one must identify the cluster. As detailed above, care should be taken to ensure that the cluster has the same point-group symmetry as the lattice. Since the calculation of

antiferromagnetism requires bipartite clusters, we select a cluster with a bipartite environment of the central site (the case of this violation will be illustrated in the example of the cluster $\sqrt{13} \times \sqrt{13}$).

The cluster Hamiltonian

$$\hat{H} = J \sum_{n\bar{\delta}} \vec{S}_n \vec{S}_{n+\bar{\delta}} = \hat{H}_u + \hat{V} \quad (1)$$

is composed of the term $\hat{V} = J \vec{S}_0 \sum_{\bar{\delta}} \vec{S}_{0+\bar{\delta}}$ describing interactions of the central spin \vec{S}_0 with the nearest neighbours at distances $\bar{\delta}$ and rest terms denoted as the Hamiltonian of the ‘environment’ \hat{H}_u . Since, by construction, the cluster retains a lattice point symmetry, its states $|iSM\Gamma\mu\rangle$ with the energies $E_{iS\Gamma}$ are labelled by the cluster total spin S with the third component M and by the irreducible representation $\Gamma\mu$ of the cluster point group. Different states with the same values SM and $\Gamma\mu$ are distinguished by the index i . In addition we need to consider the operator $O_{q1}^{1A_1} = \sum_{\bar{\delta}} (S_{0+\bar{\delta}})_q^1$ as a double irreducible tensor which transforms according to identity representation A_1 . The same arguments enable us to use the irreducible form of the central spin operator $(S_0)_q^1 \equiv (S_0)_{q1}^{1A_1}$. The part \hat{V} may be written as the inner product:

$$\hat{V} = J \sum_q (-1)^q \begin{bmatrix} A_1 & A_1 & A_1 \\ 1 & 1 & 1 \end{bmatrix} (S_0)_{q1}^{1A_1} O_{-q1}^{1A_1} \equiv [(S_0)^{1A_1} \times O^{1A_1}]_{01}^{0A_1},$$

where $\begin{bmatrix} A_1 & A_1 & A_1 \\ 1 & 1 & 1 \end{bmatrix} = 1$ is the Clebsch–Gordan coefficient of the cluster point group [32].

Let us suppose that we have found the eigenvalues $E_{i_u S_u \Gamma_u}$ and the eigenstates of the environment Hamiltonian \hat{H}_u in the form $|i_u S_u M_u \Gamma_u \mu_u\rangle$. The basis functions of the full cluster are obtained by the addition rule of spin angular momentum:

$$|i_u S_u \Gamma_u; s; SM\Gamma_u \mu_u\rangle = \sum_{\mu_u, \sigma} \begin{bmatrix} S_u & s & S \\ M_u & \sigma & M \end{bmatrix} |i_u S_u M_u \Gamma_u \mu_u\rangle |s\sigma\rangle, \quad (2)$$

where $[\dots]$ is a Clebsch–Gordan coefficient, hereinafter we use that of given in [33], and $|s\sigma\rangle$ is the wavefunction of the central spin. Since the state $|s\sigma\rangle$ is invariant under all transformations of the point-symmetry group, the cluster basis functions transform like that of the environment according to the same irreducible representations.

The calculation of matrix elements for the Hamiltonian (1) with the help of the Wigner–Eckart theorem yields (see appendix A)

$$\begin{aligned} & \langle i_u S_u \Gamma_u; s; SM\Gamma_u \mu_u | \hat{H} | i'_u S'_u \Gamma'_u; s; S' M' \Gamma'_u \mu'_u \rangle \\ &= E_{i_u S_u \Gamma_u} \delta_{i_u, i'_u} \delta_{S_u, S'_u} \delta_{\Gamma_u, \Gamma'_u} \delta_{\mu_u, \mu'_u} \delta_{S, S'} \delta_{M, M'} + J (-1)^{S'_u + S + 1/2} \begin{Bmatrix} S_u & s & S \\ s & S'_u & 1 \end{Bmatrix} \\ & \quad \times \delta_{S, S'} \delta_{M, M'} \langle s \| s \| s \rangle \langle i_u S_u \Gamma_u \| O^{1A_1} \| i'_u S'_u \Gamma'_u \rangle \delta_{\Gamma_u, \Gamma'_u} \delta_{\mu_u, \mu'_u}, \end{aligned} \quad (3)$$

where $\{\dots\}$ is a $6j$ symbol. The first reduced matrix element is $\langle s \| s \| s \rangle = \sqrt{s(s+1)(2s+1)}$ and the latter may be obtained if the environment eigenstates are known (see section 3). The energy per bond is then calculated as

$$\varepsilon_{iS\Gamma_u} = \frac{1}{z} \left(E_{iS\Gamma_u} - \sum_{i_u S_u} E_{i_u S_u \Gamma_u} |\beta_{i_u S_u \Gamma_u}^{iS\Gamma_u}|^2 \right) = \frac{1}{z} (E_{iS\Gamma_u} - \langle E_{iS\Gamma_u}^{(env)} \rangle), \quad (4)$$

where z is the number of nearest-neighbours of the central spin. The eigenfunctions

$$|iSM\Gamma\mu\rangle = \sum_{i_u S_u} \beta_{i_u S_u \Gamma_u}^{iS\Gamma} |i_u S_u \Gamma; s; SM\Gamma\mu\rangle, \quad (\Gamma\mu = \Gamma_u \mu_u) \quad (5)$$

and the energy levels $E_{iS\Gamma}$ are determined by the direct diagonalization of the cluster Hamiltonian H (equation (3)). The values $\varepsilon_{iS\Gamma}$ should be regarded as an approximation of the energy spectrum in the thermodynamical limit, whereas the energy $E_{iS\Gamma}$ divided per bond number is much less appropriate for this.

It is important to note that from equation (3) it follows that to build the cluster target state $|iSM\Gamma\mu\rangle$, we need only to know the states $|i_u S_u M_u \Gamma_u \mu_u\rangle$ of the environment with the quantum numbers $|S - s| \leq S_u \leq S + s$ and $\Gamma_u \mu_u = \Gamma \mu$.

The most important quantity typically measured in numerical simulations is the ground-state staggered magnetization M_c . The quantum mechanical observable for the z projection of the central spin is given as follows:

$$\langle iSM\Gamma\mu | S_0^z | iSM\Gamma\mu \rangle = (-1)^{1+S+s} M \sqrt{\frac{2S+1}{S(S+1)}} \langle s \| S \| s \rangle \sum_{i_u S_u} (-1)^{S_u} |\beta_{i_u S_u \Gamma_u}^{iS\Gamma}|^2 \begin{Bmatrix} S & 1 & S \\ s & S_u & s \end{Bmatrix}, \quad (6)$$

where identity (B.3) is used. The staggered magnetization M_c is determined as

$$M_c^2 = \lim_{|\vec{R}| \rightarrow \infty} 3 |\langle S^z(\vec{R}) S^z(0) \rangle|,$$

where factor 3 arises from rotational symmetry in spin space. At long distances $|\langle S^z(\vec{R}) S^z(0) \rangle| \approx \langle S^z(0) \rangle^2$ that yields our estimate of the full root-mean-square staggered magnetization per spin $M_c = \sqrt{3 \langle S_0^z \rangle^2}$.

According to equation (A.1), spin-correlation function in the states of A_1 symmetry, the ground-state symmetry as shown below, is determined as

$$\begin{aligned} \langle iSMA_1 | S_0^z S_j^z | iSMA_1 \rangle &= \frac{1}{3} \langle iSMA_1 | \vec{S}_0 \vec{S}_j | iSMA_1 \rangle = \frac{1}{3z_f} \sum_{i_u S_u} \sum_{i'_u S'_u} \\ &\times \beta_{i_u S_u A_1}^{iSA_1} \beta_{i'_u S'_u A_1}^{iSA_1} \langle s \| S \| s \rangle \langle i_u S_u \| S^{1A}(r_j) \| i'_u S'_u \rangle (-1)^{s+S+S'_u} \begin{Bmatrix} S_u & s & S \\ s & S'_u & 1 \end{Bmatrix}, \quad (7) \end{aligned}$$

where z_f is the lattice coordination number. In this calculation, it is convenient to introduce the double irreducible tensor $S_{q1}^{1A}(r_j) = \sum_j (S_j)_q^1$ summing spins at distance r_j , which transforms according to identity representation A_1 . One can see that $O_{q1}^{1A_1} = S_{q1}^{1A}(\delta)$.

2.2. Increasing cluster size

As mentioned above, the lattice point-group symmetry should be conserved with the increasing cluster size. The requirement is put into a practical computational scheme by the following algorithm. (i) At step N , we have the eigenvalues $E_{i_u S_u \Gamma_u}^{(N)}$ and eigenvectors $|i_u S_u m_u \Gamma_u \mu_u\rangle_{(N)}$ of the environment. Make a regular symmetry conserving expansion in the cluster size by adding sites from the next coordination shell. (ii) Using a scheme of coupling of angular momenta we build the set $|i_I S_I m_I\rangle$ of states with total spin S_I and third component m_I for the part that is being attached to the environment. The index i_I labels other possible quantum numbers. (iii) In general case, these functions form a basis of reducible representation of the cluster point group. Based on the projection operator technique, one build basic functions $|i_I S_I m_I \Gamma_I \mu_I\rangle$ transforming according to irreducible representations $\Gamma_I \mu_I$. (iv) Using a scheme of coupling of angular momenta build a new set $|i_I S_I m_I i_{II} S_{II} m_{II}; S_u m_u \Gamma_u \mu_u\rangle_{(N+1)}$ of states associated with the extended environment, where the notation $|i_{II} S_{II} m_{II} \Gamma_{II} \mu_{II}\rangle = |i_u S_u m_u \Gamma_u \mu_u\rangle_{(N)}$ is introduced. An interaction between the N th step environment and the part added to it can be conveniently written through the irreducible tensors $U^{1u\gamma}$ and $W^{1u_0\gamma}$ built from spin operators

of the ‘old’ and ‘new’ added parts, respectively,

$$V = J \sum_{t_u t_0} \sum_{\gamma^v} \sum_q (-1)^q \begin{bmatrix} \gamma & \gamma & A_1 \\ v & v & 1 \end{bmatrix} U_{qv}^{1t_u \gamma} W_{-qv}^{1t_0 \gamma}.$$

The indices $t_u t_0$ label different tensors of the same symmetry. The matrix elements of the extended $(N + 1)$ th step environment is

$$\begin{aligned} & \langle i_I S_I m_I i_{II} S_{II} m_{II}; S_u m_u \Gamma_u \mu_u | H_u | i'_I S'_I m'_I i'_{II} S'_{II} m'_{II}; S'_u m'_u \Gamma'_u \mu'_u \rangle \\ &= E_{i_u S_u \Gamma_u}^{(N)} \delta_{S_u S'_u} \delta_{m_u m'_u} \delta_{\Gamma_u \Gamma'_u} \delta_{\mu_u \mu'_u} + J \sum_{t_0 t_u \gamma} F(\Gamma_I \Gamma_{II} \Gamma; \Gamma'_I \Gamma'_{II} \gamma) (-1)^{S'_I + S_{II} + S_u} \\ & \times \begin{Bmatrix} S_I & S_{II} & S_u \\ S'_I & S'_{II} & 1 \end{Bmatrix} \langle i_I S_I \Gamma_I \| U^{1t_u \gamma} \| i'_I S'_I \Gamma'_I \rangle \langle i_{II} S_{II} \Gamma_{II} \| W^{1t_u \gamma} \| i'_{II} S'_{II} \Gamma'_{II} \rangle. \quad (8) \end{aligned}$$

The derivation of equation (8) and the reduced matrix elements of the operators involved in equation (8) are given in appendices A and B, respectively.

At final step, we diagonalize (8) and find the eigenvalues $E_{i_u S_u \Gamma_u}^{(N+1)}$ and eigenvectors

$$\begin{aligned} |i_u S_u m_u \Gamma_u \mu_u\rangle_{(N+1)} &= \sum \alpha_{i_I S_I \Gamma_I i_{II} S_{II} \Gamma_{II}}^{i_u S_u \Gamma_u} \begin{bmatrix} S_I & S_{II} & S_u \\ m_I & m_{II} & m_u \end{bmatrix} \begin{bmatrix} \Gamma_I & \Gamma_{II} & \Gamma_u \\ \mu_I & \mu_{II} & \mu_u \end{bmatrix} \\ & \times |i_I S_I m_I \Gamma_I \mu_I\rangle |i_{II} S_{II} m_{II} \Gamma_{II} \mu_{II}\rangle. \end{aligned}$$

The iteration is closed by recalculating reduced matrix elements of the irreducible tensors $W^{1t_u \gamma}$ in the basis of the extended environment (see appendix B). Note that following the scheme we will in some cases form intermediate clusters, unsuitable for calculations of local results, with a non-bipartite environment.

