Spin Gaps in a Frustrated Heisenberg model for CaV$_4$O$_9$

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Abstract

I report results of a density matrix renormalization group (DMRG) study of a model for the two dimensional spin-gapped system CaV$_4$O$_9$. This study represents the first time that DMRG has been used to study a two dimensional system on large lattices, in this case as large as $24 \times 11$, allowing extrapolation to the thermodynamic limit. I present a substantial improvement to the DMRG algorithms which makes these calculations feasible.

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Since the discovery of the high temperature superconductivity, condensed matter physicists have searched for other two dimensional systems with exotic “spin liquid” ground states. Thus considerable excitement accompanied the recent discovery of CaV$_4$O$_9$, a two dimensional, frustrated $S = 1/2$ Heisenberg spin system, with a substantial spin gap [1]. This system has been modeled by a depleted square lattice Heisenberg model, with both nearest and next-nearest exchange interactions [2]. It consists of a square lattice with 1/5 of the spins missing, as shown in Figure 1, and has been called a CAVO lattice [3]. It is believed that the superexchange is mediated by out-of-plane oxygen atoms, resulting in a very large next-nearest exchange: $J' \approx J/2$ [2]. This frustrating interaction helps stabilize the spin-gapped state against Néel order.

One can think of the ground state of this system as a “plaquette resonating valence bond” state [2]: no phase transition is expected if the interactions between plaquettes are adiabatically removed, and the ground state of a single plaquette is perfectly described by a resonating valence bond (RVB) variational ansatz (in the absence of frustration). In addition, in the weakly-interacting plaquette limit, pairs of holes bind on plaquettes, suggesting the possibility of a superconducting ground state upon doping. The system is reminiscent of ladder systems with even numbers of legs. In that case, somewhat more complicated RVB states have been useful in describing the spin liquid ground states of undoped ladders [4,5], and upon doping, strong pairing correlations are observed numerically [6].

A number of theoretical and numerical treatments have been performed on this model in the last year [2,7–11]. Troyer, Kontani and Ueda made the most reliable determination of the unfrustrated phase diagram using a quantum Monte Carlo loop algorithm [10]. An important conclusion of this study was that simple 1/5 depletion of the isotropic square lattice, without frustration, does not destroy Néel order. A spin liquid ground state was found when the couplings within a plaquette were about 10% greater than between plaquettes. This result contradicted earlier (non-loop) quantum Monte Carlo calculations on smaller systems [7]. It was not possible to include frustration because of sign problems. Gelfand, et. al. [3] applied series expansion techniques and were able to study the frustrated and unfrustrated
systems. Their results were in agreement with Troyer, et. al. for the unfrustrated system.
They concluded that a next-nearest neighbor interaction $J' = J/2$ was consistent with experimental results.

I present results here from density matrix renormalization group (DMRG) calculations \cite{12} for the spin gap of the frustrated CAVO lattice. The results are in agreement with those of Gelfand, et. al., and, in fact, when extrapolated to the thermodynamic limit, appear to be more accurate. Although DMRG is usually much more accurate than other numerical techniques for large one dimensional systems, these are the first reliable results for systems wide enough to be considered two dimensional—up to $24 \times 11$. These calculations are feasible because of an important improvement to the DMRG algorithms, which I present here, which increases the speed of the calculations by up to two orders of magnitude.

The improvement to DMRG involves keeping track of the wavefunction from step to step. The step referred to here is the process of adding a site to a block and requires the diagonalization of a superblock configuration of two blocks and two sites \cite{12}. In each DMRG step, an iterative sparse matrix algorithm, such as the Davidson method, is used to find the ground state of the superblock. In the original formulation of DMRG, no starting point for the Davidson procedure was specified. To ensure that the DMRG procedure is always stable and convergent, the superblock ground state usually has to be determined to rather high accuracy. (One diagonalization which converges to a low-lying eigenstate other than the ground state ruins the accuracy of the entire DMRG calculation.) Consequently, a substantial number of Davidson steps are necessary to converge to sufficient accuracy, typically 40-100. The total calculation time is proportional to the average number of Davidson steps.

