Large-scale numerical investigations of the antiferromagnetic Heisenberg icosidodecahedron

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ABSTRACT

We present up to date investigations of the antiferromagnetic Heisenberg icosidodecahedron by means of the density matrix renormalization group method. We compare our results with modern correlator product state as well as Lanczos calculations.

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1. Introduction

Thanks to advanced chemical strategies there exist several chemical realizations of icosidodecahedral magnetic molecules: $Mo_{72}Fe_{30}$ [1], $W_{72}Fe_{30}$ [2] (both $s=5/2$), $Mo_{72}Cr_{30}$ [3] ($s=3/2$), $Mo_{72}V_{30}$ [4,5], and $W_{72}V_{30}$ [6] (both $s=1/2$). These molecules are some of the largest magnetic molecules synthesized to date [7]. Icosidodecahedral magnetic molecules are of special interest because they are highly symmetric, frustrated, exist with different spin quantum numbers, and are a kind of finite-size version of the Kagomé lattice antiferromagnet [8]. Fig. 1 shows the structure of the icosidodecahedron. It is an Archimedian Solid with 12 pentagons and 20 triangles, which means that it is geometrically frustrated [9].

Experimental investigations of these molecules are in most cases measurements of the susceptibility as a function of temperature [1–6] or magnetic field [10,11], or magnetization as a function of the applied magnetic field [1,11,12]. These experimental investigations show that the icosidodecahedral magnetic molecules are antiferromagnetic with a nonmagnetic ground state. Other experimental techniques that were applied to these molecules are NMR and μSR [13–15], INS [16], diffuse (elastic) neutron scattering, as well as specific heat measurements [17].

These molecules are usually modeled using a simple Heisenberg model [1–4,6,16–23]. Anisotropic terms were considered in Refs. [22,24]. Bond disorder and distortions (i.e., more than just one exchange constant in the Heisenberg Hamiltonian) were investigated in Refs. [10,23]. However, since the icosidodecahedral molecules

comprise $N=30$ spins, the numerical exact calculation of $T=0$ properties is possible only for the $s=1/2$ case [8,25,26]. Quantum Monte Carlo suffers from the negative-sign problem so that small temperatures are not feasible [3,4,6]. Thermodynamical properties at $T>0$ can for $s=1/2$ also be calculated quasi-exactly using the finite-temperature Lanczos method [21,23,27]. For $s>1/2$, approximations are needed. For $s=3/2$ and $s=5/2$ systems, the classical Heisenberg model was used together with efficient classical Monte Carlo algorithms [1,2,10,18,19,22,28]. However, such an approximation is inappropriate at very low temperatures. The rotational band approximation was used in Refs. [1,12,16,17,20,29,30]. Although being a quantum mechanical approximation it misses important features of frustrated systems such as magnetization jumps and plateaus or low-lying singlets in the spectrum [25]. Another approximation applied to the icosidodecahedron is spin-wave theory [20,24]. However, as for the rotational band and the classical approximation, it is not clear how accurate this approximation is.

It has to be emphasized here that although there exist many theoretical studies on the icosidodecahedron for $s>1/2$, accurate numerical calculation for the *full* Heisenberg model are very rare. The density matrix renormalization group (DMRG) method allows for treating the full Heisenberg Hamiltonian but in a reduced Hilbert space [31,32]. It relies on a controlled truncation of the Hilbert space and allows for the estimation of the accuracy so that it seems to be suited for these systems. In Ref. [33] the DMRG method has already been applied to the Heisenberg icosidodecahedron with $s=5/2$. However, only up to $m=120$ density matrix eigenstates were used, so that the accuracy of the results is rather limited for such a complicated system with a geometry that is not favorable for the DMRG method.

In this paper we apply the DMRG method to the antiferromagnetic Heisenberg icosidodecahedron. We focus on the calculation

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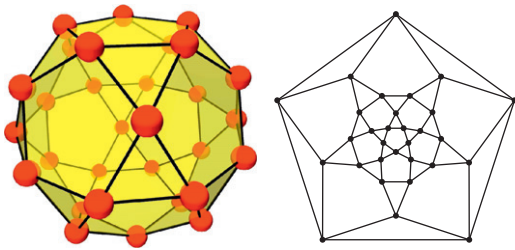
Fig. 1. Structure of the icosidodecahedron: the bullets correspond to the spin positions and the lines to interaction paths between them. The right part of the figure shows the two-dimensional projection.

of the lowest energies in subspaces of total magnetic quantum number M which allow for a calculation of the $T=0$ magnetization curve and also gaps which might be of importance for spectroscopic methods such as, e.g., inelastic neutron scattering (INS). These results are compared with very recent variational Monte Carlo calculations using correlator product states (CPS) by Neuscamman and Chan in Ref. [34]. Finally, for the case $s=1/2$ we also calculate the dynamical correlation function $S_{loc}^z(\omega)$ using the dynamical DMRG (DDMRG) [35].

2. DMRG results

The DMRG technique is best suited for open one-dimensional chain systems but can be applied to systems with an arbitrary geometry. The icosidodecahedron can be viewed as a two-dimensional lattice on a sphere (similar to the Kagome lattice, see [8]), i.e., with periodic boundary conditions. This means that the convergence is much slower than in one-dimensional systems [33]. But since DMRG is a variational method, it is clear that the ground state – or the lowest energy in a subspace – is the better the lower the corresponding energy is. Also, the truncated weight Δw offers the possibility to judge the quality of the results and an extrapolation to zero truncated weight (or $m \to \infty$) might be possible.

