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Improved upper and lower energy bounds for antiferromagnetic Heisenberg spin systems

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Abstract. Large spin systems as given by magnetic macromolecules or two-dimensional spin arrays rule out an exact diagonalization of the Hamiltonian. Nevertheless, it is possible to derive upper and lower bounds of the minimal energies, *i.e.* the smallest energies for a given total spin S. The energy bounds are derived under additional assumptions on the topology of the coupling between the spins. The upper bound follows from "n-cyclicity", which roughly means that the graph of interactions can be wrapped round a ring with n vertices. The lower bound improves earlier results and follows from "n-homogeneity", *i.e.* from the assumption that the set of spins can be decomposed into n subsets where the interactions inside and between spins of different subsets fulfill certain homogeneity conditions. Many Heisenberg spin systems comply with both concepts such that both bounds are available. By investigating small systems which can be numerically diagonalized we find that the upper bounds are considerably closer to the true minimal energies than the lower ones.

PACS. 75.10.Jm Quantized spin models – 75.50.Xx Molecular magnets – 75.50.Ee Antiferromagnetics – 75.40.Mg Numerical simulation studies

1 Introduction

Rigorous results on spin systems like the Marshall-Peierls sign rule [1] and the famous theorems of Lieb *et al.* [2,3] have sharpened our understanding of magnetic phenomena. In addition such results can serve as a basis or source of inspiration for the development of approximate models. For example, the inequalities of Lieb and Berezin [4,5] relating spectral properties of quantum systems to those of their classical counterparts provide a foundation for classical or semi-classical treatments of spin systems.

In this article we will extend the body of rigorous results on Heisenberg spin systems by generalizing the notion of "bi-partiteness", which is fundamental for the findings of Marshall *et al.* [1–3]. We will introduce two new concepts which rest on the topological properties of the interaction matrix connecting the spins of the systems.

The first concept, n-cyclicity, uses the property of many spin systems that their "net" of interactions can be wrapped round an n-cycle. The triangular lattice may serve as an example, it can be mapped onto a triangle in an oriented manner as if one would wrap it round the triangle. In such cases an upper bound of the minimal energies $E_{\min}$ in each subspace $\mathcal{H}(M)$ of total magnetic quantum number M can be derived for Heisenberg models and XXZ models in general.

The second concept which leads to lower bounds rests on n-homogeneity, *i.e.* on the fact that the set of spins can be decomposed into n subsets of equal size where the interactions inside and between spins of different subsets fulfill certain conditions.

Fortunately, many Heisenberg spin systems comply with both concepts such that both bounds are available. For all cases which were investigated it turns out that the upper bounds are rather close to the true minimal energies, whereas the lower bounds are not. Therefore, especially the upper bound can serve are a benchmark or guideline for approximate methods like DMRG or variational methods in order to rate the achieved quality.

The resulting bounds improve earlier findings of reference [4–6] especially for frustrated spin systems.

The article is organized as follows. In Section 2 upper bounds and the concept of n-cyclicity will be discussed, in Section 3 lower bounds and n-homogeneity will be introduced. Both sections start with subsections explaining the idea followed by more mathematical subsections presenting the mathematical tools. At the end of each section the resulting bounds are given. Examples are provided in Section 4. A more technical calculation is carried out in Appendix A.

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2 Upper bounds

2.1 Idea

It is obvious from the Ritz variational principle that an upper bound for the minimal energy can be provided if an appropriate trial state can be found for which the energy expectation value is known analytically and rather close to the exact ground state value. In the following we will construct such trial states for subspaces $\mathcal{H}(M)$, *i.e.* for total magnetic quantum number M. Starting point is the magnon vacuum ($M = Ns$) which is mapped by means of suitable powers of the total ladder operator into the subspace $\mathcal{H}(M)$. Since the total ladder operator commutes with the Heisenberg Hamilton operator this does not change the energy of that state. In a second step we assume a certain topological property of the spin array namely that it can be wrapped round an n-cycle and construct a generalized "Bloch operator" which is a unitary operator that adds appropriate phases to the components of the trial state. Utilizing the known action of the Bloch operator onto the Hamiltonian we can evaluate the energy expectation value analytically which results in the expression for the upper bound.

In order to motivate our definitions in the next subsection we recall the definition of a bi-partite spin system in the case of constant coupling: It is required that the spin sites can be grouped into +sites and −sites such that only $+-$ pairs are coupled but no $++$ or $--$ pairs. We suggest the following generalization: Assume that complex phase factors $\mathrm{e}^{\mathrm{i}\phi_j}$ can be attached to spin sites j such that only constant phase differences $|\phi_j - \phi_k|$ occur between adjacent (coupled) spin sites. This is the requirement needed for the above-mentioned construction of the Bloch operator. The attachment of phase factors is no longer arbitrary if there are "loops" in the coupling scheme of the spin system, *i.e.* periodic sequences of adjacent spin sites. If only even loops exist we may choose the phase differences to be $|\phi_j - \phi_k| = \pi$ and the system is bi-partite. However, in the case of odd loops it becomes necessary to "wrap" the loop around the complex unit circle and the resulting phase differences will be integer fractions of 2π. We will make this more precise in the next subsection employing the language of graph theory.