If a very good initial guess is available for the Davidson procedure, the number of Davidson steps can be reduced substantially. An ideal initial guess, for the case of the finite system DMRG algorithm, is the final wavefunction from the previous DMRG step. This wavefunction, however, is in a different basis, corresponding to a different superblock, but it can be transformed into the basis corresponding to the current superblock, as I describe below. Use of this transformation to obtain the initial state in a Davidson diagonalization
can reduce the number of Davidson steps by one half, typically, assuming that one iterates Davidson until it converges to high accuracy. Use of this initial guess has an even more important advantage: it is not necessary to converge to high accuracy, since there is no danger of converging to an incorrect low-lying eigenstate. The initial guess not only has low energy, it approximately describes the correct eigenstate, as obtained in the previous step. In fact, the algorithm can be made completely stable even if the number of Davidson steps is restricted to two or three! Thus one saves a factor of 20-50 in the time required by the Davidson procedure. The overall speedup is somewhat reduced from this factor because the calculation time to perform other parts of the DMRG procedure, such as diagonalizing the density matrix, becomes significant.

A DMRG step adds a site onto a block, constructing an appropriate basis for the new block. Let $|\alpha_l\rangle$ be the states of left block $l$, where $l$ is the rightmost site of the block. Let $|s_l\rangle$ be the states of site $l$. Then the basis states for the new left block are given by

$$
|\alpha_{l+1}\rangle = \sum_{s_{l+1},\alpha_l} L^{l+1}[s_{l+1}]_{\alpha_{l+1},\alpha_l} |\alpha_l\rangle \otimes |s_{l+1}\rangle.
$$

(1)

This notation is similar to that of Ostlund and Rommer \[13\]. The transformation matrix $L^{l+1}[s_{l+1}]_{\alpha_{l+1},\alpha_l}$ is a slightly rewritten form of the truncated matrix of density matrix eigenvectors. The states of the right block $|\beta_{l+3}\rangle$ were formed at an earlier DMRG step in a similar fashion

$$
|\beta_{l+3}\rangle = \sum_{s_{l+3},\beta_{l+4}} R^{l+3}[s_{l+3}]_{\beta_{l+3},\beta_{l+4}} |s_{l+3}\rangle \otimes |\beta_{l+4}\rangle.
$$

(2)

I do not assume any reflection symmetry for the lattice: the $L$ and $R$ matrices are independent.

A superblock basis state is written in the form

$$
|\alpha_l s_{l+1} s_{l+2} \beta_{l+3}\rangle = |\alpha_l\rangle \otimes |s_{l+1}\rangle \otimes |s_{l+2}\rangle \otimes |\beta_{l+3}\rangle.
$$

(3)

A superblock wavefunction $|\psi\rangle$ is written in this basis as

$$
|\psi\rangle = \sum_{\alpha_l s_{l+1} s_{l+2} \beta_{l+3}} \psi(\alpha_l s_{l+1} s_{l+2} \beta_{l+3}) |\alpha_l s_{l+1} s_{l+2} \beta_{l+3}\rangle.
$$

(4)
One needs to transform this wavefunction into the basis appropriate for the next DMRG step, $|\alpha_{l+1}s_{l+2}s_{l+3}\beta_{l+4}\rangle$. The transformation is not exact, since there is a truncation in going from $|\alpha ls_{l+1}\rangle$ to $|\alpha_{l+1}\rangle$. However, the states $|\alpha_{l+1}\rangle$ are formed using the density matrix to be ideally adapted for representing $|\psi\rangle$, so for the transformation of the wavefunction only, one can approximate

$$\sum_{\alpha_{l+1}} |\alpha_{l+1}\rangle\langle\alpha_{l+1}| \approx 1. \quad (5)$$

With this approximation one readily obtains

$$\psi(\alpha_{l+1}s_{l+2}s_{l+3}\beta_{l+4}) \approx \sum_{\alpha_{l}s_{l+1}\beta_{l+3}} L^{l+1}[s_{l+1}]_{\alpha_{l+1},\alpha_{l}} \psi(\alpha ls_{l+1}s_{l+2}\beta_{l+3}) R^{l+3}[s_{l+3}]_{\beta_{l+3},\beta_{l+4}}. \quad (6)$$

The most efficient way to implement this transformation numerically is to first form the intermediate wavefunction

$$\psi(\alpha_{l+1}s_{l+2}\beta_{l+3}) = \sum_{\alpha_{l}s_{l+1}\beta_{l+3}} L^{l+1}[s_{l+1}]_{\alpha_{l+1},\alpha_{l}} \psi(\alpha ls_{l+1}s_{l+2}\beta_{l+3}), \quad (7)$$

and then form the final result

$$\psi(\alpha_{l+1}s_{l+2}s_{l+3}\beta_{l+4}) = \sum_{\beta_{l+3}} \psi(\alpha_{l+1}s_{l+2}\beta_{l+3}) R^{l+3}[s_{l+3}]_{\beta_{l+3},\beta_{l+4}}. \quad (8)$$

In this form, the transformation requires very computer little time compared to other parts of the calculation.