3. An example: spin-1/2 antiferromagnet on a square lattice

The spin-half antiferromagnet on a square lattice represents an optimal playground to study the strength and limitations of the method. To implement the algorithm, we need first to build wavefunctions of the environment which are predetermined by the lattice point symmetry.

To perform calculations we start with the cluster of minimal size $\sqrt{5} \times \sqrt{5}$. The sequence of clusters involved in the calculations is shown in figure 1. Within the smallest cluster, the central spin interacts with the nearest environment consisting of the spins S_{α_1} , S_{β_1} , S_{γ_1} , S_{η_1} . The spin wavefunctions of the environment with the total spin number S_u and the third component M_u may be written as follows:

$$\begin{aligned} \left| \frac{1}{2} \frac{1}{2} (S_{\alpha_1 \beta_1}) \frac{1}{2} \frac{1}{2} (S_{\gamma_1 \eta_1}) S_u M_u \right\rangle &= \sum_{m_{\alpha_1}, m_{\beta_1}, m_{\gamma_1}, m_{\eta_1}} \sum_{m_{\alpha_1 \beta_1}, m_{\gamma_1 \eta_1}} \begin{bmatrix} 1/2 & 1/2 & S_{\alpha_1 \beta_1} \\ m_{\alpha_1} & m_{\beta_1} & m_{\alpha_1 \beta_1} \end{bmatrix} \begin{bmatrix} 1/2 & 1/2 & S_{\gamma_1 \eta_1} \\ m_{\gamma_1} & m_{\eta_1} & m_{\gamma_1 \eta_1} \end{bmatrix} \\ & \times \begin{bmatrix} S_{\alpha_1 \beta_1} & S_{\gamma_1 \eta_1} & S_u \\ m_{\alpha_1 \beta_1} & m_{\gamma_1 \eta_1} & M_u \end{bmatrix} \times |1/2 m_{\alpha_1}\rangle |1/2 m_{\beta_1}\rangle |1/2 m_{\gamma_1}\rangle |1/2 m_{\eta_1}\rangle. \end{aligned}$$

In such a description, all allowed configurations are comprised by a set $|00; 00\rangle, |11; 00\rangle, |01; 1M\rangle, |10; 1M\rangle, |11; 1M\rangle, |11; 2M\rangle$, where we have dropped the spin 1/2 arguments for notation convenience. It is easy to see that the functions $|S_{\alpha_1 \beta_1} S_{\gamma_1 \eta_1}; S_u M_u\rangle$ form (in common case) a basis of reducible representation of the group D_4 (for details see appendix C):

$$\hat{g} |S_{\alpha_1 \beta_1} S_{\gamma_1 \eta_1}; S_u M_u\rangle = D_{S'_{\alpha_1 \beta_1} S'_{\gamma_1 \eta_1}, S_{\alpha_1 \beta_1} S_{\gamma_1 \eta_1}}^{(S_u)}(\hat{g}) |S'_{\alpha_1 \beta_1} S'_{\gamma_1 \eta_1}; S_u M_u\rangle.$$

The matrices $D_{\kappa, \kappa'}^{(0)}(\hat{g})$ (the upper index denotes the spin S_u) with the multi-index $\kappa = \{S_{\alpha_1 \beta_1} S_{\gamma_1 \eta_1}\}$ are readily determined and read

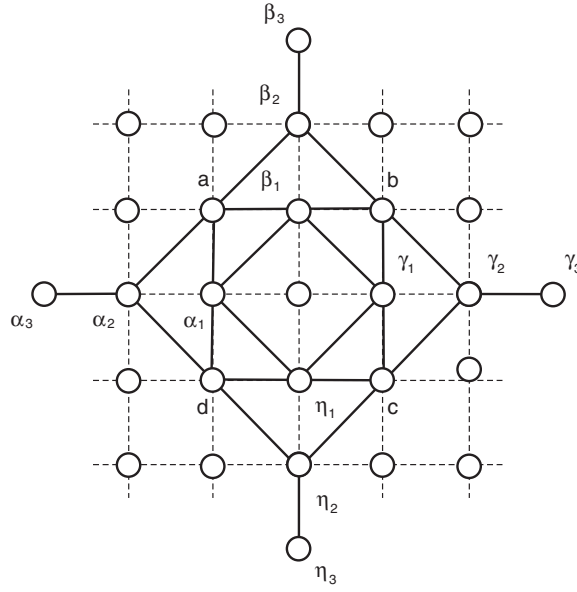


Figure 1. Clusters used in the calculations.

$$D_{\kappa,\kappa'}^{(0)}(E) = D_{\kappa,\kappa'}^{(0)}(C_4^2) = D_{\kappa,\kappa'}^{(0)}(\sigma'_v) = D_{\kappa,\kappa'}^{(0)}(\sigma''_v) = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix},$$

$$D_{\kappa,\kappa'}^{(0)}(C_4) = D_{\kappa,\kappa'}^{(0)}(C_4^3) = D_{\kappa,\kappa'}^{(0)}(C_2^x) = D_{\kappa,\kappa'}^{(0)}(C_4^y) = \begin{pmatrix} 1/2 & -\sqrt{3}/2 \\ -\sqrt{3}/2 & -1/2 \end{pmatrix}.$$

The functions $|00; 00\rangle, |11; 00\rangle$ form a basis of this two-dimensional representation. Still another representation of D_4 can be generated by means of the functions $|01; 1M\rangle, |10; 1M\rangle$ and $|11; 1M\rangle$

$$D_{\kappa,\kappa'}^{(1)}(E) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad D_{\kappa,\kappa'}^{(1)}(C_4) = \begin{pmatrix} -1/2 & -1/2 & 1/\sqrt{2} \\ -1/2 & -1/2 & -1/\sqrt{2} \\ -1/\sqrt{2} & 1/\sqrt{2} & 0 \end{pmatrix},$$

$$D_{\kappa,\kappa'}^{(1)}(C_4^2) = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}, \quad D_{\kappa,\kappa'}^{(1)}(C_4^3) = \begin{pmatrix} -1/2 & -1/2 & -1/\sqrt{2} \\ -1/2 & -1/2 & 1/\sqrt{2} \\ 1/\sqrt{2} & -1/\sqrt{2} & 0 \end{pmatrix},$$

$$D_{\kappa,\kappa'}^{(1)}(C_2^x) = \begin{pmatrix} 1/2 & 1/2 & 1/\sqrt{2} \\ 1/2 & 1/2 & -1/\sqrt{2} \\ 1/\sqrt{2} & -1/\sqrt{2} & 0 \end{pmatrix}, \quad D_{\kappa,\kappa'}^{(1)}(C_2^y) = \begin{pmatrix} 1/2 & 1/2 & -1/\sqrt{2} \\ 1/2 & 1/2 & 1/\sqrt{2} \\ -1/\sqrt{2} & 1/\sqrt{2} & 0 \end{pmatrix},$$

$$D_{\kappa,\kappa'}^{(1)}(\sigma''_v) = \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad D_{\kappa,\kappa'}^{(1)}(\sigma'_v) = \begin{pmatrix} 0 & -1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}.$$

In a similar way we find the matrices $D_{\kappa,\kappa}^{(2)}(\hat{g})$ in the basis $|11; 2M\rangle$

$$D_{\kappa,\kappa}^{(2)}(\hat{g}) = 1(\forall \hat{g} \in D_4).$$

The representations $D^{(S)}$ are the direct sums of the irreducible representations $D^{(0)} = D^{(0A_1)} \oplus D^{(0B_2)}$, $D^{(1)} = D^{(1B_1)} \oplus D^{(1E)}$, $D^{(2)} = D^{(2A_1)}$ (see appendix E). The basis functions of these irreducible representations are given by a similarity transformation

$$|S_u M_u; \Gamma \mu\rangle = \sum_{S_{\alpha_1 \beta_1}, S_{\gamma_1 \eta_1}} \hat{T}_{S_{\alpha_1 \beta_1} S_{\gamma_1 \eta_1}; \Gamma \mu}^{(S_u)} |S_{\alpha_1 \beta_1} S_{\gamma_1 \eta_1}; S_u M_u\rangle, \quad (9)$$

and the matrix $\hat{T}_{S_{\alpha_1 \beta_1} S_{\gamma_1 \eta_1}; \Gamma \mu}^{(S_u)} = \hat{T}_{S_{\alpha_1 \beta_1} S_{\gamma_1 \eta_1} S_u'; S_u \Gamma_u \mu_u} \delta_{S_u, S_u'}$ found with the aid of the projection-operator technique reads (for details see appendix D):

	$ 00; A_1 1\rangle$	$ 00; B_2 1\rangle$	$ 1M; B_1 1\rangle$	$ 1M; E 1\rangle$	$ 1M; E 2\rangle$	$ 2M; A_1 1\rangle$
$ 00; 00\rangle$	$\frac{\sqrt{3}}{2}$	$\frac{1}{2}$	0	0	0	0
$ 11; 00\rangle$	$-\frac{1}{2}$	$\frac{\sqrt{3}}{2}$	0	0	0	0
$ 01; 1M\rangle$	0	0	$\frac{1}{\sqrt{2}}$	$\frac{1}{2}$	$\frac{1}{2}$	0
$ 10; 1M\rangle$	0	0	$\frac{1}{\sqrt{2}}$	$-\frac{1}{2}$	$-\frac{1}{2}$	0
$ 11; 1M\rangle$	0	0	0	$\frac{1}{\sqrt{2}}$	$-\frac{1}{\sqrt{2}}$	0
$ 11; 2M\rangle$	0	0	0	0	0	1

Given the environment eigenfunctions $|S_u M_u; \Gamma_u \mu_u\rangle$ with the eigenvalues $E_{S_u \Gamma_u}$, the reduced matrix elements of the double irreducible tensor $O^{1A_1} = S_{\alpha_1} + S_{\beta_1} + S_{\gamma_1} + S_{\eta_1}$ can be computed straightforwardly using the Wigner–Eckart theorem and the similarity transformation (9)

$$\begin{aligned} & \begin{bmatrix} \gamma & \Gamma' & \Gamma \\ \nu & \mu' & \mu \end{bmatrix}^* \langle S \Gamma \| O^{1\gamma} \| S' \Gamma' \rangle \\ &= \sum_{S_{12}, S_{34}} \sum_{S'_{12}, S'_{34}} \hat{T}_{S_{12} S_{34} S; S \Gamma \mu}^* \hat{T}_{S'_{12} S'_{34} S'; S' \Gamma' \mu'} \langle S_{12} S_{34}; S \| O_{\nu}^{1\gamma} \| S'_{12} S'_{34}; S' \rangle, \end{aligned} \quad (10)$$

where the indices $\alpha_1, \beta_1, \gamma_1, \eta_1$ are correspondingly denoted by the numbers 1–4.