2.2 Definition of n-cyclicity

In this section we consider systems with N spin sites with spin s and constant anti-ferromagnetic coupling. Thus the complete information about the coupling scheme is encoded in some (undirected) graph $\gamma = (\mathcal{V}, \Gamma)$. The vertices of γ are the spin sites, $\mathcal{V} = \{1, \ldots, N\}$, the set of edges of γ consists of those pairs of sites which are coupled and will be denoted by Γ. We make the convention that $\langle i, j\rangle \in \Gamma$ iff $\langle j, i\rangle \in \Gamma$ and $\langle i, i\rangle \notin \Gamma$. Hence the number of members of the set Γ, denoted by $|\Gamma|$, equals twice the number of bonds. Further, we will consider *orientations* on γ, denoted by γ^+, *i.e.* we split Γ into disjoint subsets $\Gamma = \Gamma^+ \cup \Gamma^-$, such that $\langle i, j\rangle \in \Gamma^+$ iff $\langle j, i\rangle \in \Gamma^-$.

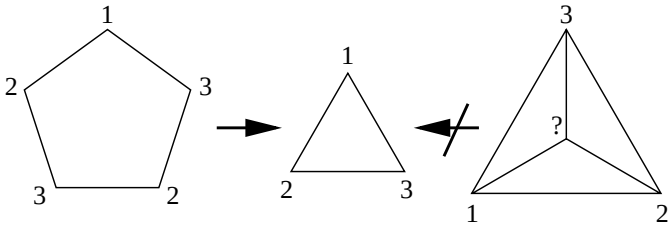

Fig. 1. The pentagon is 5-cyclic and also 3-cyclic (l.h.s.) whereas the tetrahedron is not 3-cyclic (r.h.s.), because if the four vertices of the tetrahedron are attached to the numbers $1, 2, 3$ one number must repeat and occurs at adjacent vertices, which does not happen in the 3-cycle.

Then the Hamiltonian of XXZ-type can be written in the form

$$H = \delta \sum_{\langle i,j\rangle \in \Gamma} s_i^{(3)} s_j^{(3)} + \sum_{\langle i,j\rangle \in \Gamma^+} s_i^+ s_j^- + \sum_{\langle i,j\rangle \in \Gamma^-} s_i^+ s_j^- \quad (1)$$

$$\equiv \Delta + \mathcal{G} + \mathcal{G}^\dagger, \quad (2)$$

where $\delta > 0$ and $\boldsymbol{s}_i$ denote the usual spin observables at site i with components $s_i^{(\mu)}$, $\mu = 1, 2, 3$, and $s_i^\pm \equiv s_i^{(1)} \pm i s_i^{(2)}$. Of course, only the splitting (2) depends on the orientation, not the Hamiltonian itself.

In order to define a suitable concept of n-cyclicity we consider *graph homomorphisms*, *i.e.* maps between graphs, such that vertices are mapped onto vertices and the corresponding edges onto corresponding edges. Let $\mathcal{C}_n$ denote the cyclic graph with n vertices which will be identified with the nth roots of unity

$$\mathrm{e}^{\mathrm{i}\alpha_\ell} \equiv \exp\left(\frac{2\pi \mathrm{i}\ell}{n}\right), \qquad \ell = 0, \ldots, n-1\ . \quad (3)$$

Further $\mathcal{C}_n^+$ will denote the cyclic graph with anti-clockwise orientation.

Any graph γ (or the spin system itself) will be called n-cyclic or having the cyclicity n iff there exists a graph homomorphism

$$h : \gamma \longrightarrow \mathcal{C}_n\ . \quad (4)$$

In this case $\mathcal{C}_n^+$ will induce an orientation on γ in an obvious sense.

It is only in certain cases that different cyclicities n and n' mean an essential distinctness. This is because for $n \geq 4$ any n-cyclic system is also $(n-2)$-cyclic since three successive vertices and the corresponding edges can be mapped in a forward-backward-forward way, compare the l.h.s. of Figure 1, which shows a homomorphism of a pentagon onto a triangle, as an example. Each $2m$-ring and hence any $2m$-cyclic system is n-cyclic for any positive integers m, n, since it is 2-cyclic and $\mathcal{C}_2$ can be homomorphically embedded into any n-cycle.

Hence it makes only sense to distinguish between even-cyclic systems, which will be called 2-cyclic, and $(2n+1)$-cyclic system with maximal integer n. If a spin system is 2-cyclic in our sense it will be bi-partite in the sense of

references [2,3], where, however, the theory also comprises cases with different coupling constants.

We consider some more examples which illustrate the definition of cyclicity. A triangular plane lattice with suitable periodic boundary conditions is 3-cyclic, a square lattice or cubic lattice is 2-cyclic. The kagomé lattice is 3-cyclic but not 2-cyclic. 3-cyclicity is equivalent to 3-colorability. Hence the octahedron, the dodecahedron, the cuboctahedron, and the icosidodecahedron are 3-cyclic, *cf.* [7], but the tetrahedron is not, see r.h.s. of Figure 1.

A natural basis for a matrix representation of H is provided by the product states $|\,\mathbf{m}\,\rangle = |\,m_1 \dots m_N\,\rangle$, $-s \leq m_i \leq s$ with

$$s_i^{(3)}\,|\,\mathbf{m}\,\rangle = m_i\,|\,\mathbf{m}\,\rangle\ , \tag{5}$$

and

$$s_i^{\pm}\,|\,\mathbf{m}\,\rangle = \sqrt{s(s+1) - m_i(m_i \pm 1)}\,|\,m_1, \dots, m_i \pm 1, \dots, m_N\,\rangle \tag{6}$$

for all $i \in \{1, \dots, N\}$. The state $|\,\Omega\,\rangle \equiv |\,s, s, \dots, s\,\rangle$ will be called the "magnon vacuum". Further we define

$$a \equiv \sum_{i=1}^{N} a_i = Ns - M, \quad a_i = s - m_i \ \text{ for all } i \in \{1, \dots, N\}. \tag{7}$$

We also define a mapping $\hat{h}$ of product states into complex numbers which depends on the graph homomorphism (4) by

$$\hat{h}(\mathbf{m}) \equiv \prod_{i=1}^{N} h(i)^{a_i}\ . \tag{8}$$

Then it is easily shown that if $\langle \mathbf{m}|\mathcal{G}|\mathbf{m}'\rangle \neq 0$ then $\hat{h}(\mathbf{m}) = \mathrm{e}^{2\pi \mathrm{i}/n}\hat{h}(\mathbf{m}')$.

