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Variational determination of the two-particle reduced density matrix within the doubly occupied configuration interaction space: exploiting translational and reflection invariance

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Abstract. This work incorporates translational and reflection symmetry reductions to the variational determination of the two-particle reduced density matrix (2-RDM) corresponding to the ground state of N-particle systems, within the doubly occupied configuration interaction (DOCI) space. By exploiting these symmetries within this lower-bound variational methodology it is possible to treat larger systems than those previously studied. The 2-RDM matrix elements are calculated by imposing up to four-particle N-representability constraint conditions using standard semidefinite programing algorithms. The method is applied to the one- and two-dimensional XXZ spin 1/2 model of quantum magnetism. Several observables including the energy and the spin–spin correlation functions are obtained to assess the physical content of the variationally determined 2-RDM. Comparison with quantum-Monte Carlo and matrix product state simulations shows that in most cases only requiring up to three-particle positivity conditions is enough to correctly describe the ground-state properties of these one- and two-dimensional models.

Keywords: spin chains, ladders and planes, correlation functions

Contents

1. Introduction

The exponential growth of the dimension of the Hilbert space with the number of composing particles constitutes one of the main issues in quantum many-body problems,

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including those present in condensed matter, nuclear physics, and quantum chemistry [1]. Although a complete diagonalization of the Hamiltonian describing such systems in the many-particle space provides the exact answer, it does so at a prohibitively expensive computational cost. Therefore, research efforts have been focussed on the development of approximate methods capturing the most relevant physical content of the many-body wavefunction at a feasible computational cost, i.e. with a polynomial increase with system size. Most of such methods rely onto approximations that improve over a reference many-body state or onto variational treatments on the energy. In the former case, a common approach is to start from a reference state obtained from a mean-field approximation and add perturbative corrections [2] or excitations with increasing complexity [3, 4]. However, these methods fail in the strong correlation regimes where multi-reference approximations are needed. New variational methods overcoming this issue were developed over the last few decades. For instance, variational algorithms like tensor-network-state approaches [5–8], variational Monte Carlo methods [9–12], or stochastic techniques [13–16] can be made, in principle, as accurate as the exact diagonalizations. For instance, in one dimension, the well-known matrix product state (MPS) representation of a system can accurately describe ground states at the computational cost of only a power law with the system size [5, 17, 18]. However, in practice, even though the MPS is one of the best understood tensor networks for which many efficient algorithms have been developed, for higher dimensional systems the cost of the method is shown to rapidly increase exponentially [19]. Therefore, other methods based on the concept of tensor networks are continuously being developed [20–22]. On the other hand, quantum Monte Carlo (QMC) simulations [23, 24] are not restricted to one dimension, albeit only suitable for systems not suffering from the infamous sign problem [25] and being efficient to calculate only certain magnitudes.

A very different approach to address the exponential wall problem that is applicable to any correlation regime relies on using the second-order reduced density matrix (2-RDM) [26, 27], without mediation of the wavefunction. As the energy of any pairwise-interacting system can be written as an exact but simple linear function of the 2-RDM, it can be used to variationally optimize this matrix at polynomial cost [28]. However, this optimization should be constrained to the class of 2-RDMs that can be derived from a wavefunction (or an ensemble of wavefunctions), the so-called N-representable 2-RDMs [29, 30]. Since one may use an incomplete set of necessary but not (in general) sufficient constraints on the 2-RDM, the optimization finds a lower bound to the exact ground-state energy and an approximation to the exact ground-state 2-RDM. Such an approach, known as the variational second-order reduced density matrix (v2RDM) method, has been applied with different degrees of success in quantum-chemistry [31–34], nuclear-physics [35, 36], and condensed-matter problems [37–39]. Similar procedures to that of the v2RDM methodology have been recently proposed which consist of, e.g. substituting the density-matrix description of the system for approximate finite-range correlation matrices of tunable order to limit the amount of information kept [40] or embedding highly accurate local theories to solve large-scale strongly correlated quantum many-body theories [41].

In the last few years, the computational efficiency of the v2RDM method has been substantially improved for systems whose states can be described in the Hilbert subspace

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of paired single-particle states. This lies at the heart of the so-called doubly-occupied configuration interaction (DOCI) method used in quantum chemistry to reduce the dimension of the configuration interaction Hilbert space. The assumptions in DOCI drastically simplify the structure of the 2-RDM [42, 43] and reduce the scaling of the v2RDM method [44–46] for molecular systems, while still offering an excellent method for many systems as demonstrated through the years [47–50]. A related concept, the seniority quantum number, counts the number of particles not coupled in pair of total angular momentum 0. It has been initially proposed by Racah [51] and extensively developed in nuclear physics [52]. Remarkably, the assumptions of DOCI and seniority are directly related to the pseudo-SU(2) pair algebra. Very recently, this combined approach of the v2RDM restricted to the DOCI space has been used to tackle an important class of Hamiltonians [53, 54], the pairing Hamiltonians, where the fundamental physics lies in the specific form of the paired states. The quantum integrable and exactly solvable Richardson–Gaudin models [55–58] describe pairing Hamiltonians with a large amount of free parameters in the single particle-energies and the interactions [59]. They were recently utilized to benchmark the v2RDM-DOCI methodology at various levels of N-representability conditions and they were shown to provide excellent results. Later on, the research extended to treat one-dimensional Hamiltonians presenting quantum magnetism that demonstrated the need to develop higher positivity constraints near the emergence of long-range antiferromagnetic ordering [60]. The results were very promising but limited to one-dimensional systems of small-to-moderate sizes. In this work, we include novel translational and reflection symmetry reductions onto the v2RDM-DOCI methodology that greatly overcome the previous limitations in size.

The manuscript is organized as follows. In section 2.1 we review the theoretical aspects of the variational methodology in terms of the p-particle RDMs of hard-core bosons. Section 2.2 provides a detailed description of the symmetry reductions applied to one- and two-dimensional systems. In section 3 we present the application to the Heisenberg XXZ model, with a discussion of the computational details (section 3.1) and the numerical results for the ground-state energies and spin–spin correlation functions along a comparison with other methodologies (section 3.2). Finally, in section 4 we present a summary of the main achievements and concluding remarks.

2. Methods

2.1. Theoretical background

Recently, the v2RDM approach has been applied within the restricted DOCI space, strongly reducing its computational scaling. The method is adequate for studying Hamiltonians with interactions in the DOCI space, i.e. interactions that do not change the number of paired fermions. For this class of Hamiltonians, the seniority number is also an exact quantum number, as unpaired particles do not interact with the rest of the system since seniority-zero Hamiltonians do not allow for pair breaking.

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The most general Hamiltonian within the seniority-zero subspace can be written as

$$H = \sum_i \epsilon_i n_i + \sum_{i \neq j} w_{ij} n_i n_j + \sum_{ij} v_{ij} b_i^\dagger b_j \tag{1}$$

where ϵ_i are the energies of the K single-particle levels, and w_{ij} and v_{ij} stand for the monopole and pairing interactions, respectively. The operators $b_i^\dagger$ and $(2n_i - 1)/2$ are the generators of the SU(2) pair algebra, satisfying the following hard-core boson relations

$$\left[b_i, b_j^\dagger\right] = \delta_{ij}\left(1 - 2n_i\right), \quad \left(b_i^\dagger\right)^2 = 0. \tag{2}$$

These operators can be related to the fermion creation and annihilation operators from a fermion pairing scheme involving two particles with either opposite spins $(i\uparrow, i\downarrow)$, momenta $(i, -i)$, or in general any classification of conjugate quantum numbers in doubly degenerate single-particle levels.

