Phase diagram of depleted Heisenberg model for CaV$_4$O$_9$

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We have numerically investigated the 1/5-depleted Heisenberg square lattice representing CaV$_4$O$_9$ using the Quantum Monte Carlo loop algorithm. We have determined the phase diagram of the model as a function of the ratio of the two different couplings: bonds within a plaquette and dimer bonds between plaquettes. By calculating both the spin gap and the staggered magnetization we determine the range of stability of the long range ordered (LRO) phase. At isotropic coupling LRO survives the depletion. But the close vicinity of the isotropic point to the spin gap phase leads us to the conclusion that already a small frustrating next nearest neighbor interaction can drive the system into the quantum disordered phase and thus explain the spin gap behavior of CaV$_4$O$_9$.

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The stability of the long range ordered (LRO) ground state of the planar Heisenberg model has been at the focus of investigations for a long time. The recent discovery of a spin gap in CaV$_4$O$_9$ [1] has given special importance to this question. This compound can be described by a 1/5-depleted planar antiferromagnetic Heisenberg model [2]. One of the important questions regarding this material is whether the depletion of the square lattice can account for the spin gap, or if additionally frustration effects are important.

The role of lattice defects and depletion in destabilizing LRO has been studied in a variety of contexts. One example are spin ladders which can be obtained from the planar copper oxide materials, by breaking up the planes into ladders of constant width [3]. Another way to destroy LRO is to deplete the lattice. The bonds between spins are then weakened, similar to the introduction of holes, and quantum fluctuations are enhanced, which might destroy LRO. An example is the triangular Heisenberg antiferromagnet, which exhibits LRO. Depletion of 1/4 of the spins, leads to the Kagome-lattice, which is believed to have no LRO [4].

The stability of LRO is also of relevance in the field of high temperature superconductors. There the rapid destruction of LRO upon hole doping and the possibility of realizing a doped resonating valence bond (RVB) phase [5], exhibiting a finite gap in the spin excitation spectrum (spin gap) are of great current interest. The study of lattice defects, such as depletion or the formation of ladders can give valuable insights [6].

The lattice structure of CaV$_4$O$_9$ and the 8-spin unit cell [7] used in our simulations is shown in Fig. [1]. It can be viewed as consisting of loosely connected 4-spin plaquettes. Two topologically different types of bonds can be distinguished. One are bonds within a plaquette $J_0$, the other are dimer-bonds connecting plaquettes $J_1$. Additionally CaV$_4$O$_9$ is believed to have a significant frustrating next nearest neighbor (n.n.n.) antiferromagnetic interaction $J_2$. 

Ueda et al. [2] and Imada and Katoh [8] have argued that the spin gap can be explained as originating in a plaquette RVB state, consisting of local singlets of the four spins on a plaquette. This plaquette RVB state is the exact ground state in the limit $J_1 = 0$. Second order perturbation theory around this limit suggests that it survives even at isotropic coupling [9]. A perturbation around the dimer limit $J_0 = 0$, [2] also leads to a wide range of stability of the dimer singlet phase, but a small range of the couplings exists, where no gap is observed in perturbation theory. First Quantum Monte Carlo (QMC) results by Katoh and Imada [8] also suggest the existence of a finite gap $\Delta = 0.11 \pm 0.03$ at isotropic coupling.

Linear spin wave theory (LSW) [2] and Schwinger boson mean field theory results [10] on the other hand indicate that LRO could survive at isotropic coupling despite the depletion of the lattice. Exact diagonalization results are also contradictory [11,12]. They suffer greatly from the restriction to very small clusters and the extrapolation to the infinite system size is difficult. Sano and Takano [11] and Albrecht and Mila [12] find a small spin gap, but also a substantial staggered magnetization [5]. No definite conclusions can thus be drawn from these calculations either.

To resolve these conflicting results we have determined the phase diagram (see Fig. [2]) of the non-frustrated model using the QMC loop algorithm [12]. Using this highly efficient cluster method we can investigate larger systems at lower temperatures and with a much higher accuracy than possible with the standard world line algorithm used by Katoh and Imada [8]. We have investigated lattices with up to $N = 800$ spins at temperatures down to $T = 0.02$. The QMC method suffers from no systematic errors and the results are reliable within the statistical errors.

We find a wide region of stability of the Neel-ordered phase as a function of the ratio of the couplings $\alpha = J_0/ J_1$. We estimate the lower boundary to lie between $0.55 < \alpha^c < 0.65$ and the upper boundary between $1.05 < \alpha^u < 1.1$. At isotropic coupling LRO thus survives the depletion of the lattice. The critical point $\alpha^c$ is quite close to isotropic coupling, and a small frustration.
might be sufficient to drive the system into the disordered state.