For any $\ell = 0, \dots, N-1$ we define a unitary "Bloch operator" (generalizing the corresponding definition for spin rings in reference [8])

$$U_\ell : \mathcal{H}(M) \longrightarrow \mathcal{H}(M) \tag{9}$$

by

$$U_\ell|\mathbf{m}\rangle = \hat{h}(\mathbf{m})^\ell|\mathbf{m}\rangle \tag{10}$$

and linear extension. Recall that $\alpha_\ell = 2\pi\ell/n$. Then the following relations hold:

$$U_\ell^\dagger \mathcal{G} U_\ell = \mathrm{e}^{-\mathrm{i}\alpha_\ell}\mathcal{G} \tag{11}$$

$$U_\ell^\dagger H U_\ell = \Delta + \cos\alpha_\ell(\mathcal{G} + \mathcal{G}^\dagger) - \mathrm{i}\sin\alpha_\ell(\mathcal{G} - \mathcal{G}^\dagger). \tag{12}$$

If $E_{\min}(M)$ denotes the minimal energy eigenvalue within the sector $\mathcal{H}(M)$ and $|\,\varphi\,\rangle \in \mathcal{H}(M)$ is an arbitrary normalized state we have the obvious upper bound

$$E_{\min}(M) \leq \langle\varphi|H|\varphi\rangle\ . \tag{13}$$

The problem is to find a state $|\,\varphi\,\rangle$ such that $\langle\varphi|H|\varphi\rangle$ can be explicitly calculated and represents a close bound. To this end we map the magnon vacuum $|\,\Omega\,\rangle$ by $(S^-)^a$ into $\mathcal{H}(M)$, which remains an eigenstate of H with the largest eigenvalue in the Heisenberg case $\delta = 1$, and change its phases according to the Bloch operator. More precisely, let

$$|\Omega_M\rangle \equiv C_M(S^-)^a|\Omega\rangle\ , \tag{14}$$

where C_M is the positive normalization factor, compare (52), ensuring $\langle\,\Omega_M\,|\,\Omega_M\,\rangle = 1$ and define

$$|\,\varphi\,\rangle \equiv |\,\varphi_{M,\ell}\,\rangle \equiv U_\ell|\Omega_M\rangle. \tag{15}$$

Then we obtain

$$\langle\Omega_M|\Delta|\Omega_M\rangle = \frac{\delta|\Gamma|}{N}\left\{Ns^2 - \frac{2sa(2Ns-a)}{2Ns-1}\right\}\ . \tag{16}$$

As it must be, this result has the obvious value $\delta|\Gamma|s^2$ for $a = 0$ and remains unchanged under $a \leftrightarrow 2Ns - a$. The proof of equation (16) is given in Appendix A.

Now consider

$$\langle\varphi|H|\varphi\rangle = \langle\Omega_M|U_\ell^\dagger H U_\ell|\Omega_M\rangle \tag{17}$$

$$= \langle\Omega_M|\Delta + \cos\alpha_\ell(\mathcal{G} + \mathcal{G}^\dagger)|\Omega_M\rangle \tag{18}$$

$$= \cos\alpha_\ell\langle\Omega_M|\frac{\Delta}{\delta} + (\mathcal{G} + \mathcal{G}^\dagger)|\Omega_M\rangle \tag{19}$$

$$+(1 - \frac{\cos\alpha_\ell}{\delta})\langle\Omega_M|\Delta|\Omega_M\rangle$$

$$= |\Gamma|s^2\cos\alpha_\ell \tag{20}$$

$$+(1 - \frac{\cos\alpha_\ell}{\delta})\frac{\delta|\Gamma|}{N}\left(Ns^2 - \frac{2sa(2Ns-a)}{2Ns-1}\right)\ .$$

In line (18) we used (12) and the fact that $|\,\Omega_M\,\rangle$ and $\mathcal{G}$ are real in the product basis of the $|\mathbf{m}\rangle$ whence $\langle\Omega_M|(\mathcal{G} - \mathcal{G}^\dagger)|\Omega_M\rangle = 0$. Equation (20) follows by equation (16) and the observation that $\frac{\Delta}{\delta} + (\mathcal{G} + \mathcal{G}^\dagger)$ is a Heisenberg Hamiltonian which has the eigenstate $|\,\Omega_M\,\rangle$ with eigenvalue $|\Gamma|s^2$.

For spin rings and $a = 1$, $|\,\varphi\,\rangle$ is nothing else but the relative ground state. Generally for spin rings, $|\,\varphi\,\rangle$ has the same shift quantum number as the relative ground state [9].