The ground-state energy of Hamiltonian (1) can then be cast as [42–44]

$$E_{\mathrm{GS}} = \sum_{ij} J_{ij}\Pi_{ij} + \sum_{i \neq j} w_{ij} D_{ij} \tag{3}$$

where $J_{ij} = \delta_{ij}\epsilon_i + v_{ij}$, and the Π_{ij} and D_{ij} matrices given by

$$\begin{aligned} &\Pi_{ij} = \langle\psi|b_i^\dagger b_j|\psi\rangle, \qquad D_{ij} = \langle\psi|n_i n_j|\psi\rangle \quad \forall\, i \neq j \\ &D_{ii} = \Pi_{ii} = \rho_i = \langle\psi|n_i|\psi\rangle \end{aligned} \tag{4}$$

define the seniority blocks of the 2-RDM, which are Hermitian and fulfil the normalization and contraction relations

$$\sum_i \Pi_{ii} = \sum_i D_{ii} = M, \tag{5}$$

$$\sum_j D_{ij} = M\Pi_{ii}. \tag{6}$$

where M is the number of hard-core bosons. In principle, equation (3) indicates that the ground-state energy of the system may be computed by direct variation of the 2-RDM. However, this is not the case, because not every computed 2-RDM derives from the integration of an N-particle wave function [30, 61]. This difficulty leads to the well-known N-representability problem which aims to define a set of necessary and sufficient conditions, ensuring a reduced density matrix stems from an N-particle physical system. In general, the p particle reduced density matrix (p-RDM) must be Hermitian, properly normalized and related by contraction mappings such as equations (5) and (6) to other order p-RDMs. Additionally, a hierarchy of necessary, albeit not sufficient, set of constraints on the p-RDM constitutes the p-positivity N-representability conditions (p-POS) [33, 62]. These conditions, whose application leads to energies closer to the exact one according to the sequence

$$E_{p-\mathrm{POS}} \leqslant E_{q-\mathrm{POS}} \leqslant E_{\mathrm{GS}} \quad (1 \leqslant p < q \leqslant N), \tag{7}$$

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derive from the positive semidefinite property of a class of Hamiltonians of the form

$$H = B^\dagger B \tag{8}$$

where $B^\dagger$ is a p-particle operator. The expectation value of such Hamiltonians must be non-negative and hence their matrix representations must be positive semidefinite. Therefore, different forms of $B^\dagger$ lead to different p-positivity conditions. For instance, in the two-particle space, these yield the well-known 2-$\mathcal{P}$, 2-$\mathcal{Q}$ and 2-$\mathcal{G}$ two-positivity conditions (2-POS) [30, 63] which require the positive semidefiniteness of the two-particle, two-hole, and particle–hole representations of the 2-RDM. Within the DOCI subspace, these conditions can be formulated in terms of the seniority blocks of the 2-RDM as follows [42–46, 53, 64]: while the 2-$\mathcal{P}$ condition is given by

$$\Pi_{ij} \succeq 0, \tag{9}$$

$$D_{ij} \geqslant 0, \quad \forall\ i < j, \tag{10}$$

where the symbol $\succeq$ means that the corresponding matrix is semidefinite positive, the 2-$\mathcal{Q}$ condition is

$$\Pi_{ij} + \delta_{ij}(1 - 2\rho_i) \succeq 0, \tag{11}$$

$$D_{ij} + 1 - \rho_i - \rho_j \geqslant 0, \quad \forall\ i < j, \tag{12}$$

and the 2-$\mathcal{G}$ condition is given by

$$D_{ij} \succeq 0 \tag{13}$$

$$\begin{pmatrix} \rho_a - D_{ab} & -\Pi_{ab} \\ -\Pi_{ba} & \rho_b - D_{ab} \end{pmatrix} \succeq 0, \quad \forall\ a < b. \tag{14}$$

Each of these conditions possesses $O(1)$ blocks of dimension $O(K \times K)$ and $O(K^2)$ blocks of dimension $O(1 \times 1)$.

Similarly to the 2-POS conditions, the three- (3-POS) and four-positivity (4-POS) conditions in the seniority-zero subspace can be expressed in terms of the seniority blocks of the 3- and 4-RDMs, defined as

$$D_{ijk} = \langle\psi|n_i n_j n_k|\psi\rangle, \quad \forall\ i \neq j \neq k \tag{15}$$

$$\Pi^i_{jk} = \langle\psi|b_j^\dagger n_i b_k|\psi\rangle = \Pi^i_{kj} = \langle\psi|b_k^\dagger n_i b_j|\psi\rangle, \quad \forall\ i \neq j, k \tag{16}$$

and

$$D_{ijkl} = \langle\psi|n_i n_j n_k n_l|\psi\rangle, \quad \forall\ i \neq j \neq k \neq l \tag{17}$$

$$\Pi^{ij}_{kl} = \langle\psi|b_k^\dagger n_i n_j b_l|\psi\rangle, \quad \forall\ i \neq j \neq k \neq l \tag{18}$$

$$\Pi_{ijkl} = \langle\psi|b_i^\dagger b_j^\dagger b_l b_k|\psi\rangle, \quad \forall\ i \neq j \neq k \neq l. \tag{19}$$

respectively. The complete set of constraints on these blocks recently reported in [54, 60], are presented in appendix A.

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The variational optimization of the 2-RDM in the DOCI space may be formulated as a semidefinite programing (SDP) problem in which the energy, being a linear function of the 2-RDM, is minimized over the intersection of a linear affine space (3) and the convex cone of block-diagonal positive semidefinite matrices corresponding to the above sets of N-representability conditions. It should be noted that, as the commutation relations of the hard-core boson operators ensure certain symmetries among the different seniority sectors of the p-RDMs, e.g. hermiticity of Π_{ij} and D_{ij}, etc, we have explicitly restricted the p-POS conditions to terms involving unique parts of the corresponding p-RDMs and avoid any repetition. In practice, this amounts to choosing specific orderings of the indexes appearing in the different seniority blocks. In the next section, we will discuss additional simplifications of this methodology for translation and reflection invariant systems.

2.2. Reductions for translation and reflection invariant systems

The variational procedure can be significantly simplified when the Hamiltonian (1) is translational and reflection invariant. In this case, the single-particle level index may be identified with the position of a site on a lattice and the ground state of H is guaranteed to either be translationally invariant or degenerate. In the latter case, one can choose a superposition of the degenerate ground-state wavefunctions that would itself be translationally invariant. We can, therefore, impose translational invariance on the p-RDMs and thus reduce the number of variational parameters to be determined. Similar considerations follow from the reflection symmetry. The variational calculation of the 2-RDM for periodic molecular systems has been previously tackled by using irreducible representations of the translational operator for low order N-representability conditions [65], and very recently also in presence of strong correlations [66]. In this work, we directly apply the symmetry reductions onto the full 2-POS, 3-POS, and 4-POS N-representability conditions in coordinate space.

In this section, the indices of the p-RDMs will be explicitly separated by commas to highlight the mathematical operation performed on each index. The case of one-dimensional lattices is the simplest one. Let us consider the two seniority blocks of the 2-RDM. On the one hand, there is only one reduced variable $\rho_i = \rho_0 \,\forall\, i$ that corresponds to the single-site occupation of the translationally invariant system. On the other hand, the variable reductions are straightforward in the $D_{i,j}$ and $\Pi_{i,j}$ matrices as, assuming $i < j$, the translational symmetry entails

$$D_{i,j} = D_{0,j-i} = D_{i-j,0}, \tag{20}$$

$$\Pi_{i,j} = \Pi_{0,j-i} = \Pi_{i-j,0}, \tag{21}$$

where we have numbered the L sites from 0 to $L-1$ and we understand that a negative $-q$ index means a site at $L-q$. This reduces the number of variational parameters in the order of $O(L)$ times. The reflection symmetry does not add a further reduction in these blocks because it is equivalent to an index exchange followed by a translation.

The two seniority blocks of the 3-RDM, $D_{i,j,k}$ and $\Pi^i_{j,k}$, can also be reduced using the translational invariance. However, in this case $D_{i,j,k}$ and $\Pi^i_{j,k}$ have to be treated differently as the i index in $\Pi^i_{j,k}$, corresponding to a number operator index n_i (cf

equation (16)), is not equivalent to the j and k indices, corresponding to hard-core boson creation ($b_j^\dagger$) and annihilation (b_k) operator indices. On the one hand, choosing the ordering $i < j < k$ in all the instances of $D_{i,j,k}$, the reflection and translational invariance warrants that

$$D_{i,j,k} = D_{i-n,j-n,k-n} = D_{n-i,n-j,n-k}, \quad \forall\ 0 \leqslant n < L \tag{22}$$

where the last equality involves variables obtained by applying the reflection symmetry around site 0. This amounts to a reduction factor of about $2L$ in the variational parameters entering $D_{i,j,k}$. For a faster computation, it is enough to find a representative variable that has one index equal to 0

$$\begin{aligned} D_{i,j,k} &= D_{0,j-i,k-i} = D_{i-j,0,k-j} = D_{i-k,j-k,0} \\ &= D_{0,i-j,i-k} = D_{j-i,0,j-k} = D_{k-i,k-j,0}. \end{aligned} \tag{23}$$

Furthermore, a similar procedure on the $D_{i,j,k,l}$ yields a factor of $2L$.

On the other hand, choosing the ordering $j < k$ in all the instances of $\Pi^i_{j,k}$, the reflection and translational invariance on equation (16) implies

$$\Pi^i_{j,k} = \Pi^{i-n}_{j-n,k-n} = \Pi^{n-i}_{n-j,n-k}, \quad \forall\ 0 \leqslant n < L \tag{24}$$

and the variable reduction amounts to a factor of $2L$. To find a representative variable it is enough to search in the variables

$$\Pi^i_{j,k} = \Pi^0_{j-i,k-i} = \Pi^0_{i-j,i-k}. \tag{25}$$

Analogously, for the four-index matrices $\Pi^{i,j}_{k,l}$ and $\Pi_{i,j,k,l}$, equations (18) and (19), we obtain a reduction factor of $2L$.

