Ground state properties of antiferromagnetic
Heisenberg spin rings

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Abstract

Exact ground state properties of antiferromagnetic Heisenberg spin rings with isotropic next neighbour interaction are presented for various numbers of spin sites and spin quantum numbers. Earlier work by Peierls, Marshall, Lieb, Schultz and Mattis focused on bipartite lattices and is not applicable to rings with an odd number of spins. With the help of exact diagonalization methods we find a more general systematic behaviour which for instance relates the number of spin sites and the individual spin quantum numbers to the degeneracy of the ground state. These numerical findings all comply with rigorous proofs in the cases where a general analysis could be carried out. Therefore it can be plausibly conjectured that the ascertained properties hold for ground states of arbitrary antiferromagnetic Heisenberg spin rings. These general rules help to explain the low temperature behaviour of recently synthesized spin rings.

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I. INTRODUCTION AND SUMMARY

Synthesized molecules containing relatively small numbers of paramagnetic ions and their magnetic properties are of great current interest\textsuperscript{1-4}. Some of them appear as rings of localized single-particle magnetic moments which are adequately described by the Heisenberg model\textsuperscript{5-9} with – in many cases – isotropic antiferromagnetic coupling.

In this article properties of the ground state for antiferromagnetically (AF) coupled spin rings will be presented. Exact diagonalization methods\textsuperscript{5,10-13} make it possible to investigate small spin rings for various numbers $N$ of spin sites and spin quantum numbers $s$. The obvious symmetries allow to decompose the Hilbert space $\mathcal{H}$ into a set of mutually orthogonal subspaces $\mathcal{H}(S,M,k)$ according to the quantum numbers of the total spin $S$, the total magnetic quantum number $M$ and the shift quantum number $k$ of the cyclic shift operator.

In view of our exact numerical results (table I), several conjectures on general properties of the antiferromagnetic ground state suggest themselves. The two most important ones are:

- If $N \cdot s$ is integer, then the ground state is non-degenerate.
• If $N \cdot s$ is half integer, then the ground state is fourfold degenerate.

These findings exceed those derived from the theorem of Lieb, Schultz and Mattis\textsuperscript{14,15} and establish rules also for spin rings of an odd number of spin sites. Though, for the time being, rigorous proofs or refutations of our statements in the undecided cases remain a challenge, the numerical results and partial proofs so far provide a strong evidence of general validity.

The presented general ground state properties are very helpful in order to understand the low temperature behaviour of small spin rings, since only a few states dominate the properties at low temperature because of the finite system size. In addition the found general rules allow statements about spin rings of larger size whose Hamilton operator cannot be diagonalized any longer. As an example the behaviour of the zero-field susceptibility is discussed. Our results also open a new view on frustration as they show that there are systems which one would like to call frustrated, but which do not possess the typical ground state degeneracy\textsuperscript{16}.

II. OBSERVATIONS

The Hamilton operator of the Heisenberg model with antiferromagnetic, isotropic next-neighbor interaction between spins of equal spin quantum number $s$ reads

$$H = -2J \sum_{x=1}^{N} \vec{s}(x) \cdot \vec{s}(x+1), \quad \forall x : s(x) = s, \quad J < 0,$$  \hspace{1cm} (1)

the spin sites being enumerated by $x$ modulo $N$. This Hamilton operator commutes with the total spin $\vec{S}$ and its three-component $S^3$. In addition $H$ is invariant under cyclic shifts. These are represented by a unitary cyclic shift operator $T_x$ defined by its action on the product basis (3)

$$T_x \left| m_1, \ldots, m_{N-1}, m_N \right> = \left| m_N, m_1, \ldots, m_{N-1} \right>,$$  \hspace{1cm} (2)

where the product basis of single-particle eigenstates of all $\vec{s}(x)$ obeys

$$\vec{s}(x) \left| m_1, \ldots, m_x, \ldots, m_N \right> = m_x \left| m_1, \ldots, m_x, \ldots, m_N \right>.$$  \hspace{1cm} (3)

For a shorter notation the basis elements are sometimes abbreviated as

$$\left| m \right> := \left| m_1, \ldots, m_x, \ldots, m_N \right>.$$  \hspace{1cm} (4)

The eigenvalues of $T_x$ are the $N$-th roots of unity,

$$z = \exp \left\{ -\frac{2\pi i k}{N} \right\},$$  \hspace{1cm} (5)

where $k$ will be called shift quantum number, and $k$ can assume values $k = 0, \ldots, N - 1$ modulo $N$. For every eigenstate of $H$ and $T$ with $k \neq 0$ and $k \neq N/2$ there exists another eigenstate with translational quantum number $N-k$ with the same energy eigenvalue, which
is just the result of invariance under complex conjugation. For later use we define a “cycle” as the linear span of all product states which result from a given product state by multiple repetition of the cyclic shift operator $T^{12}$.