This transformation is used for one half of the DMRG steps, when a site is being added to the left block. An analogous transformation is used for adding a site to the right block.

Implementing this transformation requires saving all the matrices $L$ and $R$, which is ordinarily not done. The storage for these matrices is typically 20-30% of the storage required for the blocks themselves, so the extra storage is not a major concern. In an efficient DMRG implementation for a typical machine, such as a Cray or a workstation, both the blocks and the transformation matrices should be stored on disk. The calculations described here sometimes required more than a gigabyte of scratch disk storage, but never more than 80-90 megabytes of RAM.
In many one-dimensional systems, DMRG converges in one or two sweeps through the system. In quasi-one or two-dimensional systems, the number of sweeps needed can easily grow to five to 10. Another important improvement in efficiency comes from gradually increasing the number of states kept per block as one performs the sweeps \[^{[14]}\]. In this case the calculation time is dominated by the last sweep, during which the number of Davidson steps per DMRG step can be constrained to be only two or three. Compared to a DMRG calculation keeping a constant number of states and without the wavefunction transformation, the speedup can be over two orders of magnitude.

If the lattice is reflection-symmetric, an additional factor of two can be saved in both calculation time and memory, but a different wavefunction transformation is needed for the DMRG step where the superblock is symmetric. Also, it is possible to adopt somewhat similar methods to obtain good initial guesses for the wavefunction in the infinite system method. These techniques will be reported elsewhere \[^{[15]}\].

The beginning of any DMRG calculation of a 2D system is a mapping of the 2D lattice onto a 1D chain—basically, one must choose an order to traverse the sites. It is standard to use the scanline mapping—fix \(x\), step through all values of \(y\), then increment \(x\), etc.—which has the advantage of keeping the blocks as contiguous as possible. In treating the CAVO lattice, it is more advantageous to modify this slightly so that all the sites in a plaquette are traversed in succession. This incorporates the fact that for the parameters I consider here, correlations within a plaquette are strongest.

I consider the Heisenberg Hamiltonian

\[
H = J_{ij} \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j
\]

defined on an \(L_x \times L_y\) CAVO lattice with \(S = \frac{1}{2}\). As shown in Fig. 1, I take all nearest-neighbor \(J_{ij}\) to be identical, with value \(J_1 = 1\), setting the energy scale. All next-nearest-neighbor \(J_{ij}\) are also identical, with value \(J_2\). All other \(J_{ij}\) are zero.

I have studied open systems, although putting periodic boundary conditions in the short direction \(y\) is not particularly difficult. Having open boundary conditions allows a variety
of sizes to be studied. We consider a set of systems of length 24 and width up to 11. Since interactions are strongest within plaquettes, only full plaquettes are included. We keep up to \( m = 600 \) states per block, with truncation error at worst about \( 10^{-5} \). Typical errors in the total energy, for the larger systems, were less than about \( 10^{-3} \). For systems of width 8 and larger, an extrapolation to \( m \to \infty \) was used, assuming an exponential fall off in the error in the energy as a function of \( m \) \cite{ref1, ref2}. Corrections were at most about \( 10^{-3} \). For each system we calculate the ground state energies with quantum numbers \( S_z = 0 \) and \( S_z = 1 \). The spin gap \( \Delta \) is the difference in energies. The largest system, \( 24 \times 11 \), took about 15 - 20 hours of workstation time (rated at 135 SPECfp92) for one value of \( S_z \).

Figure 2 shows some of the results, for various widths of the system \( L_y \). From the width 7 data, we see that the spin gap is peaked at \( J_2 = 0.5 \), which also happens to be appropriate for CaV\(_4\)O\(_9\). For larger values of \( J_2 \) it falls rapidly towards zero. (The gap for \( J_2 = 0.8 \) was consistent with zero, within uncharacteristically large error bars of about 0.05.)