2.3 Analytical expression for the upper bound

The best bound for $E_{\min}(M)$ is obtained if $\cos\alpha_\ell$ in (20) is as low as possible, *i.e.* $\ell = \frac{n}{2}$ and $\cos\alpha_\ell = -1$ for even n and $\ell = \frac{n\pm1}{2}$ for odd n. Therefore the upper bound is given by

$$E_{\min}(M) \leq c|\Gamma|s^2 + (1 - \frac{c}{\delta})\frac{\delta|\Gamma|}{N}\left(Ns^2 - \frac{2sa(2Ns-a)}{2Ns-1}\right)\ , \tag{21}$$

where $c = -1$ in the case of even n and $c = -\cos\frac{\pi}{n}$ for odd n. Let $\delta = 1$ and Ns be integer. Then the total ground state lies in the sector $M = 0$. In this case we obtain

$$E_{\min}(0) \leq c|\Gamma|s^2 + \frac{(-1+c)|\Gamma|s^2}{2Ns-1}\ , \tag{22}$$

which improves the upper Berezin-Lieb bound $E_{\min}^{\text{classical}}s^2$, see [4,5], if $c|\Gamma| = E_{\min}^{\text{classical}}$.

3 Lower bounds

3.1 Idea

For the lower bound to be derived in the following section the interaction matrix $\mathbb{J} \equiv (J_{\mu\nu})$ describing the coupling between spins at sites μ and ν must have certain homogeneity properties. The matrix must be symmetric and must have constant row sum. This alone is sufficient to derive some lower bounds [6], which can be improved using the topological structure of the interactions, as will be shown in the following.

The derivation works by constructing another "averaged" Hamiltonian having an analytically diagonalizable interaction matrix $\widetilde{\mathbb{J}}$, which nevertheless has only eigenvalues already present for the original interaction matrix $\mathbb{J}$. Since, by construction, $\mathbb{J} \geq \widetilde{\mathbb{J}}$, this relation also holds for the related Hamiltonians, and we arrive at a lower bound. Extending this idea the obtained lower bounds could be improved for particular systems. However, in this article we will confine ourselves to deriving bounds for general classes of systems.

Using the topological structure of the interactions will further enable us to determine the degeneracy of some eigenvalues of $\mathbb{J}$ and therefore improve the calculations of reference [6] where this information was not exploited.

3.2 Definition of n-homogeneity

The Hamiltonian used in this section is different from that of the previous section and assumed to be of the form

$$H = \sum_{\mu\nu} J_{\mu\nu} \boldsymbol{s}_\mu \cdot \boldsymbol{s}_\nu \ . \tag{23}$$

The matrix $\mathbb{J}$ of coupling constants $J_{\mu\nu}$ is assumed to be symmetric and having constant row sums j. The latter property can be viewed as a kind of gauge condition, since adding a diagonal matrix with vanishing trace to $\mathbb{J}$ does not change the Hamiltonian (23), see reference [10].

Being symmetrical, $\mathbb{J}$ has a complete set of (ordered) eigenvalues $j_1, \ldots, j_N$. One of them is the row sum j with $\mathbf{1} \equiv \frac{1}{\sqrt{N}}(1, 1, \ldots, 1)$ as the corresponding eigenvector. Let $\mathbb{J}'$ denote the matrix $\mathbb{J}$ restricted to the subspace orthogonal to $\mathbf{1}$, and $j_{\min}$ the smallest eigenvalue of $\mathbb{J}'$. $j_{\min}$ may be m-fold degenerate. Further, we will denote the αth normalized eigenvector of $\mathbb{J}$ by $(c_{1\alpha}, \ldots, c_{N\alpha})$, *i.e.*

$$\sum_\nu J_{\mu\nu} c_{\nu\alpha} = j_\alpha c_{\mu\alpha} \ , \ \sum_\mu \overline{c_{\mu\alpha}} c_{\mu\beta} = \delta_{\alpha\beta} \qquad \alpha, \beta, \mu = 1, \ldots, N \ , \tag{24}$$

where we also allow for the possibility to choose complex eigenvectors. Sums over $\alpha = 1, \ldots, N$ excluding α_j will be denoted by $\sum'$, where α_j denotes the index (within the ordered set of all eigenvalues) of the eigenvalue j belonging to the eigenvector $\mathbf{1}$.

For later use we will consider a transformation of the spin observables analogous to the transformation onto the eigenbasis of $\mathbb{J}$ and define

$$\boldsymbol{T}_\alpha \equiv \sum_\mu \overline{c_{\mu\alpha}} \boldsymbol{s}_\mu, \text{ and } Q_\alpha \equiv \boldsymbol{T}_\alpha^\dagger \cdot \boldsymbol{T}_\alpha \ , \qquad \alpha = 1, \ldots, N. \tag{25}$$

The inverse transformation then yields

$$\boldsymbol{s}_\mu = \sum_\alpha c_{\mu\alpha} \boldsymbol{T}_\alpha, \quad \mu = 1, \ldots, N \ . \tag{26}$$

In particular, $\boldsymbol{T}_{\alpha_j} = \boldsymbol{S}/\sqrt{N}$. It then follows directly from the definitions that

$$Ns(s+1) = \sum_\mu (\boldsymbol{s}_\mu)^2 = \sum_\alpha Q_\alpha = \frac{1}{N}\boldsymbol{S}^2 + \sum_\alpha{}' \, Q_\alpha, \tag{27}$$

$$H = \sum_{\mu\nu\alpha\beta} J_{\mu\nu} \overline{c_{\mu\alpha}} c_{\nu\beta} \boldsymbol{T}_\alpha^\dagger \cdot \boldsymbol{T}_\beta = \sum_\beta j_\beta Q_\beta = \frac{j}{N}\boldsymbol{S}^2 + \sum_\beta{}' \, j_\beta Q_\beta. \tag{28}$$

For a later use we also need a relation between Hamiltonians with different coupling matrices. Therefore, let H and $\widetilde{H}$ be two Hamiltonians of the form (23) with coupling matrices $\mathbb{J}$ and $\widetilde{\mathbb{J}}$, such that $\mathbb{J} \geq \widetilde{\mathbb{J}}$. Then $H \geq \widetilde{H}$. As usual the ordering "$\geq$" of operators is defined by the corresponding inequality for arbitrary expectation values. Since H depends linearly on $\mathbb{J}$ it suffices to show that $\mathbb{J} \geq 0$ implies $H \geq 0$. But this is obvious in view of (28): $H = \sum_\beta j_\beta Q_\beta$ with $j_\beta \geq 0$ and $Q_\beta \geq 0$.