In two-dimensional systems, the site indices correspond to vectors in a lattice. For a square lattice of side L, $i = (i_x, i_y)$ with $0 \leqslant i_x < L$ and $0 \leqslant i_y < L$. In this case the translational invariance corresponds to a torus topology, i.e. periodic boundary conditions along both x and y directions. Similarly to the one-dimensional case in (22), for the variables in the $D_{i,j,k}$ matrix this implies

$$\begin{aligned} D_{i,j,k} &= D_{i-n,j-n,k-n} \\ &= D_{-(i-n),-(j-n),-(k-n)} \\ &= D_{\sigma_x(i-n),\sigma_x(j-n),\sigma_x(k-n)} \\ &= D_{\sigma_y(i-n),\sigma_y(j-n),\sigma_y(k-n)} \\ &\qquad \forall\ n = (n_x, n_y), \quad 0 \leqslant n_x, n_y < L \end{aligned} \tag{26}$$

where now the subtraction operation means a vectorial subtraction. The first line in (26) involves only the translation symmetry, the second one the effect of the inversion around site $0 \equiv (0, 0)$. In the third and fourth lines the reflection symmetries are taken into account separately on each direction, using the reflection matrices $\sigma_x = \mathrm{diag}(-1, 1)$ and $\sigma_y = \mathrm{diag}(1, -1)$. This leads to a stronger variable reduction with a factor of $4L^2$

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instead of $2L$ as in the one-dimensional case. As before, it is more efficient to search the representative variable by choosing that one of the indexes is at site 0 and in that case

$$\begin{aligned} D_{i,j,k} &= D_{0,j-i,k-i} = D_{i-j,0,k-j} = D_{i-k,j-k,0} \\ &= D_{0,-(j-i),-(k-i)} = D_{-(i-j),0,-(k-j)} = D_{-(i-k),-(j-k),0} \\ &= D_{0,\sigma_x(j-i),\sigma_x(k-i)} = D_{\sigma_x(i-j),0,\sigma_x(k-j)} = D_{\sigma_x(i-k),\sigma_x(j-k),0} \\ &= D_{0,\sigma_y(j-i),\sigma_y(k-i)} = D_{\sigma_y(i-j),0,\sigma_y(k-j)} = D_{\sigma_y(i-k),\sigma_y(j-k),0}. \end{aligned} \tag{27}$$

Analogously, for the variables in the $\Pi^i_{j,k}$ matrix this implies that, as in equation (24),

$$\begin{aligned} \Pi^i_{j,k} &= \Pi^{i-n}_{j-n,k-n} \\ &= \Pi^{-(i-n)}_{-(j-n),-(k-n)} \\ &= \Pi^{\sigma_x(i-n)}_{\sigma_x(j-n),\sigma_x(k-n)} \\ &= \Pi^{\sigma_y(i-n)}_{\sigma_y(j-n),\sigma_y(k-n)} \\ &\qquad \forall\ n = (n_x, n_y), \quad 0 \leqslant n_x, n_y < L. \end{aligned} \tag{28}$$

This also leads to variable reductions with a factor of $4L^2$ instead of $2L$ as in the one-dimensional case. It is thus enough to select the translation that moves site i to 0 to obtain

$$\begin{aligned} \Pi^i_{j,k} &= \Pi^0_{j-i,k-i} \\ &= \Pi^0_{-(j-i),-(k-i)} \\ &= \Pi^0_{\sigma_x(j-i),\sigma_x(k-i)} \\ &= \Pi^0_{\sigma_y(j-i),\sigma_y(k-i)}. \end{aligned} \tag{29}$$

The same generalized approach is useful for the other matrices and this causes a reduction of the free variables of L^2 in ρ_i, of $2L^2$ in $D_{i,j}$ and $\Pi_{i,j}$, and a $4L^2$ reduction in $D_{i,j,k}$, $D_{i,j,k,l}$, $\Pi^i_{j,k}$, $\Pi_{i,j,k}$, and $\Pi^{i,j}_{k,l}$.

Finally, applying the one- and two-dimensional reductions described above leads to several of the N-representability conditions to become equivalent, thus yielding additional computer time and memory savings during the solution of the SDP problem. For instance, if we consider the contraction relation $\sum_j D_{ij} = M\rho_i, \forall i$, it yields K times the same constraint $\sum_j D_{0j} = M\rho_0$. In table 1 we summarize the number of free variables (leading terms) in the SDP problem for both the standard and the symmetry-reduced p-POS conditions in one and two dimensions.

3. Results and discussion

In this section, we will discuss the computational details of the variational methodology followed by the calculation of the ground-state energy in section 3.2.1, and the spin–spin

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Table 1. The number of free variables (leading terms) in the SDP problem within the p-POS conditions. $K = L$ and L^2 for one- and two-dimensional systems, respectively.

SDP problem	2-POS	3-POS	4-POS
Without symmetry reductions	K^2	K^3	K^4
With symmetry reductions	K	K^2	K^3

correlation functions in section 3.2.2, both for one- and two-dimensional Heisenberg XXZ models.

3.1. Computational details

In this manuscript, we have considered a dual SDP problem formulation of the variational optimization of the 2-RDM in the DOCI space under three sets of N-representability constraints: the 2-POS, 3-POS, and 4-POS conditions. We have developed codes to efficiently formulate and solve the SDP problem exploiting the sparse structure of the matrices from the three sets of p-positivity conditions induced by the structure of the seniority-zero wave functions and the reductions due to translation and reflection symmetries, thus allowing to investigate larger systems than those previously studied at the same precision level. In our numerical calculations, we use the semidefinite programing algorithm (SDPA) code reported in [67, 68], which solves semidefinite problems by means of the Mehrotra-type predictor-corrector primal-dual interior-point method, providing ground-state energies and the corresponding 2-RDMs. Since the SDPA code does not allow for the equality constraints arising from the normalization, contraction, and consistency relations, they have been included by relaxing them into inequality constraints with a sufficiently small summation error δ ($\delta = 10^{-7}$), which effectively fixes the precision of the ground-state energies and 2-RDMs. In figure 1 we show the computing time per SDP iteration and the gained speedup, defined as the ratio between the computing time without and with the symmetry reductions developed in section 2.2. The results are presented as a function of the basis size K for the 2-, 3-, and 4-POS conditions. As it may be observed in the figure, the gained speedup goes up to 100 for 3- and 4-POS conditions at $K \simeq 50$. Furthermore, it is worthwhile noticing that the number of iterations needed to achieve convergence of the SDPA for the Heisenberg XXZ model lies in the range [20, 50], depending on the system size, interaction strengths, and the p-POS conditions employed. Similar values are found for the problem without the imposed symmetry reduction.

Results corresponding to QMC simulations using the stochastic series expansion [15] and the MPS calculations, both implemented in the ALPS library [69–71], have been obtained as reference values to be compared with those arising from our variational method. It is worthwhile remarking that QMC calculations are considered exact, but they usually offer access to only some magnitudes. On the other hand, MPS can be considered as exact in one dimension, but they still suffer from poor convergence scaling in two dimensions [19]. However, any observable of interest of the MPS is accessible at

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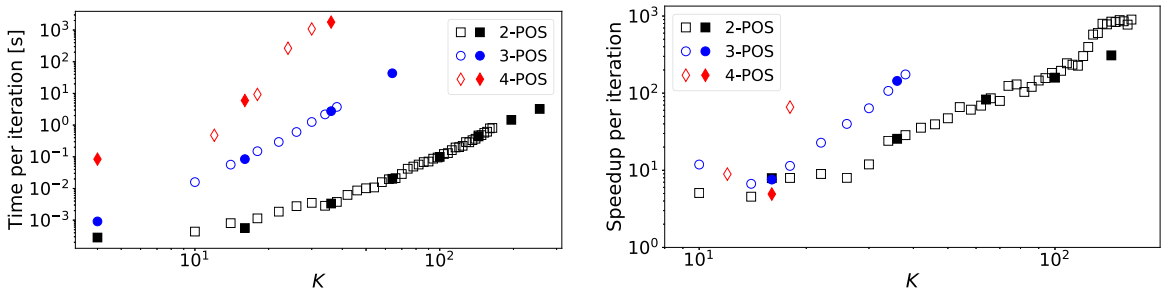

Figure 1. Time (left) and speedup (right) per iteration as a function of K for the symmetry-reduced 2-POS (squares), 3-POS (circles), and 4-POS (diamonds) conditions. Empty and solid symbols correspond to 1D and 2D systems, respectively. All measurements have been performed in an Intel Xeon CPU E5-2670 running at 2.60 GHz using a single thread for 2- and 3-POS conditions and eight threads for the 4-POS conditions.

polynomial cost. The XXZ model has been extensively studied in one-dimension, and in particular in [40, 72] it has been used to validate approximations of few-particle Green's functions or correlators for the ground state in a variational context similar to ours.