To calculate the reduced matrix element that comes onto the right-hand side of equation (10) one has to rewrite $O_{\nu}^{1\gamma}$ through the spin operators and employ their expressions for the reduced matrix elements of the spin operators:

$$\begin{aligned} \langle S_{12} S_{34}; S \| S_1 \| S'_{12} S'_{34}; S' \rangle &= (-1)^{1+S_{12}+S_{34}+S'_{12}+S'} [S_{12}, S'_{12}, S, S']^{1/2} \\ &\times \begin{Bmatrix} S'_{12} & 1 & S_{12} \\ 1/2 & 1/2 & 1/2 \end{Bmatrix} \begin{Bmatrix} S' & 1 & S \\ S_{12} & S_{34} & S'_{12} \end{Bmatrix} \langle 1/2 \| S \| 1/2 \rangle \delta_{S_{34}, S'_{34}}, \end{aligned} \quad (11)$$

$$\begin{aligned} \langle S_{12} S_{34}; S \| S_2 \| S'_{12} S'_{34}; S' \rangle &= (-1)^{1+2S_{12}+S_{34}+S'} [S_{12}, S'_{12}, S, S']^{1/2} \\ &\times \begin{Bmatrix} S'_{12} & 1 & S_{12} \\ 1/2 & 1/2 & 1/2 \end{Bmatrix} \begin{Bmatrix} S' & 1 & S \\ S_{12} & S_{34} & S'_{12} \end{Bmatrix} \langle 1/2 \| S \| 1/2 \rangle \delta_{S_{34}, S'_{34}}, \end{aligned} \quad (12)$$

$$\begin{aligned} \langle S_{12} S_{34}; S \| S_3 \| S'_{12} S'_{34}; S' \rangle &= (-1)^{1+S_{12}+2S_{34}+S} [S_{34}, S'_{34}, S, S']^{1/2} \\ &\times \begin{Bmatrix} S'_{34} & 1 & S_{34} \\ 1/2 & 1/2 & 1/2 \end{Bmatrix} \begin{Bmatrix} S' & 1 & S \\ S_{34} & S_{12} & S'_{34} \end{Bmatrix} \langle 1/2 \| S \| 1/2 \rangle \delta_{S_{12}, S'_{12}}, \end{aligned} \quad (13)$$

$$\begin{aligned} \langle S_{12} S_{34}; S \| S_4 \| S'_{12} S'_{34}; S' \rangle &= (-1)^{1+S_{12}+S_{34}+S'_{34}+S} [S_{34}, S'_{34}, S, S']^{1/2} \\ &\times \begin{Bmatrix} S'_{34} & 1 & S_{34} \\ 1/2 & 1/2 & 1/2 \end{Bmatrix} \begin{Bmatrix} S' & 1 & S \\ S_{34} & S_{12} & S'_{34} \end{Bmatrix} \langle 1/2 \| S \| 1/2 \rangle \delta_{S_{12}, S'_{12}}. \end{aligned} \quad (14)$$

Since the operator O^{1A_1} coincides with that of the environment total spin \hat{S}_u , it turns out that the matrix elements $\langle S_u \Gamma_u | O^{1A_1} | S'_u \Gamma'_u \rangle$ are diagonal:

$$\langle S_u \Gamma_u | O^{1A_1} | S'_u \Gamma'_u \rangle = \sqrt{S_u(S_u + 1)(2S_u + 1)} \delta_{S_u, S'_u} \delta_{\Gamma_u, \Gamma'_u}.$$

Table 1. Energies $E_{S\Gamma}$ and $\epsilon_{S\Gamma}$.

$S\Gamma$	$\frac{1}{2}A_1$	$\frac{1}{2}B_1$	$\frac{1}{2}B_2$	$\frac{1}{2}E$	$\frac{3}{2}A_1$	$\frac{3}{2}B_1$	$\frac{3}{2}E$	$\frac{5}{2}A_1$
$E_{S\Gamma}$	0	-J	0	-J	$-\frac{3}{2}J$	$\frac{1}{2}J$	$\frac{1}{2}J$	J
$\epsilon_{S\Gamma}$	0	$-\frac{1}{4}J$	0	$-\frac{1}{4}J$	$-\frac{3}{8}J$	$\frac{1}{8}J$	$\frac{1}{8}J$	$\frac{1}{4}J$

As a consequence, one may check that this property holds for the Hamiltonian of the total cluster:

$$\begin{aligned} & \left\langle S_u \Gamma_u; \frac{1}{2}; S M \Gamma_u \mu_u | \hat{H} | S'_u \Gamma'_u; \frac{1}{2}; S' M' \Gamma'_u \mu'_u \right\rangle \\ &= J(-1)^{S'_u+S+1/2} \begin{Bmatrix} S_u & 1/2 & S \\ 1/2 & S'_u & 1 \end{Bmatrix} \sqrt{\frac{3}{2} S_u(S_u+1)(2S_u+1)} \delta_{S,S'} \delta_{M,M'} \delta_{\Gamma_u, \Gamma'_u} \delta_{\mu_u, \mu'_u} \delta_{S_u, S'_u}. \end{aligned}$$

A direct calculation shows that the ground state belongs to the Hilbert space sector with $S = 3/2$ and $\Gamma = A_1$. Hence, only the environment state with $S\Gamma = 1A_1$ is needed to find the ground-state energy (see table 1).

Let us now consider the next step, an expansion of the current environment block due to the next coordination sphere of radius $\sqrt{2}$. After an addition of four spins S_a, S_b, S_c, S_d , the cluster becomes a square of size 3×3 with the bipartite environment of the central site (figure 1). The basis associated with the added part is

$$\begin{aligned} & \left| \frac{1}{2} \frac{1}{2} (S_{ab}) \frac{1}{2} \frac{1}{2} (S_{cd}) S_I M_I \right\rangle \\ &= \sum_{m_a, m_b, m_c, m_d} \sum_{m_{ab}, m_{cd}} \begin{bmatrix} 1/2 & 1/2 & S_{ab} \\ m_a & m_b & m_{ab} \end{bmatrix} \begin{bmatrix} 1/2 & 1/2 & S_{cd} \\ m_c & m_d & m_{cd} \end{bmatrix} \begin{bmatrix} S_{ab} & S_{cd} & S_I \\ m_{ab} & m_{cd} & M_I \end{bmatrix} \\ & \times |1/2 m_a\rangle |1/2 m_b\rangle |1/2 m_c\rangle |1/2 m_d\rangle. \end{aligned} \quad (15)$$

Repeating the basic steps in the approach we obtain the symmetry adapted basis $|S_I M_I; \Gamma_I \mu_I\rangle$. The matrix of corresponding similarity transformation has the form

$$\begin{array}{cccccc} |00; A_1 1\rangle & |00; B_1 1\rangle & |1M; B_2 1\rangle & |1M; E 1\rangle & |1M; E 2\rangle & |2M; A_1 1\rangle \\ |00; 00\rangle & \frac{\sqrt{3}}{2} & \frac{1}{2} & 0 & 0 & 0 \\ |11; 00\rangle & -\frac{1}{2} & \frac{\sqrt{3}}{2} & 0 & 0 & 0 \\ |01; 1M\rangle & 0 & 0 & \frac{1}{\sqrt{2}} & 0 & \frac{1}{\sqrt{2}} \\ |10; 1M\rangle & 0 & 0 & \frac{1}{\sqrt{2}} & 0 & -\frac{1}{\sqrt{2}} \\ |11; 1M\rangle & 0 & 0 & 0 & 1 & 0 \\ |11; 2M\rangle & 0 & 0 & 0 & 0 & 1. \end{array}$$

The environment Hamiltonian includes only interactions between the first and second coordination spheres:

$$\hat{H}_u = J[\vec{S}_{\alpha_1}(\vec{S}_d + \vec{S}_a) + \vec{S}_{\beta_1}(\vec{S}_a + \vec{S}_b) + \vec{S}_{\gamma_1}(\vec{S}_b + \vec{S}_c) + \vec{S}_{\eta_1}(\vec{S}_c + \vec{S}_d)]. \quad (16)$$

We now introduce the cluster irreducible tensors $W_{q\mu}^{1\Gamma}$ and $U_{q\mu}^{1\Gamma}$ transforming according to representations $\Gamma\mu$ of the point-symmetry group D_4 (for details see appendix D):

$$\begin{aligned} U_{q1}^{1A_1} &= \frac{1}{\sqrt{2}}(S_{aq} + S_{bq} + S_{cq} + S_{dq}), & U_{q1}^{1B_2} &= \frac{1}{\sqrt{2}}(S_{aq} - S_{bq} + S_{cq} - S_{dq}), \\ U_{q1}^{1E} &= \frac{1}{\sqrt{2}}(S_{aq} + S_{bq} - S_{cq} - S_{dq}), & U_{q2}^{1E} &= \frac{1}{\sqrt{2}}(S_{aq} - S_{bq} - S_{cq} + S_{dq}), \\ W_{q1}^{1A_1} &= \frac{1}{\sqrt{2}}(S_{\alpha_1 q} + S_{\beta_1 q} + S_{\gamma_1 q} + S_{\eta_1 q}), & W_{q1}^{1B_1} &= \frac{1}{\sqrt{2}}(S_{\alpha_1 q} - S_{\beta_1 q} + S_{\gamma_1 q} - S_{\eta_1 q}), \\ W_{q1}^{1E} &= (S_{\alpha_1 q} - S_{\gamma_1 q}), & W_{q2}^{1E} &= (S_{\eta_1 q} - S_{\beta_1 q}) \end{aligned} \quad (17)$$

and then rewrite equation (16) as

$$H_u = J \sum_{\gamma\nu} \sum_q (-1)^q \begin{bmatrix} \gamma & \gamma & A_1 \\ \nu & \nu & 1 \end{bmatrix} U_{q\mu}^{1\gamma} W_{-q\mu}^{1\gamma} = J \sum_{\gamma} [U^{1\gamma} \times W^{1\gamma}]_{01}^{0A_1}. \quad (18)$$

The reduced matrix elements of the irreducible operators that appear in equation (8) can be obtained exactly from result (10):

$$\begin{aligned} \langle S_I \Gamma_I \| U^{1A_1} \| S'_I \Gamma'_I \rangle &= \frac{1}{\sqrt{2}} \langle S_I \| S \| S_I \rangle \delta_{S_I, S'_I} \delta_{\Gamma_I, \Gamma'_I}, & \langle S_{II} \Gamma_{II} \| W^{1A_1} \| S'_{II} \Gamma'_{II} \rangle \\ &= \frac{1}{\sqrt{2}} \langle S_{II} \| S \| S_{II} \rangle \delta_{S_{II}, S'_{II}} \delta_{\Gamma_{II}, \Gamma'_{II}}, \end{aligned} \quad (19)$$

$$\langle S_I \Gamma_I \| U^{1E} \| S'_I \Gamma'_I \rangle = \begin{bmatrix} 0A_1 & 0B_1 & 1B_2 & 1E & 2A_1 \\ 0A_1 & 0 & 0 & 0 & \sqrt{2} & 0 \\ 0B_1 & 0 & 0 & 0 & -\sqrt{6} & 0 \\ 1B_2 & 0 & 0 & 0 & -\sqrt{6} & 0 \\ 1E & -1 & \sqrt{3} & -\sqrt{3} & 0 & -\sqrt{5} \\ 2A_1 & 0 & 0 & 0 & \sqrt{10} & 0 \end{bmatrix},$$

$$\langle S_{II} \Gamma_{II} \| W^{1E} \| S'_{II} \Gamma'_{II} \rangle = \begin{bmatrix} 0A_1 & 0B_2 & 1B_1 & 1E & 2A_1 \\ 0A_1 & 0 & 0 & 0 & \sqrt{2} & 0 \\ 0B_2 & 0 & 0 & 0 & \sqrt{6} & 0 \\ 1B_1 & 0 & 0 & 0 & -\sqrt{6} & 0 \\ 1E & -1 & -\sqrt{3} & -\sqrt{3} & 0 & -\sqrt{5} \\ 2A_1 & 0 & 0 & 0 & \sqrt{10} & 0 \end{bmatrix}.$$

To compute matrix elements of the Hamiltonian H_u , we construct the basis

$$\begin{aligned} |i_I S_I \Gamma_I i_{II} S_{II} \Gamma_{II}; S_u M_u \Gamma_u \mu_u\rangle &= \sum_{m_I, m_{II}} \sum_{\mu_I, \mu_{II}} \begin{bmatrix} S_I & S_{II} & S_u \\ m_I & m_{II} & M_u \end{bmatrix} \begin{bmatrix} \Gamma_I & \Gamma_{II} & \Gamma_u \\ \mu_I & \mu_{II} & \mu_u \end{bmatrix} \\ &\times |i_I S_I m_I \Gamma_I \mu_I\rangle |i_{II} S_{II} m_{II} \Gamma_{II} \mu_{II}\rangle \end{aligned} \quad (20)$$

formed from the eigenstates $|i_I S_I m_I \Gamma_I \mu_I\rangle$ and $|i_{II} S_{II} m_{II} \Gamma_{II} \mu_{II}\rangle$ of the ‘new’ and ‘old’ added parts, correspondingly. Then we obtain using equation (A.5) the expression similar to equation (8) with $E_{i_I S_I \Gamma_I} = E_{i_{II} S_{II} \Gamma_{II}} = 0$. Applying exact diagonalization to the Hamiltonian H_u one can then find the eigenfunctions

$$|i_u S_u \Gamma_u \mu_u\rangle = \sum \alpha_{i_I S_I \Gamma_I; i_{II} S_{II} \Gamma_{II}}^{i_u S_u \Gamma_u} |i_I S_I \Gamma_I i_{II} S_{II} \Gamma_{II}; S_u M_u \Gamma_u \mu_u\rangle$$

and the energy spectrum $E_{i_u S_u \Gamma_u}$ of the environment. By using the recursion relation (for details see (B.5) in appendix B)

$$\begin{aligned} \langle i_u S_u \Gamma_u \| O^{1A_1} \| i'_u S'_u \Gamma'_u \rangle &= \delta_{\Gamma_u, \Gamma'_u} \sum_{i_I, S_I, \Gamma_I} \sum_{i'_{II}, S'_{II}, \Gamma'_{II}} \sum_{i_{II}, S_{II}, \Gamma_{II}} \alpha_{i_I S_I \Gamma_I; i_{II} S_{II} \Gamma_{II}}^{i_u S_u \Gamma_u} \alpha_{i_I S_I \Gamma_I; i'_{II} S'_{II} \Gamma'_{II}}^{i'_u S'_u \Gamma'_u} \\ &\times (-1)^{1+S_I+S'_{II}+S_u} [S_u, S'_u]^{1/2} \begin{Bmatrix} S_u & 1 & S'_u \\ S'_{II} & S_I & S_{II} \end{Bmatrix} \langle i_{II} S_{II} \Gamma_{II} \| O^{1A_1} \| i'_{II} S'_{II} \Gamma'_{II} \rangle \end{aligned} \quad (21)$$

one finds the reduced matrix elements in the environment basis $|i_u S_u A_1\rangle$ that come into the matrix of the total cluster (3).