Next we turn to the suitable definition of n-homogeneity. Let the set of spin sites $\{1, \ldots, N\}$ be divided into n disjoint subsets of equal size m, $\{1, \ldots, N\} = \bigcup_{\nu=1}^n \mathcal{A}_\nu$, such that the coupling constants within each $\mathcal{A}_\nu$ are ≤ 0, but ≥ 0 between $\mathcal{A}_\nu$ and $\mathcal{A}_\mu$ for $\nu \neq \mu$. Moreover, the partial row sums are assumed to be constant:

$$\sum_{b \in \mathcal{A}_\mu} J_{ab} = \begin{cases} j^{\text{in}} \text{ if } a \in \mathcal{A}_\mu \\ j^{\text{ex}} \text{ if } a \notin \mathcal{A}_\mu \end{cases} \ . \tag{29}$$

A spin system satisfying the assumptions of this section will be called n-homogeneous, see [1–3]. Note that this notion is incommensurable to n-cyclicity defined in the previous section. However, certain rings, the triangular lattice, the kagomé lattice, and the icosidodecahedron satisfy both definitions. A necessary condition for nearest neighbor Heisenberg systems to be n-homogeneous is that the number of nearest neighbors, which is assumed to be constant, is divisible by $(n-1)$. Actually, spin rings of even N are 2-homogeneous, rings of odd N are 3-homogeneous if N is divisible by 3. n-homogeneous Heisenberg rings do not exist for $n > 3$ because they do not fulfill the homogeneity condition (29).

We recall that $\mathbf{1} = \frac{1}{\sqrt{N}}(1, 1, \ldots, 1)$ is an eigenvector of $\mathbb{J}$ with eigenvalue j. Due to n-homogeneity there are, after

a suitable permutation of the spin sites, further eigenvectors of the form

$$u^{(k)} = (m:1, m:\rho^k, m:\rho^{2k}, \ldots, m:\rho^{(n-1)k}), \quad k = 1, \ldots, n-1, \quad (30)$$

where $(m : x, \ldots)$ denotes the m-fold repetition of the entry x, and $\rho \equiv \mathrm{e}^{2\pi \mathrm{i}/n}$. The corresponding eigenvalues are $j_k = j^{\mathrm{in}} + j^{\mathrm{ex}} \sum_{p=1}^{n-1} \rho^{pk} = j^{\mathrm{in}} - j^{\mathrm{ex}}$, hence they coalesce into one $(n-1)$-fold degenerate eigenvalue. By applying the theorem of Geršgorin (*cf.* [11], 7.2) this eigenvalue is shown to be the smallest one $j_{\min}$.

Next we construct a coupling matrix $\widetilde{\mathbb{J}}$ with the same eigenspaces as $\mathbb{J}$ but only three different eigenvalues. It has the block structure

$$\widetilde{\mathbb{J}} = \begin{pmatrix} A\ C\ C \ldots \\ C\ A\ C \ldots \\ C\ C\ A \ldots \\ \vdots\ \vdots\ \vdots\ \vdots \end{pmatrix} , \quad (31)$$

where A and C are $m \times m$-matrices of the form

$$A = \begin{pmatrix} \beta & -\alpha & -\alpha \ldots \\ -\alpha & \beta & -\alpha \ldots \\ -\alpha & -\alpha & \beta \ \ldots \\ \vdots & \vdots & \vdots \ \vdots \end{pmatrix} , \quad C = \begin{pmatrix} \gamma\ \gamma\ \gamma \ldots \\ \gamma\ \gamma\ \gamma \ldots \\ \gamma\ \gamma\ \gamma \ldots \\ \vdots\ \vdots\ \vdots\ \vdots \end{pmatrix} . \quad (32)$$

The three eigenvalues of $\widetilde{\mathbb{J}}$ are

$$\tilde{j} = \beta - (m-1)\alpha + (N-m)\gamma, \quad (33)$$

$$\tilde{j}_{\min} = \beta - (m-1)\alpha - m\gamma, \quad (34)$$

$$\tilde{j}_2 = \alpha + \beta \quad (35)$$

with degeneracies $1, n-1$ and $N-n$, resp. By choosing

$$\alpha = \frac{1}{N}(nj_2 - j - (n-1)j_{\min}), \quad (36)$$

$$\beta = \frac{1}{N}((N-n)j_2 + j + (n-1)j_{\min}), \quad (37)$$

$$\gamma = \frac{j - j_{\min}}{N} , \quad (38)$$

one obtains

$$\tilde{j} = j,\ \tilde{j}_{\min} = j_{\min},\ \tilde{j}_2 = j_2 . \quad (39)$$

j_2 is the remaining smallest eigenvalue of $\mathbb{J}'$ after eliminating $(n-1)$-times $j_{\min}$ from the set of eigenvalues. Thus it can happen that $j_2 = j_{\min}$ if $j_{\min}$ is more than $(n-1)$-fold degenerate.