For the investigations throughout this article we employ the Heisenberg Hamiltonian

$$\underset{\sim}{H} = J \sum_{i<j} \underset{\sim}{\vec{s}}_i \cdot \underset{\sim}{\vec{s}}_j + g\mu_B B \sum_i \underset{\sim}{s}_i^z \tag{1}$$

with antiferromagnetic isotropic nearest neighbor exchange J only.

2.1. Numbering of the spins

When DMRG is applied to spin systems that are not one-dimensional, the usual way is to map the system on a one-dimensional chain with long-range interactions, i.e., to number the spins of the lattice [36]. However, if not very simple systems such as, e.g., ladders are investigated, it is not clear, which numbering is best suited. Such a problem also occurs, when DMRG is applied in the context of quantum chemistry, where models similar to the Hubbard model with long-range interactions appear and the ordering, i.e., the numbering of the orbitals is relevant [37–41]. Since long-range interactions diminish the accuracy of DMRG (cf. Ref. [42]) it is clear that a good ordering needs to minimize such long-range interactions.

We have tested several numberings for the icosidodecahedron. The resulting coupling matrices J_{ij} are shown in Fig. 2. The numbering used by Exler and Schnack in an earlier investigation [33] (see top left of Fig. 2) gives a very regular "interaction pattern" with rather-short-ranged interactions, but the "periodic boundaries", i.e., interactions between the first and the last spins, are clearly not optimal for the DMRG algorithm with two center

sites. As proposed in Ref. [37], we have used a variant of the reverse Cuthill–McKee algorithm [43,44], the RCMD algorithm, which aims to number the vertices such that the bandwidth of the matrix is minimized. We have also used the Sloan algorithm [45] which minimizes the "envelope size", i.e., the sum of the "row bandwidths". (The bandwidth is the maximum of the row bandwidths.) We have used these algorithms as implemented in Mathematica [46]. The figure also shows an unoptimized numbering for comparison.

The results of DMRG calculations (using the ALPS DMRG code [47]) for the different spin numberings are shown in Fig. 3. We have calculated the ground state energy of the $s=1/2$ icosidodecahedron with an increasing number of kept density matrix eigenstates (m) so that the convergence can be investigated and a comparison with the exact ground state energy (see Ref. [8]) is possible. One can see that the different optimized numberings (Exler/Schnack, RCMD, and Sloan) give almost identical results whereas the unoptimized numbering gives much worse results. These results show that a "good" numbering of the spins is absolutely essential if the DMRG method is applied to a spin system with a complicated structure. For the following results we have always used the numbering as proposed by Exler and Schnack.

2.2. Lowest energy eigenvalues and magnetization curves

As a next step we have calculated the lowest energies in the M subspaces for the icosidodecahedron with $s > 1/2$ using DMRG. The results for the $s=1/2$ system already showed that DMRG is able to produce very accurate results for this system with relative errors smaller than 10^{-3}.

Fig. 4 shows the lowest energy eigenvalues in the subspaces of total magnetic quantum number M for the icosidodecahedron with $s=1$ and $s=3/2$ as obtained by DMRG and – for the large-M subspaces ($M > 18$ for $s=1$ and $M > 33$ for $s=3/2$) – Lanczos calculations. We have used up to $m=2500$ density matrix eigenstates for the $s=1$ case and up to $m=2000$ for the $s=3/2$ case. The largest truncated weight within a sweep is of the order of 7×10^{-4} for the $M=0$ subspace of the $s=1$ icosidodecahedron and of the order of 4×10^{-4} for the $s=3/2$ case. That the truncated weight for the $s=1$ icosidodecahedron is larger than for $s=3/2$ although more states have been used for $s=1$ indicates that it cannot be reliably used for a quantitative estimate of the error. The reason for this behavior might be that the results are not yet fully converged for the value of m that we have used, although we have carried out up to 60 sweeps for the calculations.

The rotational band model predicts a behavior of the form $E_{min}(M) = aM(M+1) + b$, i.e., a parabolic dependence [12]. The insets of Fig. 4 show that this is a good approximation for the energy eigenvalues of the full Heisenberg model. The simple rotational band approximation predicts a proportionality constant of $a=0.1$. The linear fits as shown in the insets give the results $a=0.111$ for $s=1$ and $a=0.108$ for $s=3/2$, very close to the simple rotational band approximation. However, if one uses these (DMRG) data to calculate the zero-temperature magnetization curve, it becomes clear that there are some crucial deviations from the ideal parabolic dependence. If there was an ideal parabolic dependence, the resulting magnetization curve would consist of steps with constant widths. Fig. 5 shows the resulting zero-temperature magnetization curves as calculated using the DMRG data. Again, the exact diagonalization data for $s=1/2$ are taken from Ref. [8].

One can see that the magnetization curves do not consist of steps with constant widths. There are some anomalies as expected for frustrated systems. The plateaus at $\mathcal{M}/\mathcal{M}_{sat} = 1/3$ are clearly visible. The magnetization jumps due to the independent magnons [26]