Formulae (3)–(5) allow us to obtain any of the possible 54 square cluster states. Our calculation shows that the ground state belongs to the Hilbert space sector with $S = 1/2$ and $\Gamma = A_1$. Hence, only the environment states with $S\Gamma = 0A_1, 1A_1$ are needed for the

Table 2. Environment states of symmetry $0A_1$.

i_u	$\alpha_{0A_1 0A_1}^{i_u 0A_1}$	$\alpha_{1E 1E}^{i_u 0A_1}$	$\alpha_{2A_1 2A_1}^{i_u 0A_1}$	$E_{i_u 0A_1}$
1	0.071	0.449	0.890	$-3.651J$
2	0.569	0.715	-0.406	$-0.726J$
3	-0.819	0.535	-0.205	$0.377J$

Table 3. Environment states of symmetry $1A_1$.

i_u	$\alpha_{0A_3 1A_3}^{i_u 1A_1}$	$\alpha_{1A_4 0A_4}^{i_u 1A_1}$	$\alpha_{1E 1E}^{i_u 1A_1}$	$\alpha_{2A_1 2A_1}^{i_u 1A_1}$	$E_{i_u 1A_1}$
1	0.153	0.153	-0.478	-0.851	$-3.128J$
2	-0.470	-0.470	0.566	-0.487	$-1.202J$
3	-0.505	-0.505	-0.672	0.196	$1.330J$
4	-0.707	0.707	0	0	0

Table 4. Data on the ground state of the cluster 3×3 .

$\beta_{10A_1}^g$	$\beta_{20A_1}^g$	$\beta_{30A_1}^g$	$\beta_{11A_1}^g$	$\beta_{21A_1}^g$	$\beta_{31A_1}^g$	$\beta_{41A_1}^g$	E_g
-0.712	0.044	0.010	-0.695	0.0048	-0.090	0.011	-4.749

evaluation of the ground-state energy. Below we summarize the results obtained for this particular case.

Using equation (8) and the explicit expressions for the nonzero sums of Clebsch–Gordan coefficients of the point group D_4 (see equation (A.3) in appendix A)

$$F(A_1 A_1 A_1; A_1 A_1 A_1) = F(E E A_1; A_1 A_1 E) = F(E E A_1; E E A_1) = 1, \\ F(A_1 A_1 A_1; E E E) = 1/2,$$

we obtain

$$\hat{H}_u^{(0A_1)} = \begin{bmatrix} 0 & -\frac{1}{\sqrt{3}}J & 0 \\ -\frac{1}{\sqrt{3}}J & -J & -\sqrt{\frac{5}{3}}J \\ 0 & -\sqrt{\frac{5}{3}}J & -3J \end{bmatrix}$$

in the basis of the states $|0A_1 0A_1; 00A_1\rangle, |1E 1E; 00A_1\rangle, |2A_1 2A_1; 00A_1\rangle$. The diagonalization of $\hat{H}_u^{(0A_1)}$ yields three states of the $0A_1$ symmetry (see table 2).

As for the \hat{H}_u operator with $S_u = 1$, we have the following matrix representation, with the same considerations as for the $\hat{H}_u^{(0A_1)}$ operator:

$$\hat{H}_u^{(1A_1)} = \begin{bmatrix} 0 & 0 & J & 0 \\ 0 & 0 & J & 0 \\ J & J & -\frac{1}{2}J & -\frac{\sqrt{5}}{2}J \\ 0 & 0 & -\frac{\sqrt{5}}{2}J & -\frac{5}{2}J \end{bmatrix}$$

in the basis $|0B_1 1B_1; 1MA_1\rangle, |1B_2 0B_2; 1MA_1\rangle, |1E 1E; 1MA_1\rangle, |2A_1 2A_1; 1MA_1\rangle$. The states of $1A_1$ symmetry are listed in table 3.

By using the recursion relation (21) with the starting value (19), one finds the reduced matrix elements in the environment basis $|i_u S_u A_1\rangle$. Plugging them into equation (3) we get the target states $|i \frac{1}{2} MA_1\rangle$ [see equation (5)] of the cluster and their energies $E_{i \frac{1}{2} A_1}$ ($i = 1 \dots 7$). The number of states involved in determining the cluster ground state equals 7 (see table 4).

Now we list the results for observables. The energy per bond found with the help of equation (4) is $\varepsilon_g = -0.3442$ J. This result may be compared to those results of QMC [7] $\varepsilon_g = -0.3347$ J and DMRG $\varepsilon_g = -0.32679$ J for lattice of size 20×20 and for number of DMRG states 150 [6]. (Extrapolation of the DMRG results in the infinite-lattice limit yields $\varepsilon_g = -0.3321$ J). The best available DCM [21], CCM [26] and RSRG-EI [19] results are -0.33486 J, -0.33308 J and -0.33409 J, respectively. Using (6), we get the ground-state expectation value of the z component of the central spin $\langle S_0^z \rangle_0 = 0.173$ and the staggered magnetization $M = \sqrt{3\langle S_0^z \rangle_0^2} = 0.299$. For comparison, the extrapolated QMC result for the lattice magnetization $M = 0.3070$. We also provide an estimate of the spin–spin correlation functions (7):

$$\langle S_0^z S_{r=1}^z \rangle = -0.115, \quad \langle S_0^z S_{r=\sqrt{2}}^z \rangle = 0.073.$$

These estimates should be compared with the known results -0.1116 and 0.0637 , correspondingly [27].

We have made a preliminary calculations by using the small cluster 3×3 and one can see that an accuracy of the results is still insufficient. However, we have established the following important features:

- (i) The ground state of the system belongs to identity representation A_1 .
- (ii) The lowest lying environment states of the same point symmetry give a contribution to the ground state of the system with the largest weight $|\beta_{10A_1}^g|^2 + |\beta_{11A_1}^g|^2 \approx 0.989$. One can see the coefficients β^2 by noting that the diagonal matrix elements of the reduced density matrix in DMRG language [2].
- (iii) A comparison of the ground-state energy per bond as calculated by $E_g/12 = -0.396$ J and its infinite-lattice approximation (4) within our approach shows that we produce a better result.

At further step, the procedure is repeated and the environment block grows by adding the coordination sphere of radius 2. When the new spins $\vec{S}_{\alpha_2}, \vec{S}_{\beta_2}, \vec{S}_{\gamma_2}, \vec{S}_{\eta_2}$ of the sphere are added, the cluster transforms into the rhombus of size $\sqrt{13} \times \sqrt{13}$. The cluster has the non-bipartite environment; hence, it is instructive to study this case to examine the effect of non-biparticity.

The Hamiltonian of the new environment decomposes as

$$\hat{H}_u = H_u(0) + J(\vec{S}_{\alpha_1}\vec{S}_{\alpha_2} + \vec{S}_{\beta_1}\vec{S}_{\beta_2} + \vec{S}_{\gamma_1}\vec{S}_{\gamma_2} + \vec{S}_{\eta_1}\vec{S}_{\eta_2}). \quad (22)$$

$H_u(0)$ contains all interactions within the ‘old’ environment, and the second term describes all couplings between this part and the added sites.

The irreducible tensors built from the added spins are the same as those of the first coordination sphere (17):

$$\begin{aligned} W_{q1}^{1A_1} &= \frac{1}{\sqrt{2}}(S_{\alpha_2q} + S_{\beta_2q} + S_{\gamma_2q} + S_{\eta_2q}), & W_{q1}^{1B_1} &= \frac{1}{\sqrt{2}}(S_{\alpha_2q} - S_{\beta_2q} + S_{\gamma_2q} - S_{\eta_2q}), \\ W_{q1}^{1E} &= (S_{\alpha_2q} - S_{\gamma_2q}), & W_{q2}^{1E} &= (S_{\eta_2q} - S_{\beta_2q}). \end{aligned} \quad (23)$$

One can then cast the Hamiltonian (22) in a more amenable form

$$\hat{H}_u = \hat{H}_u(0) + \frac{1}{2}J[U^{1A_1} \times W^{1A_1}]_{01}^{0A_1} + \frac{1}{2}J[U^{1B_1} \times W^{1B_1}]_{01}^{0A_1} + \frac{1}{\sqrt{2}}J[U^{1E} \times W^{1E}]_{01}^{0A_1},$$

where $U^{1\gamma}$ are given by

$$\begin{aligned} U_{q1}^{1A_1} &= \frac{1}{\sqrt{2}}(S_{\alpha_1q} + S_{\beta_1q} + S_{\gamma_1q} + S_{\eta_1q}), & U_{q1}^{1B_1} &= \frac{1}{\sqrt{2}}(S_{\alpha_1q} - S_{\beta_1q} + S_{\gamma_1q} - S_{\eta_1q}), \\ U_{q1}^{1E} &= (S_{\alpha_1q} - S_{\gamma_1q}), & U_{q2}^{1E} &= (S_{\eta_1q} - S_{\beta_1q}). \end{aligned} \quad (24)$$

The matrices formed from the reduced matrix elements of $W^{1\gamma}$ tensor coincide with (19). To find those of $U^{1\gamma}$ tensor we use equation (B.5). The expressions mentioned (19) are used to initialize the calculations.

From direct calculations one can show that the quantum numbers $S = 5/2$ and $\Gamma = A_1$ are attached to the ground state of the rhombus. This state is formed from 41 environment states with the symmetry $S\Gamma_u = 2A_1$ and 22 states of symmetry $S\Gamma_u = 3A_1$. Numerical diagonalization gives the cluster ground-state energy $E_g(\frac{5}{2}A_1) = -5.779$ J that yields the ground-state energy per bond $\varepsilon_g = -0.30925$ J in the thermodynamic limit. If we compare this result with that of QMC, we see that the agreement becomes worse. Nevertheless, the conclusions made for the square cluster 3×3 hold: (i) both the ground state of the environment and that of the total cluster have the lattice point symmetry A_1 . (ii) The largest weight (is of the order 0.993) into the sum of diagonal elements in the density matrix comes from three lowest lying $2A_1$ states and one state of symmetry $3A_1$, whereas the total number of states is 63.

Monitoring energies per bond $\varepsilon_{iS\Gamma}$ for the total cluster spectrum $E_{iS\Gamma}$, we found that the minimal value $\varepsilon_{\min} \approx -0.3229$ J is reached for the lowest state of symmetry $\frac{3}{2}A_1$, however, $E(\frac{3}{2}A_1) > E_g = E(\frac{5}{2}A_1)$. A similar situation, when a minimal energy per bond belongs to a higher lying state, has been early observed in the DMRG study of antiferromagnetic chains [2]. Despite the number of sites in the cluster $\sqrt{13} \times \sqrt{13}$ is greater than that of in the cluster 3×3 , we see that the result for ε_{\min} deteriorates compared to the QMC value -0.3347 J. Close inspection allows us to suggest that this is because we are working on the cluster with a non-bipartite environment.