Let us write $\boldsymbol{S}_{\mathcal{A}} \equiv \sum_{a \in \mathcal{A}} \boldsymbol{s}_a$ for any subset $\mathcal{A} \subset \{1, \ldots, N\}$. We conclude

$$H \geq \widetilde{H} = -\alpha \left(\sum_{\nu} \boldsymbol{S}^2_{\mathcal{A}_\nu} \right) + (\alpha + \beta) N s(s+1) + \gamma \left(\boldsymbol{S}^2 - \sum_{\nu} \boldsymbol{S}^2_{\mathcal{A}_\nu} \right) \quad (40)$$

$$\geq \gamma S(S+1) - (\alpha + \gamma) n \frac{N}{n} s(\frac{N}{n} s + 1) + (\alpha + \beta) N s(s+1). \quad (41)$$

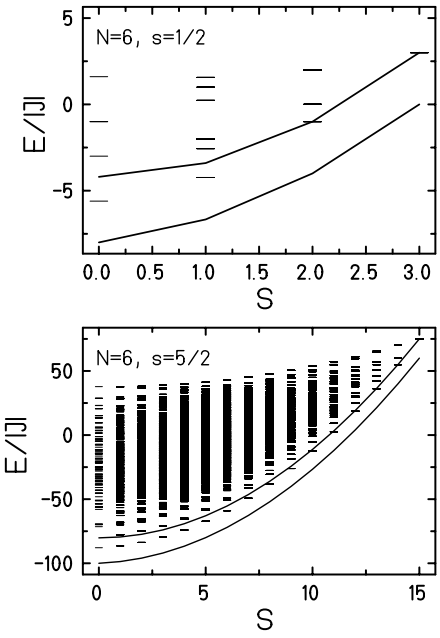

Fig. 2. Upper and lower bounds of $E_{\min}(S)$ for Heisenberg spin rings with $N = 6$ and $s = 1/2$ (top) as well as $s = 5/2$ (bottom). The solid curves display the bounds for the minimal energies considering 2-cyclicity ($s = 1/2 : \epsilon = 0.16; s = 5/2 : \epsilon = 0.05$) and 2-homogeneity ($s = 1/2 : \epsilon = 0.28; s = 5/2 : \epsilon = 0.07$).

3.3 Analytical expression for the lower bound

Hence we obtain for the lower bound

$$E \geq \frac{j - j_{\min}}{N} S(S+1) + N j_{\min} s(s+1) + (N-n)(j_2 - j_{\min}) s . \quad (42)$$

Since $j_2 - j_{\min} \geq 0$ the bound (42) is the better, the smaller n is. This is in contrast to the upper bound considered in the previous section, which is improved for large odd n.

4 Examples

In the following examples we calculate the energy eigenvalues by numerical methods as well as lower and upper bounds. All examples are Heisenberg spin systems where the total spin S is a good quantum number. It turns out that $S \mapsto E_{\min}(S)$ is always a monotonically increasing function, hence we need not to distinguish between $E_{\min}(S)$ and $E_{\min}(M)$.

In order to judge the quality of the bounds we provide the deviation of the best upper and lower bound from the exact ground state energy in relation to the energy difference between antiferromagnetic and ferromagnetic ground state, *i.e.*

$$\epsilon = \frac{|E_{\mathrm{bound},0} - E_0|}{E(Ns) - E_0} . \quad (43)$$

The first example we would like to consider is a Heisenberg spin ring with $N = 6$ and $s = 1/2$ as well as $s = 5/2$. Figure 2 shows the numerically determined energy eigenvalues (dashes) as a function of total spin S. The solid

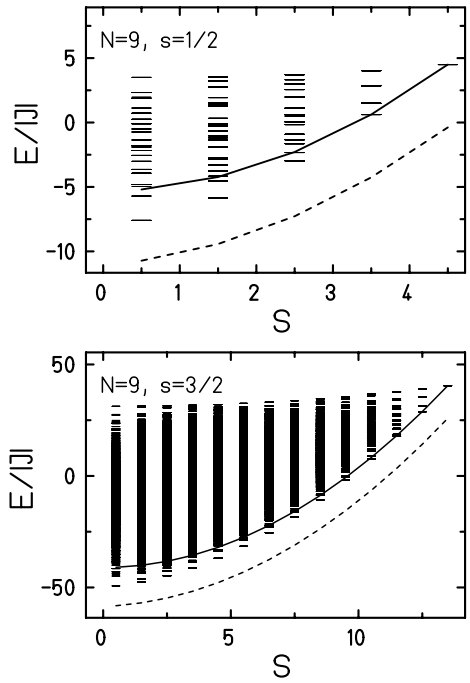

Fig. 3. Upper and lower bounds of $E_{\min}(S)$ for Heisenberg spin rings with $N = 9$ and $s = 1/2$ (top) as well as $s = 3/2$ (bottom). The solid curves display the upper bounds for the minimal energies considering 9-cyclicity ($s = 1/2 : \epsilon = 0.20; s = 3/2 : \epsilon = 0.09$), the dashed curves do the same for 3-homogeneity ($s = 1/2 : \epsilon = 0.26; s = 3/2 : \epsilon = 0.10$).