The variational optimization of the 2-RDM for the ground state of small one-dimensional spin chains following the Heisenberg XXZ model was previously analysed in [60] for the 2-POS, 3-POS, and 4-POS sets of N-representability conditions. Here, we extend these studies to larger one-dimensional periodic systems by exploiting the reductions described in section 2.2. Furthermore, we apply our variational methodology for the first time to two-dimensional systems where these reductions are specially useful. The two-dimensional system has been studied in some detail using QMC and exact diagonalizations in [73], and more recently by a high-order coupled cluster treatment in [74].

3.2. The Heisenberg XXZ model

The Heisenberg Hamiltonian is a fundamental model of quantum magnetism [75]. For spin 1/2, the XXZ version of the model is exactly solvable only in one dimension by means of the Bethe ansatz [76], and hence approximations must be considered in higher dimensions. The Hamiltonian for a general lattice $\mathcal{L}$ reads

$$H = \sum_{\langle i,j\rangle\in\mathcal{L}} \left[\frac{1}{2}\left(S_i^+ S_j^- + S_i^- S_j^+\right) + \Delta S_i^z S_j^z\right] \tag{30}$$

where $S_i^{\pm}$ and S_i^z represent the fermionic spin-ladder and spin projection operators acting at the lattice site i, $\langle i,j\rangle$ represents the nearest neighbours on the lattice and periodic boundary conditions are assumed. The parameter Δ fixes the anisotropy of the model. As a function of Δ the model has a rich phase diagram. In one dimension, for $-1 < \Delta < 1$ the system is a critical antiferromagnet with gapless excitations. For $|\Delta| > 1$ the system is gapped, being ferromagnetic for $\Delta < -1$ and antiferromagnetic for $\Delta > 1$. For the ferromagnetic phase, it can be perturbatively treated starting from a

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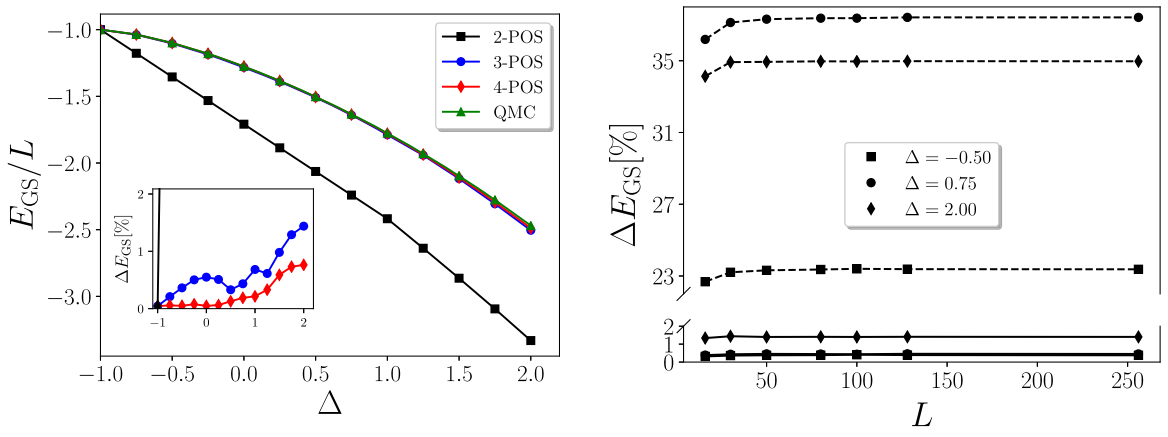

Figure 2. (Left) Ground-state energy per site E_{GS}/L as a function of the anisotropy Δ for a one-dimensional XXZ chain of $L = 30$ sites at half filling. The squares, circles, triangles and diamonds correspond to 2-POS, 3-POS, 4-POS, and QMC results. The inset shows the relative error of the ground-state energy. (Right) Relative error of the ground-state energy with respect to QMC calculations ΔE_{GS} as a function of the number of sites L for $\Delta = -0.5, 0.75$, and 2.0. The dashed and solid lines correspond to errors of the 2-POS and 3-POS calculations, respectively. The QMC simulations were calculated at an inverse temperature $\beta = 100$.

reference state where all spins are aligned in the z direction. Therefore, this phase will not be discussed in this paper and we will concentrate in the more demanding region $\Delta \geqslant -1$.

In a standard hard-core boson representation, the spin operators can be written as

$$S_i^+ = b_i^\dagger = (S_i^-)^\dagger, \qquad S_i^z = b_i^\dagger b_i - \frac{1}{2} = n_i - \frac{1}{2} \tag{31}$$

and the projection of the total spin S_{tot}^z is related to the total number of hard-core bosons by $\sum_i n_i = M = S_{tot}^z + N_s/2$, where N_s is the number of sites. Hereafter, we shall focus on a half-filled lattice, i.e. we set $M/N_s = 1/2$, which corresponds to $S_{tot}^z = 0$. In terms of these hard-core boson operators, the Hamiltonian (30) reads

$$H = E_0 + \sum_{\langle i,j\rangle \in \mathcal{L}} \left[\frac{1}{2} \left(b_i^\dagger b_j + b_j^\dagger b_i \right) + \Delta n_i n_j \right]. \tag{32}$$

This Hamiltonian, whose energy functional in terms of the Π_{ij} and D_{ij} seniority blocks of the 2-RDM is given by

$$E = E_0 + \sum_{\langle i,j\rangle \in \mathcal{L}} \left[\Pi_{i\,j} + \Delta D_{i\,j} \right], \tag{33}$$

has the same form as (1) with constant pairing and monopole interaction Δ among the nearest neighbours $\langle i, j\rangle$ of the lattice. The constant E_0 depends on the dimension and topology of the lattice and is given by $E_0 = -\frac{\gamma}{2}\Delta(M - N_s/4)$, with γ the number of nearest neighbours.

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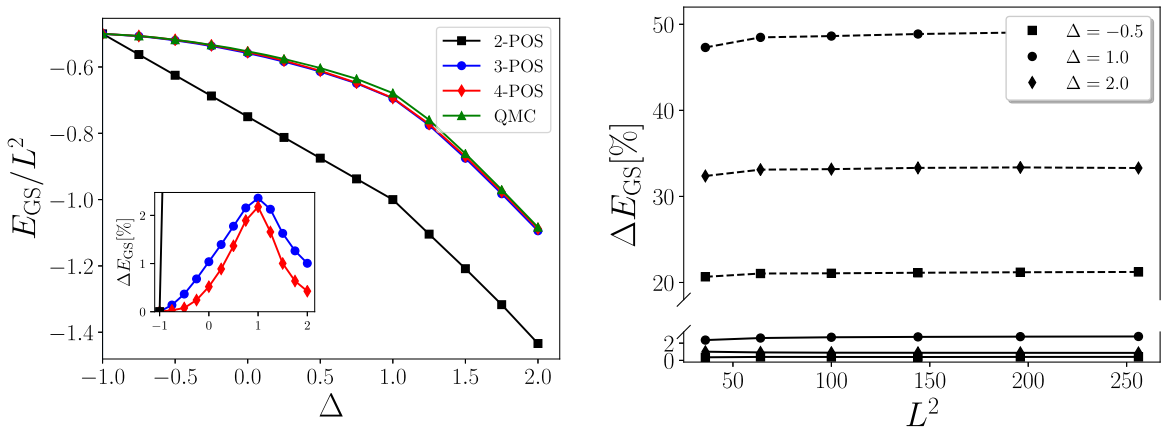

Figure 3. (Left) Ground-state energy per site E_{GS}/L^2 as a function of Δ for a 6×6 square lattice at half filling. The squares, circles, triangles, and diamonds correspond to 2-POS, 3-POS, 4-POS, and QMC results, respectively. The inset shows the relative error of the ground-state energy. (Right) Relative energy error as a function of the number of sites L^2 in a square lattice for $\Delta = -0.5, 1.0$, and 2.0. The dashed and solid lines correspond to errors of the 2-POS and 3-POS calculations, respectively. The QMC simulations were calculated at an inverse temperature $\beta = 100$.

As previously discussed in [60], for vanishing Δ, the system can also be mapped to a set of non-interacting fermions through the Jordan–Wigner transformation [77] and thus the 2-POS conditions on those fermions are sufficient N-representability conditions, whereas the 2-POS conditions on the hard-core bosons are not. On the other hand, for $\Delta = -1$ the exact ground state of the XXZ model is an antisymmetrized geminal power (AGP) and thus its ground-state energy can exactly be calculated within the 2-POS conditions in the variational treatment [53].