To proceed with increasing cluster size and satisfy the biparticity requirement we should take the square cluster 5×5 in the next step. For the 24-site environment of the cluster, an exact-diagonalization calculation of the *total* spectrum is not possible at present and so, to move on to the next-larger system, we have to elaborate a procedure for determining the states giving the best approximation to true environment states. To solve the problem and implement the condition of bipartite environment we take a system in the form of ‘decorated cross’ obtained from the former cluster $\sqrt{13} \times \sqrt{13}$ by adding four spins $\vec{S}_{\alpha_3}, \vec{S}_{\beta_3}, \vec{S}_{\gamma_3}, \vec{S}_{\eta_3}$ (figure 1). The form makes equal a number of sites in both sublattices, though it incorporates eight sites that are being attached to the cluster by single-lattice bonds. At the same time, exact diagonalization of the cluster $\sqrt{17} \times \sqrt{17}$ is allowed; hence we compare the exact diagonalization results with those obtained from a symmetry-based truncation procedure and analyse a truncation error on a number of states kept. Since the cluster increasing is similar to that used in the previous step, we present only the results of calculations. The ground state of the extended cluster environment has the symmetry $0A_1$. The total number of states with the same symmetry is 194. Together with 439 $1A_1$ states of the environment they form a ground state of the total cluster labelled by the symmetry numbers $\frac{1}{2}A_1$. Results for the ground-state energy per bond $\varepsilon = -0.3304$, the staggered magnetization $m = 0.305$ and the spin–spin correlation functions $\langle S_0^z S_{r=1}^z \rangle = -0.1101$, $\langle S_0^z S_{r=\sqrt{2}}^z \rangle = 0.0615$ agree well with the mentioned ED and QMC results and are much better than those obtained for the square cluster 3×3 . A deviation from the ED result is found for $\langle S_0^z S_{r=2}^z \rangle = 0.0169$. This discrepancy arises from finite size effects and an imperfect topology of the cluster.

We now describe the low-energy spectrum of the environment. As the dynamics of the Néel order parameter is one of the free rotators, the low-energy levels scale as $E(S) \sim S(S+1)/N$, where the inertia of that rotator is proportional to the number of sites [28, 29]. The environment lowest energy levels (tower of states) belonging to different irreducible representations of the lattice point group are shown in figure 2 for different S

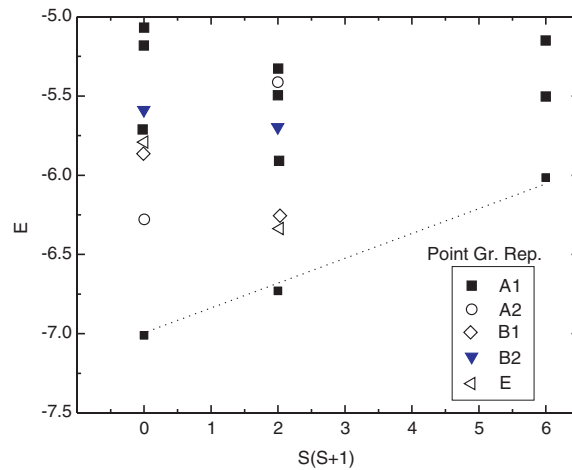


Figure 2. The lowest energy spectrum of the environment for the cluster $\sqrt{17} \times \sqrt{17}$ on the square lattice. The $SU(2)$ symmetry breaks and a long-range Néel order appear as a set of A_1 states with an energy scaling as $E(S) \sim S(S+1)$ (dashed line). The symbols represent the irreducible representations of the different eigenstates.

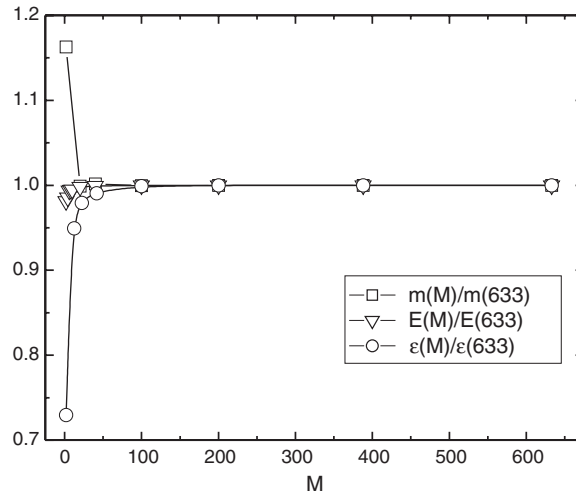


Figure 3. The cluster ground-state energy E , the energy per bond ϵ , and the staggered magnetization m convergence for the $\sqrt{17} \times \sqrt{17}$ cluster versus number of environment states kept.

sectors. The $SU(2)$ breaking due to the long-range Néel order appears as a set of A_1 states, lying off from other levels, with an energy scaling as $E(S) \sim S(S+1)$.

In the remainder of this section, we describe a version of the truncation procedure. The main idea will be illustrated on an example of the ground-state properties. An inspection of results for the current and previous clusters reveals that one has to take the lowest lying environment eigenstates both in the $0A_1$ and $1A_1$ sectors. As for the number of kept states it seems to be most simple to take M states equally from the both subspaces, albeit the choice may not be optimal. To prove that this concept works we recalculate the observables found above on the various numbers of environment states kept (see table 5). As can be seen from figure 3 the convergence of the results is exponentially fast in M . Merely keeping 100 basis states may

Table 5. Convergence of the ground-state properties versus the number of environment states kept.

$M(0A_1)$	$M(1A_1)$	E_0/J	ε/J	m	$\langle S_0^z S^z(1) \rangle$	$\langle S_0^z S^z(\sqrt{2}) \rangle$
1	1	-7.9010	-0.2410	0.354 897	-0.080 333	0.065 647
5	5	-8.1018	-0.3136	0.304 148	-0.104 533	0.071 420
10	10	-8.1282	-0.3238	0.304 928	-0.107 933	0.073 201
20	20	-8.1378	-0.3279	0.305 707	-0.109 300	0.073 872
50	50	-8.1425	-0.3301	0.305 101	-0.110 033	0.074 247
100	100	-8.1429	-0.3303	0.305 187	-0.110 100	0.074 289
194	194	-8.1430	-0.3304	0.305 187	-0.110 133	0.074 300

be as efficient as keeping of all 633 environment states intact. We regard the resulting better than 0.01% agreement for ε and m as support for the efficiency of our truncation procedure. We note that the truncation procedure becomes inevitable for the next step system of size 5×5 with a bipartite environment, when the ground-state subspace has a dimension 93034.

4. Conclusions

In this paper, we present a quantum cluster solver for the spin- S Heisenberg model on a two-dimensional lattice. The formalism is based on the real-space renormalization procedure and uses the lattice point group-theoretical analysis and non-Abelian $SU(2)$ spin symmetry technique. Let us summarize advantages of the approach as follows:

- (i) The cluster spin states are decomposed into parts belonging to different irreducible representations of the lattice point group and to different values of the total spin. Due to the embedded group-theoretical analysis, our approach can handle each of the cluster target states independently that offers a distinct advantage for parallel computation.
- (ii) An extension of MPM destined for quantum spin chains to higher dimensions has inspired the construction of variational methods for the ground states of 2D spin Hamiltonians (vertex state models [34], tensor product variational approach [35], tensor product ansatz [36]). Since, the trial states are represented by the two-dimensional product of local weights, these approaches are faced with severe limitations concerning their applicability because of the relation between a spin value and lattice topology. The shortcoming lacks in our formalism.
- (iii) Large sparse-matrix diagonalization algorithms (Lanczos technique, for example) used in DMRG and ED methods converge to maximum and minimum eigenvalues of a model Hamiltonian, i.e. to eigenvalues at the edges of the spectrum. Our approach gives access to eigenstates of an entire spectrum.
- (iv) Combined with a decimation procedure of the environment states like those used in the DMRG technique the group-theoretical analysis allows us to overcome the exponential growth of computational efforts with an increase of the system size. Our approach using the total spin S and the irrep index $\Gamma\mu$ as good quantum numbers yields a rather reliable truncation procedure of the Hilbert space of the model Hamiltonian.
- (v) Calculation of observables for the central spin involving a density matrix of the environment reduces edge effects which are inevitable on finite-size clusters.

The major drawback of the formalism is that it does not allow an easy implementation: a complexity in construction of basic sets via the repeated evaluation of $6j$ and 6Γ symbols, the calculation involves two matrix diagonalizations etc. The performance gains from implementing the $SU(2)$ and lattice point symmetries are not impressive in comparison with gains from exploiting just the simple $U(1)$ symmetry leading to total magnetization as a good

quantum number. Their use in studies with larger clusters without truncation cannot help to alleviate the problem of the exponential growth of computational efforts.

In the method that we suggest, short-range correlations on the scale of the cluster are taken into account, while correlations on a scale larger than the cluster size are neglected. To overcome this shortcoming we need to restore translational symmetry of the lattice. The results of these investigations will be reported elsewhere. In this connection, we note that the translational invariance holds for the DCM and CCM methods.

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Appendix A

Let $|i_I S_I \Gamma_I i_{II} S_{II} \Gamma_{II}; S M \Gamma \mu\rangle$ is a state with total spin S , third component M , and transforming according to irreducible representation $\Gamma \mu$. This state appears in the tensor product decomposition $(i_I S_I \Gamma_I) \times (i_{II} S_{II} \Gamma_{II})$, where $(i S \Gamma)$ denotes a state with total spin S , irreducible representation Γ and i labels other possible quantum numbers.

We need to compute the matrix element

$$\begin{aligned} & \langle i_I S_I \Gamma_I i_{II} S_{II} \Gamma_{II}; S M \Gamma \mu | [U^{1\gamma} \times W^{1\gamma}]_{01}^{0A_1} | i'_I S'_I \Gamma'_I i'_{II} S'_{II} \Gamma'_{II}; S' M' \Gamma' \mu' \rangle \\ &= \sum_{qv} \sum_{\{m, \mu\}} (-1)^q \begin{bmatrix} \gamma & \gamma & A_1 \\ \nu & \nu & 1 \end{bmatrix} \begin{bmatrix} \Gamma_I & \Gamma_{II} & \Gamma \\ \mu_I & \mu_{II} & \mu \end{bmatrix}^* \\ & \times \begin{bmatrix} \Gamma'_I & \Gamma'_{II} & \Gamma' \\ \mu'_I & \mu'_{II} & \mu' \end{bmatrix} \begin{bmatrix} S_I & S_{II} & S \\ m_I & m_{II} & M \end{bmatrix} \begin{bmatrix} S'_I & S'_{II} & S' \\ m'_I & m'_{II} & M' \end{bmatrix} \\ & \times \langle i_I S_I m_I \Gamma_I \mu_I | U_{qv}^{1\gamma} | i'_I S'_I m'_I \Gamma'_I \mu'_I \rangle \\ & \times \langle i_{II} S_{II} m_{II} \Gamma_{II} \mu_{II} | W_{-qv}^{1\gamma} | i'_{II} S'_{II} m'_{II} \Gamma'_{II} \mu'_{II} \rangle. \end{aligned} \quad (\text{A.1})$$

The Wigner–Eckart theorem for a double irreducible tensor reads

$$\begin{aligned} & \langle i_I S_I m_I \Gamma_I \mu_I | W_{qv}^{1\gamma} | i'_I S'_I m'_I \Gamma'_I \mu'_I \rangle \\ &= (-1)^{S_I - m_I} \begin{pmatrix} S_I & 1 & S'_I \\ -m_I & q & m'_I \end{pmatrix} \begin{bmatrix} \gamma & \Gamma'_I & \Gamma_I \\ \nu & \mu'_I & \mu_I \end{bmatrix}^* \langle i_I S_I \Gamma_I || W^{1\gamma} || i'_I S'_I \Gamma'_I \rangle, \end{aligned} \quad (\text{A.2})$$

where the $3j$ symbol is related to the Clebsch–Gordan coefficient by

$$\begin{pmatrix} S_1 & S_2 & S_3 \\ m_1 & m_2 & m_3 \end{pmatrix} = \frac{1}{\sqrt{2S_3 + 1}} (-1)^{S_1 + S_2 - m_3} \begin{bmatrix} S_1 & S_2 & S_3 \\ m_1 & m_2 & -m_3 \end{bmatrix}.$$