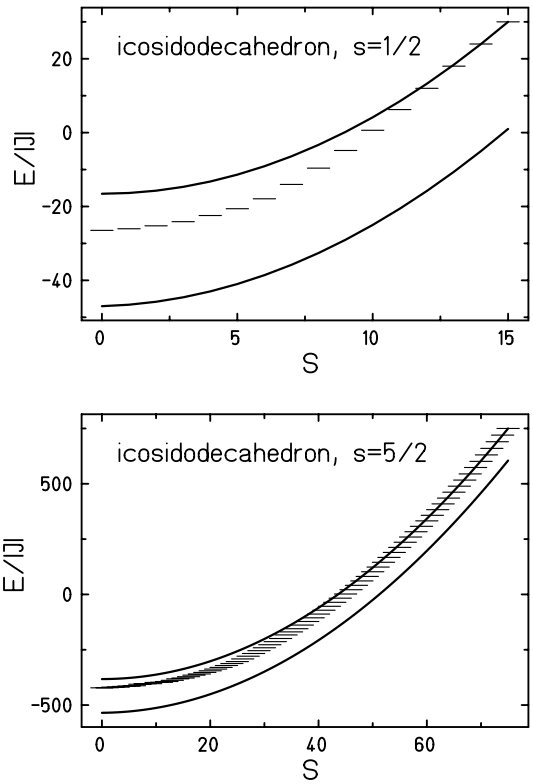

Fig. 4. Upper and lower bounds of $E_{\min}(S)$ for Heisenberg spin systems with icosidodecahedral structure, *i.e.* $N = 30$ and $s = 1/2$ (top) as well as $s = 5/2$ (bottom). The solid curves display the bounds for the minimal energies considering 3-cyclicity ($s = 1/2 : \epsilon = 0.18; s = 5/2 : \epsilon = 0.03$) and 3-homogeneity ($s = 1/2 : \epsilon = 0.36; s = 5/2 : \epsilon = 0.10$).

curves display the bounds for the minimal energies considering 2-cyclicity and 2-homogeneity.

As a second example we take a frustrated Heisenberg ring with $N = 9$ and $s = 1/2$ as well as $s = 3/2$. The results are presented in Figure 3. The solid curves display the upper bounds for the minimal energies considering 9-cyclicity, the dashed curves do the same for 3-homogeneity. Without using the concept of n-homogeneity the lower bounds are much poorer for frustrated systems [6].

Another example, an icosidodecahedral Heisenberg spin system, is related to magnetic molecules, which can be synthesized in such structures. One species is given by $\{Mo_{72}Fe_{30}\}$, a molecule where 30 Fe^{3+} paramagnetic ions ($s = 5/2$) occupy the sites of a perfect icosidodecahedron [12] and interact *via* isotropic nearest-neighbor antiferromagnetic Heisenberg exchange [13]. Not much is known about the spectrum of such giant structures since the Hilbert space assumes a very large dimension of $6^{30} \approx 10^{23}$. So far only DMRG calculations could approximate the minimal energies [14].

Figure 4 shows as dashes on the l.h.s. the minimal energies for $s = 1/2$ which are determined numerically by J. Richter with a Lánczos method [15,16] and on the r.h.s. the minimal DMRG energies [14]. The icosidodecahedral Heisenberg spin system is 3-cyclic as well as 3-homogeneous. The corresponding bounds are displayed by solid curves. Especially the upper bound for the case of $s = 5/2$ is very close to the "true" (DMRG) minimal energies and thus could be used to justify approximations of the low-lying spectrum as used in reference [17]. The lower bounds are worse than expected, but this behavior is explained by the 10-fold degeneracy of $j_{\min}$, therefore $j_2 = j_{\min}$, and the last term in (42) yields zero, unfortunately.

The last example discusses the triangular spin lattice which is one of the frustrated two-dimensional spin systems. The triangular spin lattice is 3-homogeneous and 3-cyclic, if the periodic boundary conditions are suitably chosen. Figure 5 displays the energy levels for $N = 12$ and $s = 1/2$ (l.h.s.) as well as $s = 1$ (r.h.s.). The bounds of $E_{\min}(S)$ are given by solid curves. In both cases the upper bound is very close to the exact minimal energies.

For the thermodynamic limit $N \to \infty$ of the triangular lattice with $\delta = 1$ we rewrite the bounds by introducing a continuous spin variable $S_c = S/N$ running from 0 to s and using $j = 6$, $j_{\min} = -3$ with twofold degeneracy, and $\lim_{N\to\infty} j_2 = -3$. After dividing by N the resulting bounds are separated only by $3s$:

$$9S_c^2 - 3s^2 - 3s \leq \lim_{N\to\infty} \frac{E_{\min}(S)}{N} \leq 9S_c^2 - 3s^2 \ . \quad (44)$$

We thank Johannes Richter for fruitful discussions.

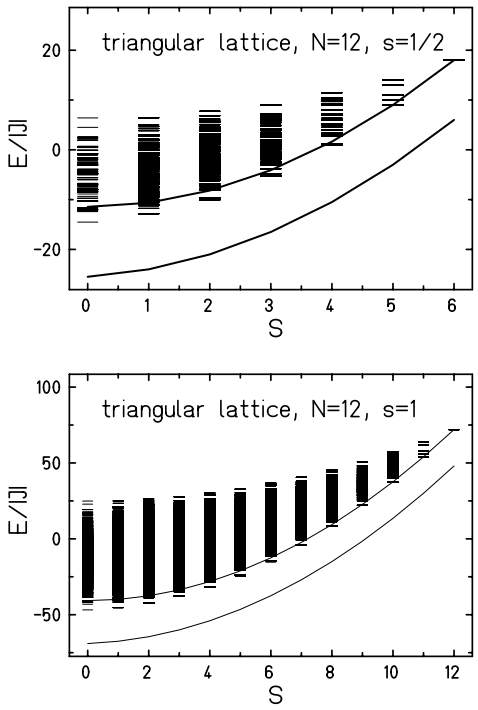

Fig. 5. Upper and lower bounds of $E_{\min}(S)$ for the triangular spin lattice with $N = 12$ and $s = 1/2$ (top) as well as $s = 1$ (bottom). The solid curves display the bounds for the minimal energies considering 3-cyclicity ($s = 1/2 : \epsilon = 0.09; s = 1 : \epsilon = 0.05$) and 3-homogeneity ($s = 1/2 : \epsilon = 0.34; s = 1 : \epsilon = 0.19$).