The Hamilton operator remains also invariant under spin flips provided by the operator $C$, which turns all $m_x$ into $-m_x$. In addition $C$ commutes with $S^2$, but anti-commutes with $S^3$. The eigenvalues of $C$ are $\pi = \pm 1$.

Exact diagonalization methods$^{12}$ enable us to evaluate eigenvalues and eigenvectors of $H$ for small spin rings of various numbers $N$ of spin sites and spin quantum numbers $s$. Our results for ground state properties are summarized in table 1. For spin $s = 1/2$ they are consistent with Ref.$^{17}$ and for spin $s = 1$ and even $N$ with Ref.$^{18}$. Without exception we find:

1. The ground state belongs to the subspace $H(S)$ with the smallest possible total spin quantum number $S$; this is either $S = 0$ for $N \cdot s$ integer, then the total magnetic quantum number $M$ is also zero, or $S = 1/2$ for $N \cdot s$ half integer, then $M = \pm 1/2$.

2. The restricted ground state within a subspace of constant total magnetic quantum number $M$ belongs to $H(S)$ with $S$ attaining its smallest value $S = |M|$.

3. If $N \cdot s$ is integer, then the ground state is non-degenerate.

4. If $N \cdot s$ is half integer, then the ground state is fourfold degenerate.

5. If $s$ is integer or $N \cdot s$ even, then the shift quantum number is $k = 0$.

6. If $s$ is half integer and $N \cdot s$ odd, then the shift quantum number turns out to be $k = N/2$.

7. If $N \cdot s$ is half integer, then $k = [(N + 1)/4]$ and $k = N - [(N + 1)/4]$ is found. $[(N + 1)/4]$ symbolizes the greatest integer less or equal to $(N + 1)/4$.

8. Non-degenerate ground states are also eigenstates of the spin flip operator $C$. For half integer $s$ and necessarily even $N$ we find that $\pi = -1$ if $N/2$ is odd and $\pi = +1$ if $N/2$ is even. The situation is more complicated for integer $s$. Here rows of alternating $\pi$ for odd $s$ change with rows of $\pi = -1$ for even $s$. This behaviour was also checked for $s = 3$ and $s = 4$, but not displayed in the table.

9. Non-degenerate ground states with $k \neq 0$ or $\pi \neq -1$ overlap with all product states of the Hilbert subspace with $M = 0$. For ground states with $k = 0$ and $\pi = -1$ those product states, which remain in the same cycle after application of the spin flip, have coefficients zero.

III. PROOFS AND SUGGESTIONS

Although some properties, especially the first one, appear natural, we cannot prove all of them rigorously and generally, but we can prove special cases and provide some general arguments:
• For $N \leq 4$ one can evaluate the eigenvalues and eigenstates of (1) analytically, because the Hamilton operator can be drastically simplified

\[
N = 2 : \mathcal{H} = -2J \left( \tilde{s}_z^2 - \tilde{s}_1^2 - \tilde{s}_2^2 \right) , \tag{6}
\]

\[
N = 3 : \mathcal{H} = -J \left( \tilde{s}_z^2 - \tilde{s}_1^2 - \tilde{s}_2^2 - \tilde{s}_3^2 \right) , \tag{7}
\]

\[
N = 4 : \mathcal{H} = -J \left( \tilde{s}_z^2 - \tilde{s}_1^2 - \tilde{s}_2^2 - \tilde{s}_{13}^2 - \tilde{s}_{24}^2 \right) , \quad \tilde{s}_{13} = \tilde{s}(1) + \tilde{s}(3) , \quad \tilde{s}_{24} = \tilde{s}(2) + \tilde{s}(4) . \tag{8}
\]

One realizes that the energy eigenvalues depend monotonically on the total spin quantum number $S^{5,19}$, so that the AF ground state must belong to the minimal total spin $S = S_{\text{min}}$ which is either 0 or 1/2 with the respective magnetic quantum numbers $M$.