3.2.1. Ground-state energy. We first analyse the accuracy of the ground-state energy obtained from our variational methodology using the symmetry-reduced 2-POS, 3-POS and 4-POS conditions in a linear chain of $L = 30$ sites and a square lattice of $L = 6$ side. In the left panels of figures 2 and 3 the energy values per site are compared to QMC simulations for the range $\Delta \in [-1, 2]$. The results for the 2-POS calculations on the one-dimensional system present a maximum relative error of 37% at $\Delta = 0.5$ and a minimum at the transition point $\Delta = -1$, where the variational ground-state energy becomes exact as expected for an AGP exact eigenstate [61, 78, 79]. Similarly, 2-POS calculations on the two-dimensional system present a maximum relative error of 47% at $\Delta = 0.75$ and a minimum at $\Delta = -1$. These results show that this set of N-representability conditions is inadequate to achieve acceptable energy precision. On the other hand, the 3-POS and 4-POS conditions provide higher quality results for the energy, showing negligible differences between them, with errors below 2% and 2.5% in one and two dimensions, respectively. These results are consistent with previous observations in the literature [40, 60, 72, 79, 80].

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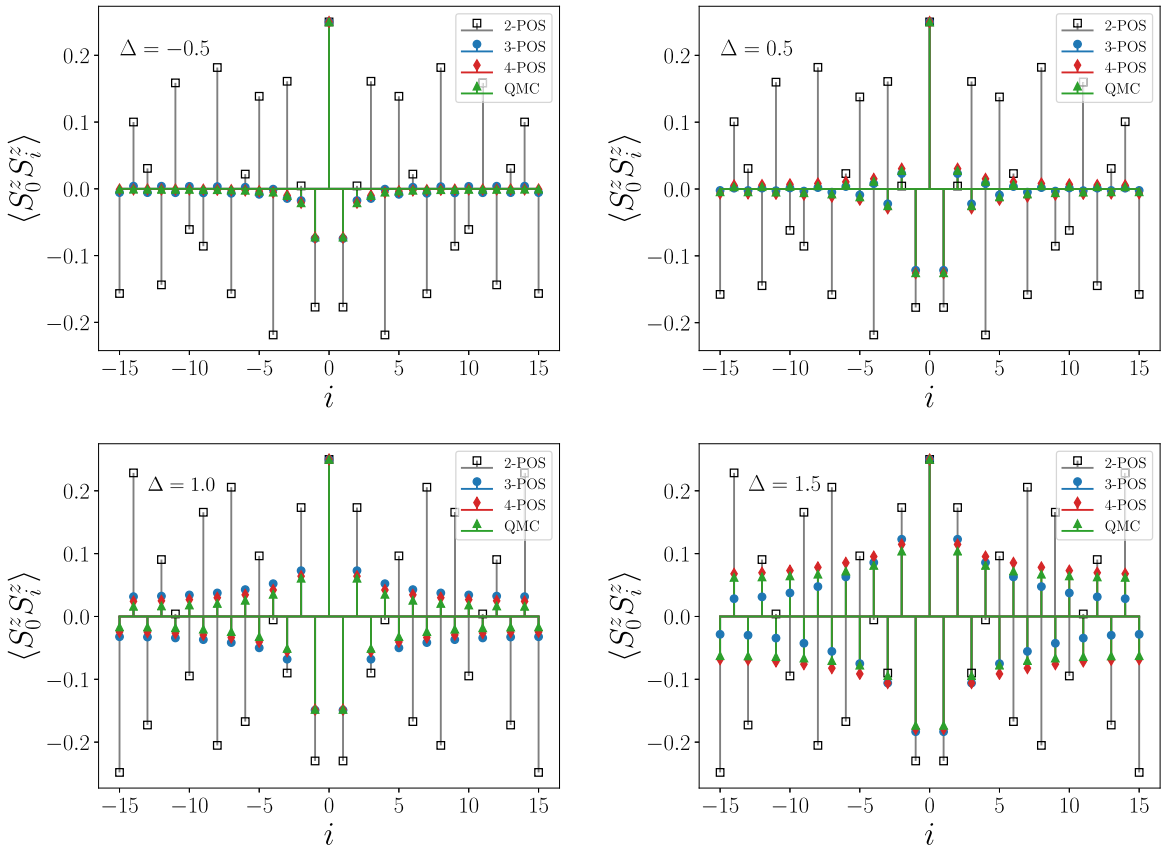

Figure 4. Spin–spin correlations along a one-dimensional chain of $L = 30$ sites at half filling for $\Delta = -0.5, 0.5, 1.0$, and 1.5. The squares, circles, diamonds, and triangles correspond to 2-POS, 3-POS, 4-POS, and QMC results, respectively.

The symmetry reductions allow us to examine the energy error of the variational calculation as a function of the system size for a broad range of lengths L. To this aim, we have investigated the accuracy of the ground-state energy computation in the 2-POS and 3-POS methodology as a function of the number of sites with chains and square lattices of up to 256 sites. In the right panels of figures 2 and 3 we present the results for the energy error $\Delta E_{\mathrm{GS}} = (E_{\mathrm{QMC}} - E_{\mathrm{v2RDM-DOCI}})/E_{\mathrm{QMC}}$ at three values of Δ. For the linear chains the relative error for the 3-POS calculation remains below 0.5% in the gapless phase of the XXZ antiferromagnet, and it increases up to about 2% in the Néel antiferromagnet at $\Delta = 2$. In the case of the square lattices, the errors follow a similar trend, with maximum relative values of about 3% for $\Delta = 1.0$. Importantly, these results for one- and two-dimensional systems show that the relative energy error does not increase with system size but it rather goes to a constant value.

3.2.2. Spin–spin correlation functions. To emphasize the validity and convenience of the variational methodology applied to moderately large systems with periodic boundary conditions, we have also computed spin–spin correlation functions. In the language of

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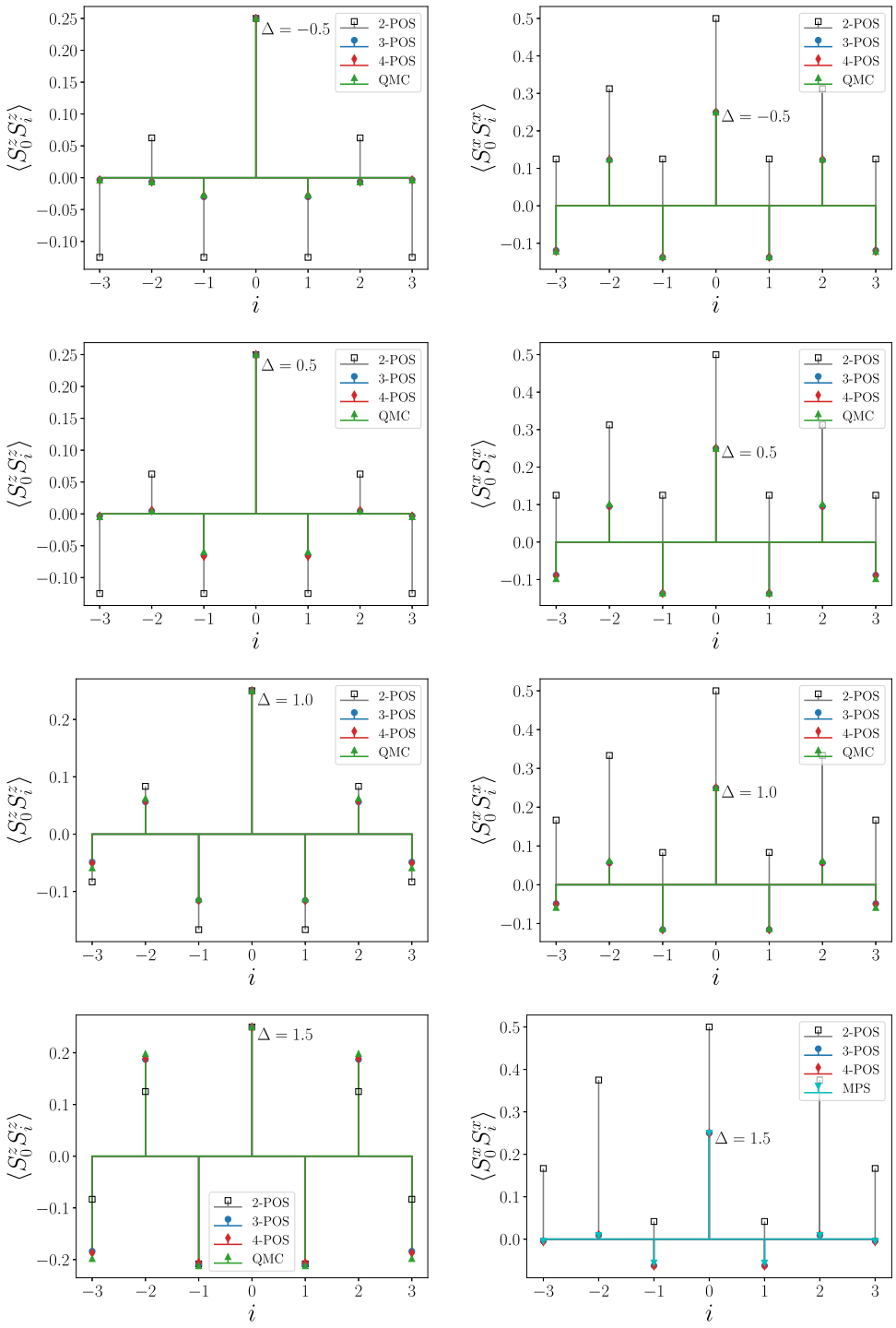

Figure 5. Spin–spin correlations $\langle S_0^k S_i^k \rangle$ in a 6×6 square lattice at half filling along an horizontal chain for $\Delta = -0.5, 0.5, 1.0$, and 1.5. Left panels correspond to the $k = z$ direction and right panels to $k = x$. The squares, circles, and diamonds correspond to 2-POS, 3-POS, and 4-POS, respectively; while the green and cyan triangles correspond to QMC and MPS results, respectively.