Appendix A: Proof of equation (16)

For easy readability we repeat equation (16)

$$\langle\Omega_M|\Delta|\Omega_M\rangle = \frac{\delta|\Gamma|}{N}\left\{Ns^2 - \frac{2sa(2Ns-a)}{2Ns-1}\right\} . \quad (45)$$

Since Ω_M is invariant w. r. t. arbitrary permutations of spin sites it suffices to choose $\Delta = s_1^{(3)}s_2^{(3)}$ and to multiply the result for $\langle\Omega_M|\Delta|\Omega_M\rangle$ by $\delta|\Gamma|$. We note that

$$\left[\Delta, S^-\right] = -(s_1^{(3)}s_2^- + s_1^-s_2^{(3)}), \quad (46)$$

and

$$\left[\left[\Delta, S^-\right], S^-\right] = 2s_1^-s_2^- , \quad (47)$$

but higher commutators vanish. Hence

$$\left[\Delta, (S^-)^a\right] = a(S^-)^{a-1}\left[\Delta, S^-\right] + \binom{a}{2}(S^-)^{a-2}\left[\left[\Delta, S^-\right], S^-\right] . \quad (48)$$

Further we define $\lambda(a,k)$ by

$$(S^+)^a(S^-)^a(S^-)^k \,|\, \Omega\,\rangle = \lambda(a,k)(S^-)^k \,|\, \Omega\,\rangle . \quad (49)$$

Using $S^+S^- = S^2 - S^{(3)}(S^{(3)}-1)$ one derives the recursion relation

$$\lambda(a+1,k) = (2Ns-a-k)(a+k+1)\lambda(a,k)\cdot \quad (50)$$

Together with $\lambda(0,k) = 1$ it can be solved and yields

$$\lambda(a,k) = \frac{(2Ns-k)!}{(2Ns-a-k)!}\frac{(a+k)!}{k!} . \quad (51)$$

Obviously,

$$C_M^2 = \lambda(a,0)^{-1} = \frac{(2Ns-a)!}{(2Ns)!\;a!} , \quad (52)$$

hence

$$C_M^2\lambda(a-1,1) = \frac{(2Ns-a)!}{(2Ns)!\;a!}\frac{(2Ns-1)!\;a!}{(2Ns-a)!\;1!} = \frac{1}{2Ns} , \quad (53)$$

and

$$C_M^2\lambda(a-2,2) = \frac{(2Ns-a)!}{(2Ns)!\;a!}\frac{(2Ns-2)!\;a!}{(2Ns-a)!\;2!} = \frac{1}{4Ns(2Ns-1)}. \quad (54)$$

Now we are prepared to calculate $\langle\Omega_M|\Delta|\Omega_M\rangle$:

$$\langle\Omega_M|\Delta|\Omega_M\rangle = C_M^2\langle\Omega_M|(S^-)^a\Delta + \left[\Delta,(S^-)^a\right]|\Omega\rangle \quad (55)$$

$$= s^2 + aC_M^2\langle\Omega_M|(S^-)^{a-1}\left[\Delta,S^-\right]|\Omega\rangle \quad (56)$$

$$+ \binom{a}{2}C_M^2\langle\Omega_M|(S^-)^{a-2}\left[\left[\Delta,S^-\right],S^-\right]|\Omega\rangle$$

$$= s^2 + aC_M^2\langle(S^+)^{a-1}(S^-)^{a-1}S^-\Omega|\left[\Delta,S^-\right]|\Omega\rangle \quad (57)$$

$$+ \binom{a}{2}C_M^2\langle(S^+)^{a-2}(S^-)^{a-2}(S^-)^2\Omega|\left[\left[\Delta,S^-\right],S^-\right]|\Omega\rangle$$

$$= s^2 + aC_M^2\lambda(a-1,1)\langle S^-\Omega|\left[\Delta,S^-\right]|\Omega\rangle \quad (58)$$

$$+ \binom{a}{2}C_M^2\lambda(a-2,2)\langle(S^-)^2\Omega|\left[\left[\Delta,S^-\right],S^-\right]|\Omega\rangle$$

$$= s^2 + \frac{a}{2Ns}(-4s^2) + \binom{a}{2}\frac{1}{4Ns(2Ns-1)}16s^2 \quad (59)$$

$$= \frac{1}{N}\left\{Ns^2 - \frac{2sa(2Ns-a)}{2Ns-1}\right\}. \quad (60)$$

In line (56) we used (48). (59) is obtained by means of (53, 54) and the identities

$$\langle S^-\Omega|\left[\Delta,S^-\right]|\Omega\rangle = \langle S^-\Omega| - (s_1^{(3)}s_2^- + s_1^-s_2^{(3)})|\Omega\rangle \quad (61)$$

$$= -2\langle s_2^-\Omega|s_1^{(3)}s_2^-|\Omega\rangle = -2s(s(s+1)-s(s-1))$$

$$= -4s^2 ,$$

and

$$\langle(S^-)^2\Omega|\left[\left[\Delta,S^-\right],S^-\right]|\Omega\rangle = \langle(S^-)^2\Omega|2s_1^-s_2^-|\Omega\rangle \quad (62)$$

$$= 4\langle s_1^-s_2^-\Omega|s_1^-s_2^-|\Omega\rangle$$

$$= 4(s(s+1)-s(s-1))^2$$

$$= 16s^2 .$$

This completes the proof.

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