• For $N \leq 4$ one can in addition explain the degeneracy of the ground state exactly using angular momentum coupling. It is obvious for $N = 2$ that one can couple the two spins to a total spin running from $S = 0$ to $S = 2s$ with the Hilbert space of $S = 0$ having dimension one. For $N = 3$ one can couple the first two spins to integers $S = 0, \ldots , 2s$. Then for $s$ being half integer there are two possibilities to couple to $S = 1/2$ which results in a fourfold degenerate ground state. If $s$ is integer, one is left with only one possibility to couple to zero giving a non-degenerate ground state. The situation is more involved for $N = 4$. Here one needs to consider that all spin operators $\tilde{s}_z, \tilde{s}_{13}$ and $\tilde{s}_{24}$ commute, therefore the lowest energy is only given by the coupling where $S = 0$ and $S_{13} = S_{24} = 2s$, which results in a non-degenerate ground state.

• Non-degenerate eigenstates of $\mathcal{H}$ remain invariant under all unitary transformations $U$ that commute with $\mathcal{H}$. In the case of rotational symmetry this implies that the eigenstate spans a one-dimensional irreducible representation of the rotation group, hence belongs to $S = 0$. In the case of the $k \leftrightarrow N - k$ symmetry one concludes by an analogous argument that $k = 0$ or $k = N/2$ (only if $N$ even). Because of the spin flip symmetry non-degenerate ground states are also eigenstates of $\mathcal{C}$.

• Using results of Peierls and Marshall$^{20}$ and Lieb, Schultz and Mattis$^{14,15}$ one can moreover prove that for even numbers $N$ of spin sites the ground state is non-degenerate and therefore must have $S = 0$. The proof rests on the fact that the coefficients $c(m)$ of the ground state $| \Psi_0 \rangle$ with respect to the product basis

\[
| \Psi_0 \rangle = \sum_m c(m) \, | m \rangle , \tag{9}
\]

with $\sum_{i=1}^{N} m_i = 0$ for the ground state, possess a specific sign property, namely

\[
c(m) = (-1)^{\left( \frac{N}{2} - \sum_{i=1}^{N} m_i \right)} a(m) \tag{10}
\]

with all $a(m)$ proven to be non-zero, real and of equal sign. Expressing the ground state energy $\langle \Psi_0 | \mathcal{H} | \Psi_0 \rangle$ in terms of the coefficients $a(m_1, m_2, \ldots , m_N)$ one also realizes
that only one set of coefficients can minimize the energy because having two sets one could create a new one as the set of differences which should belong to the same eigenspace but violates (10).

The sketched property and its proof hold more generally in any Hilbert subspace with constant \( M \). For rings of an odd number of spins a sign rule cannot be established because the Hamilton operator connects basis states \( |m\rangle \) such, that one returns to the starting state after an odd number of steps. The simplest example for \( N = 3 \), \( s = 1/2 \) reads: \( |++-\rangle \rightarrow |+-+\rangle \rightarrow |--+\rangle \rightarrow |++-\rangle \).

- **A useful byproduct of the sign rule (10)** is that it also explains the sequence of \( k \)-values for even \( N \). Consider the action of the cyclic shift operator on the basis states (2).
  
  The change in sign of the coefficient, whose absolute value is not altered, then is
  
  \[
  \frac{c(m_1, \ldots, m_{N-1}, m_N)}{c(m_N, m_1, \ldots, m_{N-1})} = (-1)^{\left(\sum_{i=1}^{N/2} m_{2i} - \sum_{i=1}^{N/2} m_{2i-1}\right)} = (-1)^{Ns}.
  \]

  Thus for odd \( N\cdot s \) we find \( k = N/2 \), whereas even \( N\cdot s \) implies \( k = 0 \).

- **For even \( N \)** the ground state has non-zero components with respect to product states of the form \( |m, -m, m, -m, \ldots\rangle \), \( m \neq 0 \). This explains the correlation between \( k \) and \( \pi \), namely \( k = 0 \leftrightarrow \pi = + \) and \( k = N/2 \leftrightarrow \pi = - \), because cyclic shift and spin flip operator have the same effect on these product states. One may also observe that an alternating product state \( |m, -m, m, -m, \ldots\rangle \) generates a cycle of dimension 2 (cf.\(^{12}\)) which in turn implies the selection rule \( k = 0 \) or \( k = N/2 \).