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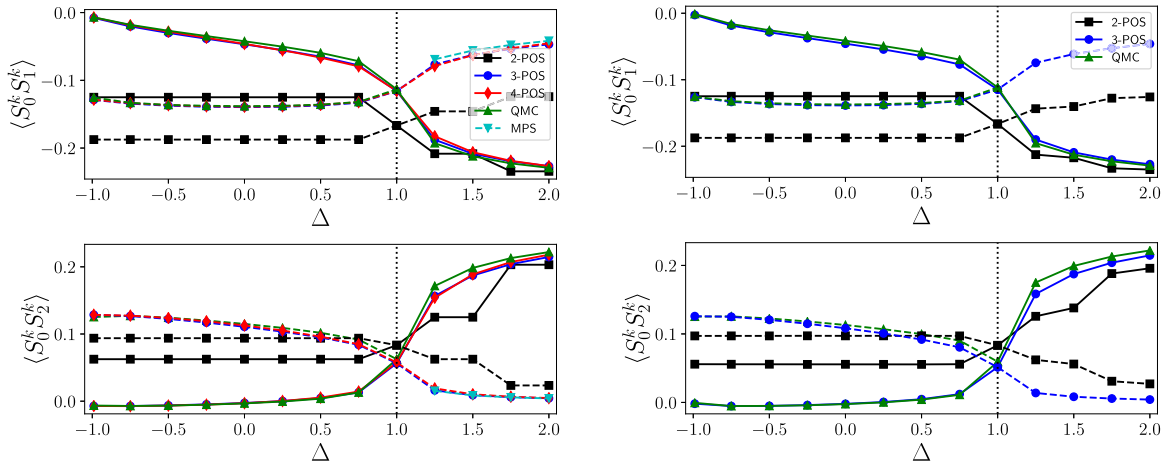

Figure 6. Spin–spin correlations $\langle S_0^k S_i^k \rangle$ as a function of Δ for a 6×6 (left column), and 16×16 (right column) square lattices. The first ($i = 1$) and second neighbours ($i = 2$) correlations are depicted in the first and second row, respectively for $k = z$ (solid lines) and $k = x$ (dashed lines) directions. The squares, circles, and diamonds correspond to 2-POS, 3-POS, and 4-POS, respectively; while the green and cyan triangles correspond to QMC and MPS results, respectively.

hard-core bosons, these correlation functions are byproducts of our methodology as

$$\langle S_i^x S_j^x \rangle = \langle S_i^y S_j^y \rangle = \frac{1}{2} \left[\Pi_{ij} + \delta_{ij}(1 - 2\rho_i) \right] \tag{34}$$

and

$$\langle S_i^z S_j^z \rangle = D_{ij} - \frac{1}{2}(\rho_i + \rho_j) + \frac{1}{4}. \tag{35}$$

The Π_{ij} seniority block of the variational 2-RDM has been deeply studied for one dimension in [60], and thus here we focus our attention on the D seniority block of the 2-RDM determining the $\langle S_i^z S_j^z \rangle$ correlation function. Under translational invariance, these functions depend only on the distance between sites i and j, so that without loss of generality we can analyse it with respect to a fixed position which we arbitrarily choose as site 0. In figure 4 we compare the correlation function at several levels of p-POS conditions for a chain of $L = 30$ sites at several values of Δ. Again the 2-POS results are incorrect for all Δ, except at $i = 0$ where its value is fixed from the hard-core boson condition $n_i^2 = n_i$ (cf equation (2)). On the other hand, the 3-POS is able to capture the spatial dependence of the correlation very well on the gapless antiferromagnet, whereas it underestimates the long-range correlation on the Néel side of the phase diagram. In accordance with previous findings in one dimension [60], near the phase transition at $\Delta = 1$, the accuracy of the two-particle correlation functions is more sensitive to the order of the p-positivity conditions utilized and therefore one may need to resort to 4-POS conditions to obtain an accurate description of the ground state.

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Let us now consider the correlation functions for the two-dimensional systems. In this case, both $\langle S_i^x S_j^x \rangle$ and $\langle S_i^z S_j^z \rangle$ functions have been studied in detail. We have calculated both correlators from the QMC simulations and the MPS calculations. Using the loop algorithm of the QMC method as implemented in the ALPS library, we computed $\langle S_i^z S_j^z \rangle$ in the full range of studied parameters, and the off-diagonal correlator $\langle S_i^x S_j^x \rangle$ for $-1 \leqslant \Delta \leqslant 1$. The MPS calculations were performed by choosing MPS parameters so as to obtain essentially identical results as those provided by QMC. We first compare the spatial dependence of the correlation corresponding to sites along a horizontal chain. The results for a 6×6 lattice at different values of Δ using 2-POS, 3-POS, 4-POS, QMC and MPS are summarized in figure 5. In all the cases, the agreement of 3-POS and 4-POS with both QMC and MPS is very good. Furthermore, we find that the 3-POS correlations differ slightly with the 4-POS results even at large distances, at variance with the one-dimensional case for $\Delta > 1$.

The determination of the spin–spin correlation functions of square lattices as a function of Δ poses a stringent test for any method. Since positive values of Δ favour the formation of antiferromagnetic domains, it is important to determine not only the first neighbour correlation, but also the second neighbour and subsequent ones in order to establish whether the system may possess a long-range order. In figure 6 we report the first- ($\langle S_0^k S_1^k \rangle$) and second-neighbour ($\langle S_0^k S_2^k \rangle$) spin-spin correlation functions for a 6×6 and a 16×16 square lattice at different N-representability levels of the variational approach. The case of the 6×6 lattice is thoroughly compared with results obtained from both QMC and MPS, while in the case of the 16×16 lattice, only QMC results are reported. As it may be seen, at $\Delta = 1$ there is a notable change in the correlators which is correctly predicted by the 3-POS and 4-POS calculations. Indeed, the first-neighbour correlation function, $\langle S_0^z S_1^z \rangle$, monotonically decreases in the whole explored range from $\Delta = -1$ to $\Delta = 2$, approaching the limiting value of $-1/4$ valid at infinite Δ. On the other hand, the second-neighbour correlation function, $\langle S_0^z S_2^z \rangle$, increases from approximately vanishing values for $\Delta < 1$ towards the maximum possible value of $1/4$. Similar agreements are observed for the other correlators $\langle S_0^x S_{1,2}^x \rangle$ in the case of the 6×6 lattice.

4. Summary and concluding remarks

We have extended the variational determination of the two-particle reduced density matrix by applying symmetry reductions stemming from the translational and reflection invariance of the ground state of an N-particle system, within a doubly-occupied-configuration-interaction space. The formalism has been presented in terms of hard-core boson operators representing the seniority-zero nature of the configuration space and applied to the spin 1/2 Heisenberg XXZ model with zero total spin along z. We have focussed on the calculation of the ground-state energy and spin–spin correlation functions for one- and two-dimensional systems of varied size.

The symmetry reductions incorporated in the calculations allowed us to determine the two-, and three-particle reduced density matrices for spin chains of lengths up to $L = 256$, and square lattices of up to $L = 16$ side. Importantly, the quality of the energy

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calculation showed that the results arising from imposing both three and 4-POS conditions do not deteriorate at increasing system size, and that the relative energy error remains below 2% in the one-dimensional model and 3% in the two-dimensional case, for the full range of Hamiltonian parameters explored. Furthermore, we have found a good agreement between the proposed variational formulation and the QMC and MPS simulations in the behaviour of the spin–spin correlation function in two dimensions already using 3-POS conditions. However, as shown in previous works with small size systems, the long-range correlations in the Neel phase of the one-dimensional XXZ Hamiltonian are underestimated unless the 4-POS conditions are taken into account. These findings suggest that 3-POS conditions may be enough to treat two-dimensional systems where correlations play a role.

In summary, we have demonstrated that the symmetry-reduced variational formulation based on the p-particle reduced density matrices provides a direct and accurate method to determine ground-state properties of moderately large systems obeying the pseudo-SU(2) algebra, as those composed by hard-core bosons. This methodology could be used as a starting point to study excited states on these systems by applying, e.g. random phase approximations on top of such correlated ground states [81, 82].