A full contraction of five Clebsch–Gordan coefficients of the point group may be written via the 6Γ symbol:

$$\begin{aligned} F(\Gamma_I \Gamma_{II} \Gamma; \Gamma'_I \Gamma'_{II} \gamma) &= \sum_{\nu \mu_I \mu_{II} \mu'_I \mu'_{II}} \begin{bmatrix} \gamma & \gamma & A_1 \\ \nu & \nu & 1 \end{bmatrix} \begin{bmatrix} \Gamma_I & \Gamma_{II} & \Gamma \\ \mu_I & \mu_{II} & \mu \end{bmatrix}^* \\ & \times \begin{bmatrix} \Gamma'_I & \Gamma'_{II} & \Gamma' \\ \mu'_I & \mu'_{II} & \mu' \end{bmatrix} \begin{bmatrix} \gamma & \Gamma'_I & \Gamma_I \\ \nu & \mu'_I & \mu_I \end{bmatrix}^* \begin{bmatrix} \gamma & \Gamma'_{II} & \Gamma_{II} \\ \nu & \mu'_{II} & \mu_{II} \end{bmatrix}^* \\ & \sim \begin{Bmatrix} \Gamma_I & \Gamma_{II} & \Gamma \\ \Gamma'_{II} & \Gamma'_I & \gamma \end{Bmatrix} \begin{bmatrix} A_1 & \Gamma' & \Gamma \\ 1 & \mu' & \mu \end{bmatrix} \delta_{\Gamma \Gamma'} \delta_{\mu \mu'}, \end{aligned} \quad (\text{A.3})$$

however, it is more convenient to find directly this sum.

Substituting (A.2) into (A.1) and performing the sum with the aid of equation (A.3) and the formula (see [33], for example)

$$\begin{aligned} & \sum_{\chi\psi\rho\sigma\tau} (-1)^{p-\psi+q-\chi+r-\rho+s-\sigma+t-\tau} \begin{pmatrix} p & a & q \\ \psi & -\alpha & \chi \end{pmatrix} \begin{pmatrix} q & r & t \\ -\chi & \rho & \tau \end{pmatrix} \\ & \times \begin{pmatrix} r & a & s \\ -\rho & \alpha' & \sigma \end{pmatrix} \begin{pmatrix} s & p & t \\ -\sigma & -\psi & -\tau \end{pmatrix} = \frac{(-1)^{a-\alpha}}{(2a+1)} \begin{Bmatrix} q & p & a \\ s & r & t \end{Bmatrix} \delta_{aa'} \delta_{\alpha\alpha'}, \end{aligned} \quad (\text{A.4})$$

we get finally

$$\begin{aligned} & \langle i_I S_I \Gamma_I i_{II} S_{II} \Gamma_{II}; SM \Gamma \mu | [U^{1\gamma} \times W^{1\gamma}]_{01}^{0A_1} | i'_I S'_I \Gamma'_I i'_{II} S'_{II} \Gamma'_{II}; S' M' \Gamma' \mu' \rangle \\ & = \delta_{SS'} \delta_{MM'} \delta_{\Gamma\Gamma'} \delta_{\mu\mu'} (-1)^{S'_I+S'_{II}+S} \begin{Bmatrix} S_I & S_{II} & S \\ S'_{II} & S'_I & 1 \end{Bmatrix} F(\Gamma_I \Gamma_{II} \Gamma; \Gamma'_I \Gamma'_{II} \Gamma) \\ & \times \langle i_I S_I \Gamma_I | U^{1\gamma} | i'_I S'_I \Gamma'_I \rangle \langle i_{II} S_{II} \Gamma_{II} | W^{1\gamma} | i'_{II} S'_{II} \Gamma'_{II} \rangle. \end{aligned} \quad (\text{A.5})$$

The reduced matrix elements appearing in (A.5) result from the previous iteration.

Appendix B

The systematic increasing cluster size requires an iterative procedure to compute the reduced matrix elements of the double irreducible tensors $U^{1\gamma}$ or $W^{1\gamma}$ (acting on the states with indices I and II , respectively) in the basis

$$\begin{aligned} |i S m \Gamma \mu\rangle & = \sum \alpha_{i_I S_I \Gamma_I; i_{II} S_{II} \Gamma_{II}}^{i S \Gamma} \begin{bmatrix} S_I & S_{II} & S \\ m_I & m_{II} & m \end{bmatrix} \begin{bmatrix} \Gamma_I & \Gamma_{II} & \Gamma \\ \mu_I & \mu_{II} & \mu \end{bmatrix} \\ & \times |i_I S_I m_I \Gamma_I \mu_I\rangle |i_{II} S_{II} m_{II} \Gamma_{II} \mu_{II}\rangle, \end{aligned} \quad (\text{B.1})$$

with an aid of the Wigner–Eckart theorem. On the other hand, one can use the basis of states (B.1) to obtain

$$\begin{aligned} \langle i S m \Gamma \mu | W_{q\nu}^{1\gamma} | i' S' m' \Gamma' \mu' \rangle & = \sum \alpha_{i_I S_I \Gamma_I; i_{II} S_{II} \Gamma_{II}}^{i S \Gamma} \alpha_{i'_I S'_I \Gamma'_I; i'_{II} S'_{II} \Gamma'_{II}}^{i' S' \Gamma'} \\ & \times \sum_{m_I m_{II} m'_I} \begin{bmatrix} S_I & S_{II} & S \\ m_I & m_{II} & m \end{bmatrix} \begin{bmatrix} S_I & S'_{II} & S' \\ m_I & m'_{II} & m' \end{bmatrix} (-1)^{S_{II}-m_{II}} \begin{pmatrix} S_{II} & 1 & S'_{II} \\ -m_{II} & q & m'_{II} \end{pmatrix} \\ & \times \sum_{\mu_I \mu_{II} \mu'_{II}} \begin{bmatrix} \gamma_I & \Gamma_{II} & \Gamma \\ \mu_I & \mu_{II} & \mu \end{bmatrix}^* \begin{bmatrix} \Gamma_I & \Gamma'_{II} & \Gamma \\ \mu_I & \mu'_{II} & \mu \end{bmatrix} \\ & \times \begin{bmatrix} \gamma & \Gamma'_{II} & \Gamma_{II} \\ \mu & \mu'_{II} & \mu_{II} \end{bmatrix}^* \langle i_{II} S_{II} \Gamma_{II} | W^{1\gamma} | i'_{II} S'_{II} \Gamma'_{II} \rangle. \end{aligned} \quad (\text{B.2})$$

The sum over m_I , m_{II} and m'_{II} is performed with the aid of the formula

$$\begin{aligned} & \sum_{\chi\psi\rho} (-1)^{p-\psi+q-\chi+r-\rho} \begin{pmatrix} p & a & q \\ \psi & \alpha & -\chi \end{pmatrix} \begin{pmatrix} q & b & r \\ \chi & \beta & -\rho \end{pmatrix} \begin{pmatrix} r & c & p \\ \rho & \gamma & -\psi \end{pmatrix} \\ & = \begin{pmatrix} a & b & c \\ -\alpha & -\beta & -\gamma \end{pmatrix} \begin{Bmatrix} a & b & c \\ r & p & q \end{Bmatrix}. \end{aligned} \quad (\text{B.3})$$

The sum of three Clebsch–Gordan coefficients of the lattice point group in turn can be transformed as follows:

$$\begin{aligned}
& \sum_{\mu_I \mu_{II} \mu'_{II}} \sum_{\bar{\nu} \bar{\mu}'} \begin{bmatrix} \Gamma_I & \Gamma_{II} & \Gamma \\ \mu_I & \mu_{II} & \mu \end{bmatrix}^* \begin{bmatrix} \Gamma_I & \Gamma'_{II} & \Gamma' \\ \mu_I & \mu'_{II} & \bar{\mu}' \end{bmatrix} \begin{bmatrix} \gamma & \Gamma'_{II} & \Gamma_{II} \\ \bar{\nu} & \mu'_{II} & \mu_{II} \end{bmatrix}^* \delta_{\bar{\nu} \bar{\nu}} \delta_{\mu' \bar{\mu}'} \\
&= \sum_{\mu_I \mu_{II} \mu'_{II}} \sum_{\bar{\nu} \bar{\mu}'} \begin{bmatrix} \Gamma_I & \Gamma_{II} & \Gamma \\ \mu_I & \mu_{II} & \mu \end{bmatrix}^* \begin{bmatrix} \Gamma_I & \Gamma'_{II} & \Gamma' \\ \mu_I & \mu'_{II} & \bar{\mu}' \end{bmatrix} \begin{bmatrix} \gamma & \Gamma'_{II} & \Gamma_{II} \\ \bar{\nu} & \mu'_{II} & \mu_{II} \end{bmatrix}^* \\
&\quad \times \sum_{\bar{\Gamma} \bar{\mu}} \begin{bmatrix} \gamma & \Gamma' & \bar{\Gamma} \\ \bar{\nu} & \mu' & \bar{\mu} \end{bmatrix}^* \begin{bmatrix} \gamma & \Gamma' & \bar{\Gamma} \\ \bar{\nu} & \bar{\mu}' & \bar{\mu} \end{bmatrix} \\
&= \sum_{\bar{\Gamma} \bar{\mu}} \begin{bmatrix} \gamma & \Gamma' & \bar{\Gamma} \\ \bar{\nu} & \mu' & \bar{\mu} \end{bmatrix}^* \sum_{\mu_I \mu_{II} \mu'_{II}} \sum_{\bar{\nu} \bar{\mu}'} \begin{bmatrix} \Gamma_I & \Gamma_{II} & \Gamma \\ \mu_I & \mu_{II} & \mu \end{bmatrix}^* \\
&\quad \times \begin{bmatrix} \Gamma_I & \Gamma'_{II} & \Gamma' \\ \mu_I & \mu'_{II} & \bar{\mu}' \end{bmatrix} \begin{bmatrix} \gamma & \Gamma'_{II} & \Gamma_{II} \\ \bar{\nu} & \mu'_{II} & \mu_{II} \end{bmatrix}^* \begin{bmatrix} \gamma & \Gamma' & \bar{\Gamma} \\ \bar{\nu} & \bar{\mu}' & \bar{\mu} \end{bmatrix}.
\end{aligned}$$

After permutation of the first and second columns in the third Clebsch–Gordan coefficient the sum over projections $\mu_I, \mu_{II}, \mu'_{II}, \bar{\nu}$, and $\bar{\mu}'$ is easily performed that gives immediately the 6Γ symbol [38]:

$$\sum_{\bar{\Gamma} \bar{\mu}} \begin{bmatrix} \gamma & \Gamma' & \bar{\Gamma} \\ \bar{\nu} & \bar{\mu}' & \bar{\mu} \end{bmatrix}^* \left\{ \begin{matrix} \gamma & \Gamma'_{II} & \Gamma_{II} \\ \Gamma_I & \Gamma & \Gamma' \end{matrix} \right\} \delta_{\Gamma \bar{\Gamma}} \delta_{\mu \bar{\mu}} \varepsilon(\Gamma_I \Gamma'_{II} \Gamma'), \quad (\text{B.4})$$

where we use the symmetry property of the Clebsch–Gordan coefficients

$$\begin{bmatrix} \Gamma_1 & \Gamma_2 & \Gamma \\ \mu_1 & \mu_2 & \mu \end{bmatrix} = \varepsilon(\Gamma_1 \Gamma_2 \Gamma) \begin{bmatrix} \Gamma_2 & \Gamma_1 & \Gamma \\ \mu_2 & \mu_1 & \mu \end{bmatrix},$$

and the sign $\varepsilon(\Gamma_1 \Gamma_2 \Gamma) = \pm 1$ depends on the point group.