- **If \( N\cdot s \) is half integer**, then the ground state, which has a finite total magnetic quantum number \( M \), must be at least twofold degenerate because of the spin flip symmetry \( \forall x: m_x \rightarrow -m_x \), i.e. \( M \rightarrow -M \). If one could show, that \( k \) could not be zero, one would explain the fourfold degeneracy. It is, however, not possible to derive the fourfold degeneracy for such cases using only symmetry arguments since the spectrum contains also twofold degenerate energy eigenvalues.

- **For \( s = 1/2 \)** the fourfold degeneracy and the special \( k \)-values can be explained using the Bethe ansatz for odd \( N^{21} \).

At the end of this section we would like to suggest an explanation for the special \( k \)-values found for half integer \( N \cdot s \). It is sufficient to restrict the investigations to the Hilbert subspace \( \mathcal{H}(M = 1/2) \). Our arguments rest on the observation that the cycle generated by the special basis state

\[
|m_0\rangle := \left| \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \cdots \right\rangle = |++--++--\cdots\rangle,
\]

which is maximally alternating but minimally undulating, contributes to the ground state. For \( s = 1/2 \) this seems to be rather natural, because this special cycle constitutes the ground state of the Ising model \(((13), \gamma = 0)\) and it is very likely that it also contributes to the ground state of the respective Heisenberg Hamiltonian \(((13), \gamma = 1)\).
\[
\mathcal{H}(\gamma) = -2J \sum_x \left\{ g^3(x) g^3(x + 1) + \frac{\gamma}{2} \left[ g^+(x) g^-(x + 1) + \zeta g^-(x) g^+(x + 1) \right] \right\} .
\] (13)

This behaviour is demonstrated by Fig. 1 for a ring of five spins. The figure also shows that this cycle is a ground state component for higher spin quantum numbers, too; which is found consistently for all investigated systems. Figure Fig. 2 suggests for the example of three spins that the weight of the special cycle is about the inverse of the dimension of the Hilbert subspace \( \mathcal{H}(M = 1/2) \). Considering the expectation value of the Hamilton operator in a ground state \( |\Psi_0; k\rangle \) with translational quantum number \( k \) and especially the contribution of the cycle generated by \( |m_0\rangle \) one finds

\[
\langle \Psi_0; k | \mathcal{H} | \Psi_0; k \rangle = -4J \left( c(m_0) \right)^2 N \cos \left( 2 \frac{2\pi k}{N} \right) + \text{remaining terms} .
\] (14)

The argument \( 2 \frac{2\pi k}{N} \) of the cosine function stems from the fact that

\[
\langle m_0 | \mathcal{H} \mathcal{T}_s | m_0 \rangle \propto \{ \delta_{\nu,2} + \delta_{\nu,N-2} \} .
\] (15)

Assuming that minimization of the first term of the r.h.s. of (14) cannot be counteracted enough by the \( k \)-dependence of the remaining terms, one understands that in the ground state \( k \) must be as close as possible to \( N/4 \) or \( 3N/4 \), hence \( k = \lfloor (N+1)/4 \rfloor \) or \( N-\lfloor (N+1)/4 \rfloor \).

It will be a subject of further research whether this \( k \) selection rule holds outside the range of \( N \) and \( s \) studied so far.

**IV. DEGENERACY AND FRUSTRATION EFFECTS**

Our results suggest a rather different behaviour for spin rings with non-degenerate and degenerate ground states.

The first example is given by the ground state energies of spin-\( \frac{1}{2} \)-rings which converge to the Bethe-Hulthén limit of \( E/(NJ) = 2 \ln(2) - 1/2^{22,23} \). As Fig. 3 shows this convergence is faster for even numbers of spins than for odd numbers. The latter systems are also frustrated.