Acknowledgments

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Appendix A. Positive semidefiniteness conditions for N-representability of the 3- and 4-RDM in the DOCI space

As mentioned in section 2.1, restricting the operator $B^\dagger$ in equation (8) to the two-particle space leads to the 2-$\mathcal{P}$, 2-$\mathcal{Q}$ and 2-$\mathcal{G}$ N-representability conditions. Similarly, restricting the operator $B^\dagger$ to the three-particle seniority-zero subspace leads to the 3-POS conditions 3-$\mathcal{P}$, 3-$\mathcal{Q}$, 3-$\mathcal{E}$ and 3-$\mathcal{F}$ [53, 54], which can be expressed in terms of the two seniority blocks of the 3-RDM, (15) and (16), as follows

- The 3-$\mathcal{P}$ condition:

$$D_{ijk} \geqslant 0, \quad \forall\ i < j < k \tag{A.1}$$

$$\Pi^a_{ij} \succeq 0, \quad \forall\ a \tag{A.2}$$

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which thus posesses $O(K^3)$ blocks of dimension $O(1\times 1)$ and $O(K)$ blocks of dimension $O(K\times K)$.

- The 3-$\mathcal{Q}$ condition:

$$1-\rho_i-\rho_j-\rho_k+D_{ij}+D_{jk}+D_{ki}-D_{ijk}\geqslant 0,\quad \forall\ i<j<k. \tag{A.3}$$

$$-\Pi^a_{ij}+\Pi_{ji}+\delta_{ij}\left(1-2\rho_i-\rho_a+2D_{ia}\right)\succeq 0,\quad \forall\ ij\neq a \tag{A.4}$$

possessing $O(K^3)$ blocks of dimension $O(1\times 1)$ and $O(K)$ blocks of dimension $O(K\times K)$.

- The 3-$\mathcal{E}$ condition:

$$\begin{pmatrix} D_{ab}-D_{abc} & \Pi^a_{bc} & \Pi^b_{ac} \\ \Pi^a_{bc} & D_{ac}-D_{abc} & \Pi^c_{ab} \\ \Pi^b_{ac} & \Pi^c_{ab} & D_{bc}-D_{abc} \end{pmatrix}\succeq 0,\quad \forall\ a<b<c \tag{A.5}$$

$$\begin{pmatrix} D_{aij}+\delta_{ij}D_{ai} & \Pi^i_{aj} & D_{ia} \\ \Pi^j_{ai} & \Pi_{ij}-\Pi^a_{ij} & \Pi_{ia} \\ D_{ai} & \Pi_{ai} & \rho_a \end{pmatrix}\succeq 0,\quad \forall\ a,\quad ij\neq a, \tag{A.6}$$

which posesses $O(K^3)$ blocks of dimension $O(1\times 1)$ and and $O(K)$ blocks of dimension $O(K\times K)$.

- The 3-$\mathcal{F}$ condition:

$$\begin{pmatrix} \rho_a-D_{ab}-D_{ac}+D_{abc} & \Pi_{ac}-\Pi^b_{ac} & \Pi_{ab}-\Pi^c_{ab} \\ \Pi_{ac}-\Pi^b_{ac} & \rho_c-D_{bc}-D_{ac}+D_{abc} & \Pi_{bc}-\Pi^a_{bc} \\ \Pi_{ab}-\Pi^c_{ab} & \Pi_{bc}-\Pi^a_{bc} & \rho_b-D_{ab}-D_{bc}+D_{abc} \end{pmatrix}\succeq 0$$

$$\forall\ a<b<c. \tag{A.7}$$

$$\begin{pmatrix} -D_{aij}+\delta_{ij}\left(\rho_i-D_{ai}\right)+D_{ij} & -\Pi^i_{aj}-\delta_{ij}\Pi_{ia} & \rho_i-D_{ia} \\ -\Pi^i_{aj}-\delta_{ij}\Pi_{ia} & \Pi^a_{ij}+\delta_{ij}\left(\rho_a-2D_{ia}\right) & -\Pi_{ia} \\ \rho_i-D_{ai} & -\Pi_{ai} & 1-\rho_a \end{pmatrix}\succeq 0$$

$$\forall\ a,\quad ij\neq a, \tag{A.8}$$

which posesses $O(K^3)$ blocks of dimension $O(1\times 1)$ and $O(K)$ blocks of dimension $O(K\times K)$.

It is important to note that, for the conditions that are split into multiple blocks, we will label the blocks with the indices $a,b,c,\ldots$, and the elements inside each block with indices $i,j,k,\ldots$.

The above 3-POS conditions are supplemented with the contraction and consistency relations

$$\rho_i=\frac{1}{M-1}\left(\sum_{j<i}\Pi^j_{ii}+\sum_{i>j}\Pi^i_{jj}\right),\quad \forall\ i \tag{A.9}$$

$$\Pi_{ij}=\frac{1}{M-1}\sum_{k\neq ij}\Pi^k_{ij},\quad \forall\ i<j \tag{A.10}$$

$$D_{ij} = D_{iij} = \Pi^{i}_{jj} = \Pi^{j}_{ii} \tag{A.11}$$

$$\Pi^{i}_{jj} = \frac{1}{M-2}\sum_{k\neq ij} D_{ijk}, \quad \forall\ i<j. \tag{A.12}$$

Five different choices of the operator $B^{\dagger}$ in equation (8) restricted to the four-particle seniority-zero subspace give rise to the 4-POS conditions 4-$\mathcal{P}$, 4-$\mathcal{Q}$, 4-$\mathcal{E}$, 4-$\mathcal{F}$ and 4-$\mathcal{G}$ [53, 54], which can be expressed in terms of the seniority blocks of the 4-RDM in equations (17)–(19) as follows:

- The 4-$\mathcal{P}$ condition:

$$D_{ijkl} \geqslant 0, \quad \forall\ i<j<k<l. \tag{A.13}$$

$$\Pi^{ab}_{ij} \succeq 0, \quad \forall\ a<b, i, j \tag{A.14}$$

$$\Pi_{ijkl} \succeq 0 \quad \forall\ i<j, \quad k<l \quad \text{with matrix indices } ij \text{ and } kl. \tag{A.15}$$

  This condition possesses $O(K^4)$ blocks of dimension $O(1\times 1)$, $O(K^2)$ blocks of dimension $O(K\times K)$, and $O(1)$ blocks of dimension $O(K^2\times K^2)$, respectively.

- The 4-$\mathcal{Q}$ condition:

$$\begin{aligned} &1-\rho_i-\rho_j-\rho_k-\rho_l+D_{ij}+D_{ik}+D_{il}+D_{jk}+D_{jl}+D_{kl}-D_{jkl}\\ &-D_{ikl}-D_{ijl}-D_{ijk}+D_{ijkl}\geqslant 0, \quad \forall\ i<j<k<l. \end{aligned} \tag{A.16}$$

$$\begin{aligned} &\Pi_{ij}-\Pi^{a}_{ij}-\Pi^{b}_{ij}+\Pi^{ab}_{ij}+\delta_{ij}\left(1-\rho_a-\rho_b-2\rho_i+D_{ab}+2D_{ia}+2D_{ib}-2D_{iab}\right)\succeq 0,\\ &\forall\ a,b,i\neq a,b, \quad j\neq a,b \end{aligned} \tag{A.17}$$

$$\begin{aligned} &\Pi_{ijkl}+\delta_{ik}\left(\Pi_{lj}-2\Pi^{i}_{lj}\right)+\delta_{jk}\left(\Pi_{li}-2\Pi^{j}_{li}\right)+\delta_{il}\left(\Pi_{kj}-2\Pi^{i}_{kj}\right)\\ &+\delta_{jl}\left(\Pi_{ki}-2\Pi^{j}_{ki}\right)+\delta_{ik}\delta_{jl}\left(1-2\rho_i-2\rho_j+4D_{ij}\right)\succeq 0,\\ &\forall\ i,j,k,l \quad \text{with matrix indices } ij \text{ and } kl. \end{aligned} \tag{A.18}$$

  This condition also possesses $O(K^4)$ blocks of dimension $O(1\times 1)$, $O(K^2)$ blocks of dimension $O(K\times K)$, and $O(1)$ blocks of dimension $O(K^2\times K^2)$, respectively.