The reduced matrix element can be computed using (B.2), (B.4) that yields the results

$$\begin{aligned}
\langle i S \Gamma \| W^{1\gamma} \| i' S' \Gamma' \rangle &= \sum \alpha_{i_i S_i \Gamma_i; i_{II} S_{II} \Gamma_{II}}^{i S \Gamma} \alpha_{i'_i S'_i \Gamma'_i; i'_{II} S'_{II} \Gamma'_{II}}^{i' S' \Gamma'} \\
&\quad \times (-1)^{1+S_i+S'_{II}+S} [S, S']^{1/2} \left\{ \begin{matrix} S & 1 & S' \\ S'_{II} & S_I & S_{II} \end{matrix} \right\} \\
&\quad \times \langle i_{II} S_{II} \Gamma_{II} \| W^{1\gamma} \| i'_{II} S'_{II} \Gamma'_{II} \rangle \left\{ \begin{matrix} \gamma & \Gamma'_{II} & \Gamma_{II} \\ \Gamma_I & \Gamma & \Gamma' \end{matrix} \right\} \varepsilon(\Gamma_I \Gamma'_{II} \Gamma'), \quad (\text{B.5})
\end{aligned}$$

and

$$\begin{aligned}
\langle i S \Gamma \| U^{1\gamma} \| i' S' \Gamma' \rangle &= \sum \alpha_{i_i S_i \Gamma_i; i_{II} S_{II} \Gamma_{II}}^{i S \Gamma} \alpha_{i'_i S'_i \Gamma'_i; i'_{II} S'_{II} \Gamma'_{II}}^{i' S' \Gamma'} \\
&\quad \times (-1)^{1+S_i+S_{II}+S'} [S, S']^{1/2} \left\{ \begin{matrix} S & 1 & S' \\ S'_I & S_{II} & S_I \end{matrix} \right\} \\
&\quad \times \langle i_I S_I \Gamma_I \| U^{1\gamma} \| i'_I S'_I \Gamma'_I \rangle \left\{ \begin{matrix} \gamma & \Gamma'_I & \Gamma_I \\ \Gamma_{II} & \Gamma & \Gamma' \end{matrix} \right\}, \quad (\text{B.6})
\end{aligned}$$

where $[S] \equiv (2S + 1)$.

Appendix C

Here, we give a detailed derivation of representation for the D_4 point group in the basis of four coupled connector spins

$$|S_1 S_2(S_{12}) S_3 S_4(S_{34}) S M\rangle = \sum \begin{bmatrix} S_1 & S_2 & S_{12} \\ m_1 & m_2 & m_{12} \end{bmatrix} \begin{bmatrix} S_3 & S_4 & S_{34} \\ m_3 & m_4 & m_{34} \end{bmatrix} \\ \times \begin{bmatrix} S_{12} & S_{34} & S \\ m_{12} & m_{34} & M \end{bmatrix} |S_1 m_1\rangle |S_2 m_2\rangle |S_3 m_3\rangle |S_4 m_4\rangle.$$

Consider first the $\pi/2$ rotation \hat{C}_4 about the z -axis:

$$\hat{C}_4 |S_1 S_2(S_{12}) S_3 S_4(S_{34}) S M\rangle = \sum_{m_1, m_2} \sum_{m_3, m_4} \sum_{m_{12}, m_{34}} \begin{bmatrix} S_1 & S_2 & S_{12} \\ m_1 & m_2 & m_{12} \end{bmatrix} \begin{bmatrix} S_3 & S_4 & S_{34} \\ m_3 & m_4 & m_{34} \end{bmatrix} \\ \times \begin{bmatrix} S_{12} & S_{34} & S \\ m_{12} & m_{34} & M \end{bmatrix} |S_2 m_2\rangle |S_3 m_3\rangle |S_4 m_4\rangle |S_1 m_1\rangle \\ = \sum_{m_1, m_2} \sum_{m_3, m_4} \sum_{m_{12}, m_{34}} \begin{bmatrix} S_1 & S_2 & S_{12} \\ m_1 & m_2 & m_{12} \end{bmatrix} \begin{bmatrix} S_3 & S_4 & S_{34} \\ m_3 & m_4 & m_{34} \end{bmatrix} \begin{bmatrix} S_{12} & S_{34} & S \\ m_{12} & m_{34} & M \end{bmatrix} \\ \times \sum_{\bar{m}_1, \bar{m}_2} \sum_{\bar{m}_3, \bar{m}_4} |S_2 \bar{m}_2\rangle |S_3 \bar{m}_3\rangle |S_4 \bar{m}_4\rangle |S_1 \bar{m}_1\rangle \delta_{\bar{m}_2 m_2} \delta_{\bar{m}_3 m_3} \delta_{\bar{m}_4 m_4} \delta_{\bar{m}_1 m_1}.$$

To proceed we use the orthogonality property for the Clebsch–Gordan coefficients:

$$\delta_{\bar{m}_2 m_2} \delta_{\bar{m}_3 m_3} \delta_{\bar{m}_4 m_4} \delta_{\bar{m}_1 m_1} = \sum_{S_{14}, m_{14}} \sum_{S_{23}, m_{23}} \begin{bmatrix} S_2 & S_3 & S_{23} \\ m_2 & m_3 & m_{23} \end{bmatrix} \begin{bmatrix} S_2 & S_3 & S_{23} \\ \bar{m}_2 & \bar{m}_3 & m_{23} \end{bmatrix} \\ \times \begin{bmatrix} S_4 & S_1 & S_{14} \\ m_4 & m_1 & m_{14} \end{bmatrix} \begin{bmatrix} S_4 & S_1 & S_{14} \\ \bar{m}_4 & \bar{m}_1 & m_{14} \end{bmatrix}.$$

Then

$$\hat{C}_4 |S_1 S_2(S_{12}) S_3 S_4(S_{34}) S M\rangle \\ = \sum_{m_1, m_2} \sum_{m_3, m_4} \sum_{m_{12}, m_{34}} \begin{bmatrix} S_1 & S_2 & S_{12} \\ m_1 & m_2 & m_{12} \end{bmatrix} \begin{bmatrix} S_3 & S_4 & S_{34} \\ m_3 & m_4 & m_{34} \end{bmatrix} \begin{bmatrix} S_{12} & S_{34} & S \\ m_{12} & m_{34} & M \end{bmatrix} \\ \times \sum_{\bar{m}_1, \bar{m}_2} \sum_{\bar{m}_3, \bar{m}_4} |S_2 \bar{m}_2\rangle |S_3 \bar{m}_3\rangle |S_4 \bar{m}_4\rangle |S_1 \bar{m}_1\rangle \sum_{S_{14}, m_{14}} \sum_{S_{23}, m_{23}} \begin{bmatrix} S_2 & S_3 & S_{23} \\ m_2 & m_3 & m_{23} \end{bmatrix} \\ \times \begin{bmatrix} S_2 & S_3 & S_{23} \\ \bar{m}_2 & \bar{m}_3 & m_{23} \end{bmatrix} \begin{bmatrix} S_4 & S_1 & S_{14} \\ m_4 & m_1 & m_{14} \end{bmatrix} \begin{bmatrix} S_4 & S_1 & S_{14} \\ \bar{m}_4 & \bar{m}_1 & m_{14} \end{bmatrix} \\ = \sum_{m_1, m_2} \sum_{m_3, m_4} \sum_{m_{12}, m_{34}} \begin{bmatrix} S_1 & S_2 & S_{12} \\ m_1 & m_2 & m_{12} \end{bmatrix} \begin{bmatrix} S_3 & S_4 & S_{34} \\ m_3 & m_4 & m_{34} \end{bmatrix} \begin{bmatrix} S_{12} & S_{34} & S \\ m_{12} & m_{34} & M \end{bmatrix} \\ \times \sum_{\bar{m}_1, \bar{m}_2} \sum_{\bar{m}_3, \bar{m}_4} |S_2 \bar{m}_2\rangle |S_3 \bar{m}_3\rangle |S_4 \bar{m}_4\rangle |S_1 \bar{m}_1\rangle \\ \times \sum_{S_{14}, m_{14}} \sum_{S_{23}, m_{23}} \sum_{\bar{m}_{14}, \bar{m}_{23}} \begin{bmatrix} S_2 & S_3 & S_{23} \\ m_2 & m_3 & m_{23} \end{bmatrix} \begin{bmatrix} S_2 & S_3 & S_{23} \\ \bar{m}_2 & \bar{m}_3 & \bar{m}_{23} \end{bmatrix} \\ \times \begin{bmatrix} S_4 & S_1 & S_{14} \\ m_4 & m_1 & m_{14} \end{bmatrix} \begin{bmatrix} S_4 & S_1 & S_{14} \\ \bar{m}_4 & \bar{m}_1 & \bar{m}_{14} \end{bmatrix} \delta_{\bar{m}_{23} m_{23}} \delta_{\bar{m}_{14} m_{14}} \\ = \sum_{m_1, m_2} \sum_{m_3, m_4} \sum_{m_{12}, m_{34}} \begin{bmatrix} S_1 & S_2 & S_{12} \\ m_1 & m_2 & m_{12} \end{bmatrix} \begin{bmatrix} S_3 & S_4 & S_{34} \\ m_3 & m_4 & m_{34} \end{bmatrix} \begin{bmatrix} S_{12} & S_{34} & S \\ m_{12} & m_{34} & M \end{bmatrix} \\ \times \sum_{\bar{m}_1, \bar{m}_2} \sum_{\bar{m}_3, \bar{m}_4} |S_2 \bar{m}_2\rangle |S_3 \bar{m}_3\rangle |S_4 \bar{m}_4\rangle |S_1 \bar{m}_1\rangle$$

$$\begin{aligned} & \times \sum_{S_{14}, m_{14}} \sum_{S_{23}, m_{23}} \sum_{\bar{m}_{14}, \bar{m}_{23}} \begin{bmatrix} S_2 & S_3 & S_{23} \\ m_2 & m_3 & m_{23} \end{bmatrix} \begin{bmatrix} S_2 & S_3 & S_{23} \\ \bar{m}_2 & \bar{m}_3 & \bar{m}_{23} \end{bmatrix} \\ & \times \begin{bmatrix} S_4 & S_1 & S_{14} \\ m_4 & m_1 & m_{14} \end{bmatrix} \begin{bmatrix} S_4 & S_1 & S_{14} \\ \bar{m}_4 & \bar{m}_1 & \bar{m}_{14} \end{bmatrix} \sum_{\bar{S}, \bar{M}} \begin{bmatrix} S_{23} & S_{14} & \bar{S} \\ m_{23} & m_{14} & \bar{M} \end{bmatrix} \begin{bmatrix} S_{23} & S_{14} & \bar{S} \\ \bar{m}_{23} & \bar{m}_{14} & \bar{M} \end{bmatrix} \end{aligned}$$

A full contraction of six Clebsch–Gordan coefficients yields the $9j$ symbol:

$$\begin{aligned} & \sum_{m_1, m_2} \sum_{m_3, m_4} \sum_{m_{12}, m_{34}} \begin{bmatrix} S_1 & S_2 & S_{12} \\ m_1 & m_2 & m_{12} \end{bmatrix} \begin{bmatrix} S_3 & S_4 & S_{34} \\ m_3 & m_4 & m_{34} \end{bmatrix} \begin{bmatrix} S_{12} & S_{34} & S \\ m_{12} & m_{34} & M \end{bmatrix} \\ & \times \begin{bmatrix} S_4 & S_1 & S_{14} \\ m_4 & m_1 & m_{14} \end{bmatrix} \begin{bmatrix} S_2 & S_3 & S_{23} \\ m_2 & m_3 & m_{23} \end{bmatrix} \begin{bmatrix} S_{23} & S_{14} & \bar{S} \\ m_{23} & m_{14} & \bar{M} \end{bmatrix} \\ & = (-1)^{S_1+S_4-S_{14}} (-1)^{S_3+S_4-S_{34}} (-1)^{S_{23}+S_{14}-\bar{S}} \\ & \times \sum_{m_1, m_2} \sum_{m_3, m_4} \sum_{m_{12}, m_{34}} \begin{bmatrix} S_1 & S_2 & S_{12} \\ m_1 & m_2 & m_{12} \end{bmatrix} \begin{bmatrix} S_3 & S_4 & S_{34} \\ m_3 & m_4 & m_{34} \end{bmatrix} \begin{bmatrix} S_{12} & S_{34} & S \\ m_{12} & m_{34} & M \end{bmatrix} \\ & \times \begin{bmatrix} S_1 & S_4 & S_{14} \\ m_1 & m_4 & m_{14} \end{bmatrix} \begin{bmatrix} S_2 & S_3 & S_{23} \\ m_2 & m_3 & m_{23} \end{bmatrix} \begin{bmatrix} S_{14} & S_{23} & \bar{S} \\ m_{14} & m_{23} & \bar{M} \end{bmatrix} \\ & = (-1)^{S_1+S_4-S_{14}} (-1)^{S_3+S_4-S_{34}} (-1)^{S_{23}+S_{14}-\bar{S}} \\ & \times [(2S_{12} + 1)(2S_{14} + 1)(2S_{23} + 1)(2S_{34} + 1)]^{1/2} \begin{Bmatrix} S_1 & S_2 & S_{12} \\ S_4 & S_3 & S_{34} \\ S_{14} & S_{23} & S \end{Bmatrix} \delta_{S\bar{S}} \delta_{M\bar{M}}, \end{aligned}$$

where we use the symmetry relation for the Clebsch–Gordan coefficients

$$\begin{bmatrix} S_1 & S_2 & S_{12} \\ m_1 & m_2 & m_{12} \end{bmatrix} = (-1)^{S_1+S_2-S_{12}} \begin{bmatrix} S_2 & S_1 & S_{12} \\ m_2 & m_1 & m_{12} \end{bmatrix}.$$