In the second example effects of the ground state degeneracy on the zero-field magnetic susceptibility are discussed. To this end we introduce the interaction of all magnetic moments with the homogeneous magnetic field \( B \) (Zeeman term) into the Hamilton operator

\[
\mathcal{H}_M = -2J \sum_{x=1}^{N} \mathcal{g}(x) \cdot \mathcal{g}(x + 1) + g\mu_B B \mathcal{S}^3 = \mathcal{H} + g\mu_B B \mathcal{S}^3 .
\] (16)

The magnetisation \( \mathcal{M} \) is defined as

\[
\mathcal{M} = \frac{1}{Z} \text{tr} \left\{ -g\mu_B S^3 e^{-\beta \mathcal{H}_M} \right\} , \quad Z = \text{tr} \left\{ e^{-\beta \mathcal{H}_M} \right\}
\] (17)

and its derivative with respect to the magnetic field \( B \) results in the zero-field susceptibility.
\[ \chi_0 = \left( \frac{\partial M}{\partial B} \right)_{B=0} = g^2 \mu_B^2 \left( \frac{1}{Z} \text{tr} \left\{ \left( \frac{S^3}{\langle S^3 \rangle} \right)^2 e^{-\beta H_M} \right\} \right)_{B=0}. \]  

Figure 4 displays the zero-field susceptibility for \( N = 3 \) and various \( s = 1/2, 1, \ldots, 9 \). For small temperatures the susceptibility diverges for half integer spins (solid lines), whereas it drops to zero for integer spin quantum numbers (dashed lines), see also\(^{24} \). The different behaviour is easy to understand. In the degenerate case two eigenstates have a total magnetic quantum number \( M = \frac{1}{2} \) and the other two have \( M = -\frac{1}{2} \). The slightest magnetic field suffices to split the degeneracy in \( M \) and thus results in a finite magnetic moment even at \( T = 0 \), because the new twofold degenerate ground state has \( M = -\frac{1}{2} \). Therefore in general, for half integer spin quantum numbers and odd \( N \)

\[ \frac{\chi_0 k_B T}{g^2 \mu_B^2} \xrightarrow{T \to 0} \frac{1}{4}. \]

In the non-degenerate cases the magnetisation at small \( B \) can only grow by thermally populating excited states with non-vanishing magnetic moment.

The classical result\(^{25} \) is given by the thick dotted line. It neither goes to infinity nor to zero but to 1/2 in the used units.

Our general results on the ground state degeneracy thus enable us to describe the zero-field susceptibility at low temperatures qualitatively, for example we know, consequently, that \( \chi_0 \) drops to zero for a ring of \( N = 11 \) and \( s = 2 \) which hardly could be calculated.

At this point some words regarding frustration might be also in order. In classical physics an antiferromagnetic spin system is called frustrated if not all spins can be paired. Then the ground state possesses a non-trivial degeneracy. An example is the spin triangle in the Heisenberg model which shows a non-trivial twofold degeneracy, see for instance\(^{16,26} \). These considerations regarding the interplay of ground state degeneracy and frustration are not naively applicable to Heisenberg rings with isotropic next neighbor interaction, although it is appealing and one could conjecture it from the first example. For quantum spin rings we find that rings with an odd number of integer spins have a non-degenerate ground state. That they are nevertheless frustrated can be seen comparing their ground state energy per spin with their even neighbours, see Fig. 5. The frustration expresses itself through a weaker binding for odd \( N \).

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TABLES

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TABLE I. Properties of the AF Heisenberg ground state, energy $E$, degeneracy $deg$, shift quantum number $k$ and spin flip parity $\pi$. Values for the empty fields could not be computed in reasonable times. † – O. Waldmann, private communication. ‡‡ – projection method\textsuperscript{37}. 

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TABLE I continued
FIG. 1. Contribution of the state $|++--\rangle$ to the ground states of the Hamiltonian $H(\gamma)$, Eq. (13).

FIG. 2. Contribution of the state $|++-\rangle$ to the ground states of the Hamiltonian $H(\gamma = 1)$ (symbols). The solid line represents the inverse of the dimension of the Hilbert subspace with $M = 1/2$. 

FIG. 3. Deviation of ground state energies (symbols) for antiferromagnetic coupled Heisenberg rings \( s = 1/2 \) from the large \( N \) limit of Bethe and Hulthén\textsuperscript{22,23}. Plus symbols are used for even \( N \), crosses for odd \( N \).

FIG. 4. Zero-field susceptibility for \( N = 3 \) and various \( s = 1/2, 1, \ldots, 9 \). The solid lines show the result for half integer spins, the dashed lines for integer spin quantum numbers. The classical result is given by the thick dotted line.
FIG. 5. Ground state energies for antiferromagnetically coupled Heisenberg rings ($s = 1$). Plus symbols are used for even $N$, crosses for odd $N$. Frustration expresses itself through a weaker binding for odd $N$. 