To determine this phase diagram we have calculated both the spin gap and the staggered magnetization. The spin gap $\Delta$ can be obtained from the low temperature behavior of the uniform susceptibility $\chi$. Figure 3 shows $\chi(T)$ for some representative points. In a gapped system it decreases exponentially as $e^{-\Delta/T}$ for low temperatures.

Any finite system exhibits a gap, and thus a careful treatment of finite size effects is necessary. For each temperature we have done calculations on clusters of different size (up to $N = 800$ spins) to see whether our results have converged to the infinite system size limit. In the regions of a large gap the convergence is quite rapid and it is no problem to obtain the gap $\Delta$ from a fit of the low temperature behavior of the uniform susceptibility $\chi$ to an exponential decay $e^{-\Delta/T}$. In case of a vanishing or very small gap on the other hand the susceptibility decreases linearly down to the lowest temperatures $T_0$ we could study reliably on our finite clusters. In these cases we cannot definitely decide about the existence of a gap, but can only give an upper bound $\Delta < T_0$.

In Fig. 2 we plot the gap obtained in this way together with the perturbation theory estimates [3]. Perturbation theory is surprisingly accurate, but overestimates the gap slightly. Specifically at the isotropic point we do not see any indication for a gap, in contradiction to Katoh and Imada [3]. Their calculation of $\chi(T)$ is for a much smaller lattice (80 spins), and their gap may be due to finite size effects [13].

The existence of LRO can be checked by calculating the staggered magnetization $m_s$:

$$m_s^2 = \langle \psi | \sum_r S_r (-1)^{|r|} | \psi \rangle.$$  

$m_s$ vanishes in the infinite system size limit in case of purely short range correlations, while it is finite for LRO. The finite size scaling of $m_s$ is known and a reliable extrapolation possible [13]:

$$m_s(N) = m_s(\infty) + O \left( \frac{1}{\sqrt{N}} \right).$$

Figure 4 shows the system size dependence of $m_s$. Let us first discuss couplings in the spin gap regime. There the finite result can be extrapolated linearly in $1/\sqrt{N}$ to zero moment in the infinite system [Fig. 4(a)]. In the double logarithmic plot [Fig. 4(b)] it can clearly be seen that the results for finite clusters bend down and approach the linear decrease (slope 1). The results for couplings in the LRO phase on the other hand bend up and reach a constant value asymptotically. At the critical coupling itself we expect a power law with a critical exponent different from the $1$-behavior of the gapped phase. A rough estimate shows an exponent of the order 0.5, as expected from the mapping to the non-linear $\sigma$ model [15]. This exponent is indicated as a dotted line. A more detailed investigation, to obtain a reliable estimate for the exponent and a better estimate for the critical coupling is currently under progress.

Although the system size dependence is asymptotically linear in $N^{-1/2}$, our lattices are not yet large enough to be really in that limit. To get an estimate for the quality of our extrapolations we extrapolate both $m_s$ and $m_s^2$. In case of LRO both extrapolations should be linear. We observe that, as seen in Fig. 4(a) the system size dependence is not perfectly linear, but still bends down a little bit. Thus we take the value obtained from this fit as an upper bound. In a plot of $m_s^2$ on the other hand a slight upwards bend can be observed and we take that extrapolation as a lower bound. Both extrapolations agree well. In the phase diagram (Fig. 5) we show the average value, with the error bars indicating these upper and lower bounds. We have tested this procedure for the square lattice, where our result of $m_s = 0.306(3)$ agrees perfectly with the most accurately known value $m_s = 0.3074(4)$ [14]. Again close to the critical points the moment is very small and a definite decision about a nonzero magnetization difficult. The magnitude of the staggered moment compares well with the results of linear spin wave theory (LSW) (also shown in Fig. 2), but the range of stability of the LRO phase is overestimated by the LSW.

The conclusions obtained from the estimation of the gap and the staggered moment are perfectly consistent. Starting from the dimer limit we see a decrease of the gap as $J_0$ is increased. At $\alpha = J_0/J_1 = 0.55$ we can still find a finite gap, while at $\alpha = 0.65$ we observe a finite staggered magnetization and a zero or small gap. Thus we conclude that at a critical coupling $0.55 < \alpha^c < 0.65$ the dimer singlet phase becomes unstable and the model exhibits LRO. The critical coupling is probably close to $\alpha = 0.6$. There we cannot definitely decide about the existence of a gap or LRO from our finite cluster results. Starting from the plaquette side the gap also decreases as we increase $J_1$, but the plaquette RVB state is stable for a wider range of couplings than the dimer state. This is quite natural, as each spin is connected to one dimer bond, but two plaquette bonds. Perturbation theory predicts that the isotropic point is still in the range of stability of the plaquette RVB state, but our QMC simulations show that LRO sets in at $1.05 < \alpha^p < 1.11$. At the isotropic point we observe a substantial nonzero staggered magnetization $m_s = 0.178(8)$.