By introducing the state with the recoupled four spins

$$\begin{aligned} |S_2 S_3 (S_{23}) S_4 S_1 (S_{14}) \bar{S} \bar{M}\rangle & = \sum_{\bar{m}_1, \bar{m}_2} \sum_{\bar{m}_3, \bar{m}_4} \sum_{\bar{m}_{14}, \bar{m}_{23}} \begin{bmatrix} S_2 & S_3 & S_{23} \\ \bar{m}_2 & \bar{m}_3 & \bar{m}_{23} \end{bmatrix} \begin{bmatrix} S_4 & S_1 & S_{14} \\ \bar{m}_4 & \bar{m}_1 & \bar{m}_{14} \end{bmatrix} \\ & \times \begin{bmatrix} S_{23} & S_{14} & \bar{S} \\ \bar{m}_{23} & \bar{m}_{14} & \bar{M} \end{bmatrix} |S_2 \bar{m}_2\rangle |S_3 \bar{m}_3\rangle |S_4 \bar{m}_4\rangle |S_1 \bar{m}_1\rangle, \end{aligned}$$

we get finally

$$\begin{aligned} \hat{C}_4 |S_1 S_2 (S_{12}) S_3 S_4 (S_{34}) S M\rangle & = \sum_{S_{14}, S_{23}} (-1)^{S_1+S_4-S_{14}} (-1)^{S_3+S_4-S_{34}} (-1)^{S_{23}+S_{14}-\bar{S}} \\ & \times [(2S_{12} + 1)(2S_{14} + 1)(2S_{23} + 1)(2S_{34} + 1)]^{1/2} \\ & \times \begin{Bmatrix} S_1 & S_2 & S_{12} \\ S_4 & S_3 & S_{34} \\ S_{14} & S_{23} & S \end{Bmatrix} |S_2 S_3 (S_{23}) S_4 S_1 (S_{14}) S M\rangle, \end{aligned}$$

that is transformation from a coupling scheme to another. Then an action of the operator \hat{C}_4 is defined by a linear transformation of the basis $\kappa' = \{S'_1 S'_2 (S'_{12}) S'_3 S'_4 (S'_{34})\}$

$$\hat{C}_4 |\kappa\rangle = \sum_{\kappa'} D_{\kappa'\kappa}^{(S)}(\hat{C}_4) |\kappa'\rangle,$$

where the matrix $D_{\kappa'\kappa}^{(S)}(\hat{C}_4)$ is determined by the expression

$$D_{\kappa'\kappa}^{(S)}(\hat{C}_4) = \delta_{S_2 S_1'} \delta_{S_3 S_2'} \delta_{S_4 S_3'} \delta_{S_1 S_4'} \delta_{S_{23} S_{12}'} \delta_{S_{14} S_{34}'} [(2S_{12} + 1)(2S_{14} + 1)(2S_{23} + 1)(2S_{34} + 1)]^{1/2} \\ \times (-1)^{S_1 + S_4 - S_{14}} (-1)^{S_3 + S_4 - S_{34}} (-1)^{S_{23} + S_{14} - \bar{S}} \begin{Bmatrix} S_1 & S_2 & S_{12} \\ S_4 & S_3 & S_{34} \\ S_{14} & S_{23} & S \end{Bmatrix}.$$

We can handle analogously another symmetry operations in the D_4 group.

Appendix D

The standard method for constructing an irreducible basis is to use the projection operator

$$P_\mu^\Gamma = \frac{[\Gamma]}{[G]} \sum_{\hat{g} \in G} D_{\mu\mu}^{(\Gamma)}(\hat{g}) \hat{g}, \quad (\text{D.1})$$

and the shift operator

$$P_{\mu\nu}^\Gamma = \frac{[\Gamma]}{[G]} \sum_{\hat{g} \in G} D_{\mu\nu}^{(\Gamma)}(\hat{g}) \hat{g}, \quad (\text{D.2})$$

where $[G]$ is the order of the group G , $[\Gamma]$ is the dimension of the irreducible representation Γ and $D_{\mu\nu}^{(\Gamma)}(\hat{g})$ are the irreducible matrix elements, μ or ν is an index enumerating the basis. Supposing that ψ is one of the reducible basis vectors of G , an irreducible basis might be obtained by applying

$$P_\mu^\Gamma \psi = (\psi_\mu^\Gamma \cdot \psi) \psi_\mu^\Gamma. \quad (\text{D.3})$$

If $\{\psi_\nu^\Gamma\}$ is the basis for the irrep Γ then

$$P_{\mu\nu}^\Gamma \psi_\nu^\Gamma = \psi_\mu^\Gamma. \quad (\text{D.4})$$

Let us construct irreducible tensors $U_{q\mu}^{1\Gamma}$ from the operators $\{S_a, S_b, S_c, S_d\}$ forming the nearest environment of the central site. The transformations of one of the given spins under the elements of the group D_4 are

$$E S_a = S_a, \quad C_4 S_a = S_d, \quad C_4^2 S_a = S_c, \quad C_4^3 S_a = S_b, \\ C_2^x S_a = S_b, \quad C_2^y S_a = S_d, \quad C_v' S_a = S_a, \quad C_v'' S_a = S_c,$$

that together with (D.1) give immediately the irreducible tensors $U_{q\mu}^{1\Gamma}$ of the one-dimensional representations:

$$U_{q1}^{1A_1} = N_{A_1} (S_a + S_b + S_c + S_d), \quad U_{q1}^{1A_2} = 0, \\ U_{q1}^{1B_1} = 0, \quad U_{q1}^{1B_2} = N_{B_2} (S_a - S_b + S_c - S_d).$$

To find the irreducible basis for the two-dimensional representation E , we construct the projection operator P_1^E and then apply it to the spin S_a that yields

$$U_{q1}^{1E} = N_E (S_a + S_b - S_c - S_d).$$

Using the shift operator (D.2) and acting according to the rule (D.4) we obtain the second irrep basis vector

$$U_{q2}^{1E} = N_E (S_a - S_b - S_c + S_d).$$

We choose the coefficients N_Γ ($\Gamma = A_1, E$) so that the Hamiltonian written through the irreducible tensors coincides with the initial spin operator form.

It is not always possible to construct all irreducible bases from one chain. The theory says that one has to choose another starting function. In a computer realization, therefore, we build all chains generated by all vectors of a reducible basis:

$$\hat{g}\psi_i = \sum_j D_{ji}(\hat{g})\psi_j, \quad (\text{D.5})$$

and form the matrix \hat{X}^Γ from the chains

$$\hat{X}_{ji}^\Gamma = N_\Gamma \sum_g D_{\mu\mu}^\Gamma(\hat{g})D_{ji}(\hat{g}). \quad (\text{D.6})$$

The rank of this matrix

$$C_\Gamma = \frac{1}{[\Gamma]} \sum_g \chi(g)\chi^\Gamma(g)$$

determines a number of linear independent columns, where the character $\chi(\hat{g}) = \sum_i D_{ii}(\hat{g})$. After these columns are established, orthogonalized and normalized with the help of the Schmidt–Gram procedure we get the first C_Γ columns of the transformation matrix $\hat{T}_{i,\Gamma\mu}$. By running over all irreducible representations and repeating the basic steps in the approach we obtain the square matrix of corresponding similarity transformation onto the symmetry adapted basis:

$$\psi_\mu^\Gamma = \sum_i \hat{T}_{i,\Gamma\mu} \psi_i.$$

As an example we calculate $\hat{T}_{S_{12}S_{34},\Gamma\mu}^{(1)}$ for the nearest-neighbour environment of the central site. The characters of three-dimensional representation can be read off from the 3×3 matrices $D^{(1)}$ given in section 3. We thus obtain

$$\begin{array}{cccccc} & E & C_4, C_4^3 & C_4^2 & C_2^x, C_2^y & \sigma'_v, \sigma''_v \\ \chi & 3 & -1 & -1 & -1 & 1 \end{array}$$

whence we conclude $D^{(1)} = D^{(1B_1)} \oplus D^{(1E)}$. A direct calculation of \hat{X}^Γ matrices from equation (D.6) yields

$$\hat{X}_{ji}^{B_1} = \frac{1}{8} \sum_g \chi^{B_1}(\hat{g})D_{ji}^{(1)}(\hat{g}) = \begin{pmatrix} \frac{1}{2} & \frac{1}{2} & 0 \\ \frac{1}{2} & \frac{1}{2} & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

$$\hat{X}_{11}^E = \frac{2}{8} \sum_g D_{11}^E(\hat{g})D_{ji}^{(1)}(\hat{g}) = \begin{pmatrix} \frac{1}{4} & -\frac{1}{4} & \frac{\sqrt{2}}{4} \\ -\frac{1}{4} & \frac{1}{4} & -\frac{\sqrt{2}}{4} \\ \frac{\sqrt{2}}{4} & -\frac{\sqrt{2}}{4} & \frac{1}{2} \end{pmatrix},$$

and

$$\hat{X}_{21}^E = \frac{2}{8} \sum_g D_{21}^E(\hat{g})D_{ji}^{(1)}(\hat{g}) = \begin{pmatrix} \frac{1}{4} & -\frac{1}{4} & \frac{\sqrt{2}}{4} \\ -\frac{1}{4} & \frac{1}{4} & -\frac{\sqrt{2}}{4} \\ -\frac{\sqrt{2}}{4} & \frac{\sqrt{2}}{4} & -\frac{1}{2} \end{pmatrix}.$$

By noting that ranks of the matrices equal to unity, we find finally via the Schmidt–Gram procedure the transformation matrix $\hat{T}_{S_{\alpha\beta}S_{\gamma\eta};\Gamma\mu}^{(1)}$:

$$\begin{array}{l} |01; 1M\rangle \\ |10; 1M\rangle \\ |11; 1M\rangle \end{array} \begin{array}{ccc} |1M; B_1\rangle & |1M; E1\rangle & |1M; E2\rangle \\ \frac{1}{\sqrt{2}} & \frac{1}{2} & \frac{1}{2} \\ \frac{1}{\sqrt{2}} & -\frac{1}{2} & -\frac{1}{2} \\ 0 & \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} \end{array} .$$

Appendix E

For the reader convenience we give the character table of the group D_4

D_4	E	C_4, C_4^{-1}	C_4^2	C_2^x, C_2^y	C'_v, C''_v
A_1	1	1	1	1	1
A_2	1	1	1	-1	-1
B_1	1	-1	1	1	-1
B_2	1	-1	1	-1	1
E	2	0	-2	0	0

and the matrices of double irreducible representation taken in the basis xy (see [32], for example)

$$\begin{array}{l} D^{(E)}(E) = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \\ D^{(E)}(C_4^3) = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}, \\ D^{(E)}(C'_v) = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \end{array} \quad \begin{array}{l} D^{(E)}(C_4) = \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}, \\ D^{(E)}(C_2^x) = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \\ D^{(E)}(C''_v) = \begin{pmatrix} 0 & -1 \\ -1 & 0 \end{pmatrix}. \end{array} \quad \begin{array}{l} D^{(E)}(C_4^2) = \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix}, \\ D^{(E)}(C_2^y) = \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}, \end{array}$$

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