Finite size extrapolation is crucial to determine the spin gap for smaller values of \( J_2 \). Excellent extrapolations can be obtained if one assumes the low lying spin excitations obey a \emph{relativistic} dispersion relation

\[
\Delta(k)^2 = \Delta^2 + v^2 k^2, \quad (10)
\]

where \( \Delta \) is the bulk gap and the velocity \( v \) corresponds to the speed of light. Lorentz invariant low energy excitations are common (but not universal) in gapped, one dimensional spin systems, reflecting Lorentz invariance of the corresponding nonlinear sigma model. Considerations of simple particle-in-a-box systems with various boundary conditions indicate that a generic boundary condition at the edge of an open system fixes the logarithmic derivative of the wavefunction \( \psi \), \( \frac{d\psi}{dx}/\psi = \text{const} \). This can be shown to imply that the lowest value of \( k \) allowed in a 1-D box of size \( L \) is given by \( \pi/(L - a) \), where \( a \) depends on boundary effects but is independent of \( L \). This leads to the following form for the gap as a function of system size

\[
\Delta(L_x, L_y)^2 = \Delta^2 + \frac{\pi^2 v^2}{(L_x - a)^2} + \frac{\pi^2 v^2}{(L_y - a)^2}, \quad (11)
\]
I have found that for $J_2 < \sim 0.3$, we can set $a = 0$, and still obtain excellent fits. This is indicated by linear behavior when $\Delta^2$ is plotted versus $1/L_x^2$, with $L_x$ fixed. Results are shown in Figure 3 for typical values of $J_2$. For $J_2 = 0.5$, the fits were poor for $a = 0$, and mediocre with $a$ nonzero, because the data was slightly irregular. Gelfand, et.al. [4] observed that the gap minimum can move away from $(\pi, \pi)$ for larger values of $J_2$. This data supports that proposition for $J_2 = 0.5$. In the case of an incommensurate gap minimum, we would expect irregular behavior of the gap as a function of $L$, as the value of $k$ allowed by the lattice which is closest to the minimum would jump about as $L$ increased.

Using the fits shown, I corrected for the finite value of $L_x$ and obtained results for $\Delta$ in the thermodynamic limit. Figure 4 shows the results as a function of $J_2$. The extrapolation using Eq. (11) yielded imaginary gaps for $J_2 = 0$ and 0.05, which we interpret to mean $\Delta = 0$. The transition to a spin gapped state appears at $J_2 = 0.06(1)$. This result is in agreement with previous work indicating that the system with $J_2 = 0$ is close to the disordered phase. The value at $J_2 = 0.5$, $\Delta = 0.515(15)$, is somewhat lower than the series results of Gelfand, et.al., $\Delta = 0.57(3)$. However, the results are completely consistent if the shift in the gap minimum results in an overestimate in the series results by about 0.05, as Gelfand, et.al. suggest.

I have presented the first results using DMRG on a two dimensional system for lattices wide enough to allow extrapolation to the thermodynamic limit. DMRG still is primarily a one dimensional technique, in that the accuracy falls off rapidly as the system’s width increases. The CAVO system studied here was less difficult than many other two dimensional systems, both because of the existence of a gap and the depleted character of the lattice. Nevertheless, as DMRG and computational resources improve, I expect it will become a standard numerical technique for two dimensional systems, including doped fermion systems.

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FIGURES

FIG. 1. A $12 \times 7$ open CAVO lattice. The solid lines are nearest-neighbor bonds with exchange $J_1$, and the dotted lines are next-nearest-neighbor bonds with exchange $J_2$. Only complete plaquettes which fit within a $12 \times 7$ rectangle are retained.

FIG. 2. Spin gap as a function of $J_2$ for various widths $L_y$, with $L_x = 24$.

FIG. 3. Gap squared as a function of $L_y^{-2}$. The solid lines are linear fits, excluding the $L_y = 4$ point for $J' = 0.2$. The dashed line is an alternative fit which includes an $L_y^{-3}$ term.

FIG. 4. Spin gap extrapolated to the thermodynamic limit, as a function of $J_2$. Where not shown, errors are comparable to the point size.
Fig. 1
White
Fig. 2

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Fig. 3

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Fig. 4

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