- The 4-$\mathcal{E}$ condition:

$$\begin{pmatrix} D_{abc}-D_{abcd} & \Pi^{bc}_{ad} & \Pi^{ac}_{bd} & \Pi^{ab}_{cd}\\ \Pi^{bc}_{ad} & D_{bcd}-D_{abcd} & \Pi^{cd}_{ab} & \Pi^{bd}_{ac}\\ \Pi^{ac}_{bd} & \Pi^{cd}_{ab} & D_{cda}-D_{abcd} & \Pi^{ad}_{bc}\\ \Pi^{ab}_{cd} & \Pi^{bd}_{ac} & \Pi^{ad}_{bc} & D_{dab}-D_{abcd} \end{pmatrix} \succeq 0, \tag{A.19}$$

$$\forall\ a<b<c, d\neq abc.$$

$$\begin{pmatrix} D_{ijab} & \Pi^{ia}_{jb} & \Pi^{ib}_{aj}\\ \Pi^{ja}_{ib} & \Pi^{a}_{ij}-\Pi^{ab}_{ij} & \Pi_{iajb}\\ \Pi^{jb}_{ia} & \Pi_{ibja} & \Pi^{b}_{ij}-\Pi^{ab}_{ij} \end{pmatrix} \succeq 0, \quad \forall\ ij\neq ab. \tag{A.20}$$

  This condition results in $O(K^4)$ blocks of dimension $O(1\times 1)$, and $O(K^2)$ blocks of dimension $O(K\times K)$.

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- The 4-$\mathcal{F}$ condition:

$$\begin{pmatrix} (a) & \Pi_{ab} - \Pi^{c}_{ab} - \Pi^{d}_{ab} + \Pi^{cd}_{ab} & \Pi_{ac} - \Pi^{b}_{ac} - \Pi^{d}_{ac} + \Pi^{bd}_{ac} & \Pi_{ad} - \Pi^{b}_{ad} - \Pi^{c}_{ad} + \Pi^{bc}_{ad} \\ \dots & (b) & \Pi_{bc} - \Pi^{a}_{bc} - \Pi^{d}_{bc} + \Pi^{ad}_{bc} & \Pi_{bd} - \Pi^{a}_{bd} - \Pi^{c}_{bd} + \Pi^{ac}_{bd} \\ \dots & \dots & (c) & \Pi_{cd} - \Pi^{a}_{cd} - \Pi^{b}_{cd} + \Pi^{ab}_{bd} \\ \dots & \dots & \dots & (d) \end{pmatrix} \succeq 0, \tag{A.21}$$

where the matrix is symmetric and the notation () represents

$$(a) = \rho_a - D_{ab} - D_{ac} - D_{ad} + D_{abc} + D_{abd} + D_{acd} - D_{abcd}. \tag{A.22}$$

and

$$\begin{pmatrix} D_{ij} - D_{ija} - D_{ijb} + D_{ijab} & \Pi^{i}_{ja} - \Pi^{ib}_{ja} + \delta_{ij}\left(\Pi_{ia} - \Pi^{b}_{ia}\right) & a_1 \\ \Pi^{j}_{ia} - \Pi^{jb}_{ia} + \delta_{ij}\left(\Pi_{ja} - \Pi^{b}_{ja}\right) & \Pi^{a}_{ij} - \Pi^{ab}_{ij} + \delta_{ij}\left(\rho_a - D_{ab} - 2D_{ia} + 2D_{iab}\right) & a_2 \\ \Pi^{j}_{ib} - \Pi^{ja}_{ib} + \delta_{ij}\left(\Pi_{jb} - \Pi^{a}_{jb}\right) & \Pi_{aijb} + \delta_{ij}\left(\Pi_{ab} - 2\Pi^{i}_{ab}\right) & a_3 \end{pmatrix} \succeq 0, \tag{A.23}$$

with

$$\begin{aligned} a_1 &= \Pi^{i}_{jb} - \Pi^{ia}_{jb} + \delta_{ij}\left(\Pi_{ib} - \Pi^{a}_{ib}\right), \\ a_2 &= \Pi_{ajib} + \delta_{ij}\left(\Pi_{ab} - 2\Pi^{i}_{ab}\right), \quad \text{and} \\ a_3 &= \Pi^{b}_{ij} - \Pi^{ab}_{ij} + \delta_{ij}\left(\rho_b - D_{ab} - 2D_{ib} + 2D_{iab}\right). \end{aligned} \tag{A.24}$$

This condition also results in $O(K^4)$ blocks of dimension $O(1 \times 1)$, and $O(K^2)$ blocks of dimension $O(K \times K)$.

- The 4-$\mathcal{G}$ condition: the mixed two-particle-two-hole representation yields

$$\begin{pmatrix} (ab) & \Pi^{a}_{bd} - \Pi^{ac}_{bd} & \Pi^{b}_{ad} - \Pi^{bc}_{ad} & \Pi^{a}_{bc} - \Pi^{ad}_{bc} & \Pi^{b}_{ac} - \Pi^{bd}_{ac} & \Pi_{abcd} \\ \Pi^{a}_{bd} - \Pi^{ac}_{bd} & (ad) & \Pi^{d}_{ab} - \Pi^{cd}_{ab} & \Pi^{a}_{cd} - \Pi^{ab}_{cd} & \Pi_{adbc} & \Pi^{d}_{ac} - \Pi^{bd}_{ac} \\ \Pi^{b}_{ad} - \Pi^{bc}_{ad} & \Pi^{d}_{ab} - \Pi^{cd}_{ab} & (bd) & \Pi_{bdac} & \Pi^{b}_{cd} - \Pi^{ab}_{cd} & \Pi^{d}_{bc} - \Pi^{ad}_{bc} \\ \Pi^{a}_{bc} - \Pi^{ad}_{bc} & \Pi^{a}_{cd} - \Pi^{ab}_{cd} & \Pi_{acbd} & (ac) & \Pi^{c}_{ab} - \Pi^{cd}_{ab} & \Pi^{c}_{ad} - \Pi^{bc}_{ad} \\ \Pi^{b}_{ac} - \Pi^{bd}_{ac} & \Pi_{bcad} & \Pi^{b}_{cd} - \Pi^{ab}_{cd} & \Pi^{c}_{ab} - \Pi^{cd}_{ab} & (bc) & \Pi^{c}_{bd} - \Pi^{ac}_{bd} \\ \Pi_{cdab} & \Pi^{d}_{ac} - \Pi^{bd}_{ac} & \Pi^{d}_{bc} - \Pi^{ad}_{bc} & \Pi^{c}_{ad} - \Pi^{bc}_{ad} & \Pi^{c}_{bd} - \Pi^{ac}_{bd} & (cd) \end{pmatrix} \succeq 0, \tag{A.25}$$

with, for example,

$$\begin{aligned} (ab) &= D_{ab} - D_{abc} - D_{abd} + D_{abcd} \\ (bc) &= D_{bc} - D_{abc} - D_{bcd} + D_{abcd}, \end{aligned} \tag{A.26}$$

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$$\begin{pmatrix} D_{ijb} - D_{ijab} & \Pi^{ij}_{ab} & \Pi^{ib}_{ja} + \delta_{ij}\Pi^{b}_{aj} & \Pi^{i}_{jb} - \Pi^{ia}_{jb} \\ \Pi^{ij}_{ab} & D_{ija} - D_{ijab} & \Pi^{ia}_{jb} + \delta_{ij}\Pi^{a}_{bj} & \Pi^{i}_{ja} - \Pi^{ib}_{ja} \\ \Pi^{jb}_{ia} + \delta_{ij}\Pi^{b}_{aj} & \Pi^{aj}_{ib} + \delta_{ij}\Pi^{a}_{bj} & \Pi^{ab}_{ij} + \delta_{ij}\left(D_{ab} - 2D_{iab}\right) & \Pi_{abij} \\ \Pi^{j}_{ib} - \Pi^{ja}_{ib} & \Pi^{j}_{ia} - \Pi^{jb}_{ia} & \Pi_{ijba} & \Pi_{ij} - \Pi^{a}_{ij} - \Pi^{b}_{ij} + \Pi^{ab}_{ij} \end{pmatrix} \succeq 0, \tag{A.27}$$

and

$$\Pi_{ilkj} + \delta_{jl}\left(\Pi_{ik} - 2\Pi^{j}_{ik}\right) \succeq 0, \quad \text{with matrix indices } ij \text{ and } kl. \tag{A.28}$$

This last condition results in $O(K^4)$ blocks of dimension $O(1 \times 1)$, $O(K^2)$ blocks of dimension $O(K \times K)$, and $O(1)$ blocks of dimension $O(K^2 \times K^2)$.

The elements of the seniority-zero blocks of the 4-RDM must satisfy the following contraction and consistency relations with the 2- and 3-RDMs:

$$\sum_{j \neq il} \Pi^{ij}_{kl} = (M-2)\Pi^{i}_{kl} \tag{A.29}$$

$$\sum_{i \neq jkl} D_{ijkl} = (M-3)D_{jkl} \tag{A.30}$$

$$\Pi_{ikkj} = \Pi^{k}_{ij}, \qquad \Pi_{ijij} = D_{ij}, \tag{A.31}$$

$$\Pi^{ij}_{kk} = D_{ijk}, \quad D_{ijkk} = D_{ijk}. \tag{A.32}$$

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