Comparing our results to previous calculations we find that the region of stability of LRO is larger than estimated by second order perturbation theory [3], but smaller than estimated by linear spin wave theory and Schwinger boson mean field theory [10]. Our results also agree well with the exact diagonalization estimates of the staggered magnetization [10], while the extrapolation of the spin gap data by exact diagonalization is unreliable.
By varying the ratio of the couplings in the 1/5-th depleted square lattice we can study both the LRO phase and the disordered phase of a two dimensional quantum antiferromagnet, without having to introduce frustration or to break symmetries, as in the dimerized square lattice model \[15\]. This model is thus ideal to study the critical behavior and to test the predictions made by Chakravarty, Halperin and Nelson based on the 2 + 1-dimensional non-linear σ model \[15\].

In comparison to experimental results on CaV$_4$O$_9$ we conclude that the depletion of the square lattice alone is not sufficient to destroy LRO in the Heisenberg antiferromagnet, but it is very close to the critical point. An additional frustrating next nearest neighbor coupling is needed to drive the system into the gapped plaquette RVB phase. All estimates from perturbation theory \[2\] and exact diagonalization \[11\] agree that the next nearest neighbor coupling could even be larger than the nearest neighbor coupling. This question is of no relevance to the validity of our results here, since the lattice obtained by considering only the n.n.n. coupling are just two non-interacting copies of the same lattice. The inclusion of the n.n. coupling on the other hand would then lead to a different frustrating coupling, but the main effect, frustration would be the same.

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[11] K. Sano and K. Takano, preprint cond-mat/9510160.
[12] H.G. Evertz, G. Lana, and M. Marcu, Phys. Rev. Lett. 70, 875 (1993).
[13] Katoh and Imada \[8\] have also estimated the spin gap from the difference of the energies of the $S^z = 1$ and $S^z = 0$ subspaces. While this method is correct in the $T \to 0$ limit, their temperature of $T \approx 0.1$, which is of the order of their gap estimate, seems to be not low enough to accurately obtain the ground state properties.

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Care must be taken in the extrapolation, since the shape of the cluster is different for each cluster size. In fact the 16 and 24 site clusters studied by Sano and Takano \[11\] consist of 2 × 2 and 2 × 3 plaquettes. This extrapolation should thus rather be viewed as a one-dimensional one to the 2 × ∞ limit.

[14] M. Troyer, preprint ETH Zürich.

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FIG. 1. The lattice structure of the depleted Heisenberg lattice describing CaV$_4$O$_9$. Indicated are the two different types of bonds, plaquette-bonds $J_0$ and dimer bonds $J_1$.

FIG. 2. Phase diagram as a function of the ratio $J_0/J_1$. (a) shows the whole range of couplings. The leftmost point corresponds to the dimer limit $J_0 = 0$ and the rightmost point to the plaquette limit $J_1 = 0$. (b) A detail of the phase diagram around the isotropic point plotted as function of $J_0/J_1$. Circles indicate our QMC results for the spin gap, normalized by $J_0 + J_1$. In the gapless region the error bar indicates an upper limit for the gap. Diamonds show the staggered magnetization. The error bars indicate the upper and lower bound, as described in more detail in the text. As reference we have included the perturbation theory estimates for the gap \[2\] and the linear spin wave theory (LSW) estimates for the staggered moment.

FIG. 3. Temperature dependence of the uniform susceptibility $\chi$ for different ratios of the couplings $J_0/J_1$. For each temperature the system size was taken large enough to see the value for the infinite system. The lowest temperatures were calculated on a $N = 512$ spin lattice. As a reference we have included results for the square lattice Heisenberg model. The temperature is in units of the larger of the couplings $J_0$ and $J_1$.

\[7\] Paul A. Lee, preprint cond-mat/9510049.
FIG. 4. System size dependence of the staggered magnetization $m_s$ for different ratios of the couplings $J_0/J_1$. For each system size the temperature was chosen low enough to see the ground state properties. The largest systems contain $N = 800$ spins. (a) $m_s$ plotted as a function of $N^{-1/2}$. A linear extrapolation gives the bulk value. (b) A double logarithmic plot clearly shows the existence of long range order or the linear decrease with system size respectively. Included as guides to the eye are two straight lines corresponding to power law decays with powers 1 and 0.5.
Figure 1, Troyer et al.
Figure 2, Troyer et al.

(a) dimer J_0/J_1 vs. plaquette

(b) isotropic coupling J_0/J_1 vs. plaquette

- Spin gap Δ/(J_0+J_1) by QMC
- Spin gap in perturbation theory
- Staggered moment m_s
- Staggered moment in LSW theory
Figure 3, Troyer et al.
Figure 4, Troyer et al.