Phase Diagram of Depleted Heisenberg Model for CaV_4O_9

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Using the quantum Monte Carlo loop algorithm we have determined the phase diagram of the 1/5-depleted Heisenberg square lattice model representing CaV_4O_9 as a function of the ratio of the two different couplings: bonds within a plaquette and dimer bonds between plaquettes. At isotropic coupling long range order *survives* the depletion, but the system is close to the quantum critical point. Already a small frustration can thus drive the system into the quantum disordered phase and explain the spin gap behavior of CaV_4O_9. [S0031-9007(96)00205-0]

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The stability of the long range ordered (LRO) ground state of the planar Heisenberg model has been the focus of investigations for a long time. The recent discovery of a spin gap in CaV_4O_9 [1] has given special importance to this question. This compound can be described by a 1/5-depleted planar antiferromagnetic Heisenberg model [2,3]. 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 is spin ladders which can be obtained from the planar copper oxide materials, by breaking up the planes into ladders of constant width [4]. 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 kagomé lattice, which is believed to have no LRO [5].

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 [6], 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 [7].

The lattice structure of CaV_4O_9 and the 8-spin unit cell [8] used in our simulations are 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 is bonds within a plaquette J_0; the other is dimer bonds connecting plaquettes J_1. Additionally CaV_4O_9 is believed to have a significant frustrating next nearest neighbor (NNN) antiferromagnetic interaction [9].

Ueda *et al.* [2] and Katoh and Imada [3] 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 [2,3]. 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 [3] 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 [10,11]. 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 [10] find a small spin gap, but also a substantial staggered magnetization [10]. No definite conclusions can thus be drawn from these calculations either.

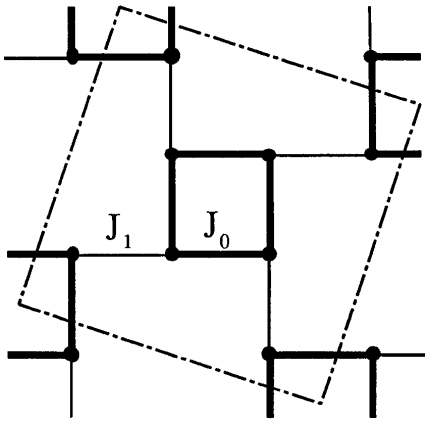

FIG. 1. The lattice structure of the depleted Heisenberg lattice describing CaV_4O_9. Indicated are the two different types of bonds, plaquette bonds J_0 and dimer bonds J_1.

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To resolve these conflicting results we have determined the phase diagram (see Fig. 2) of the nonfrustrated 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 [3]. 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.

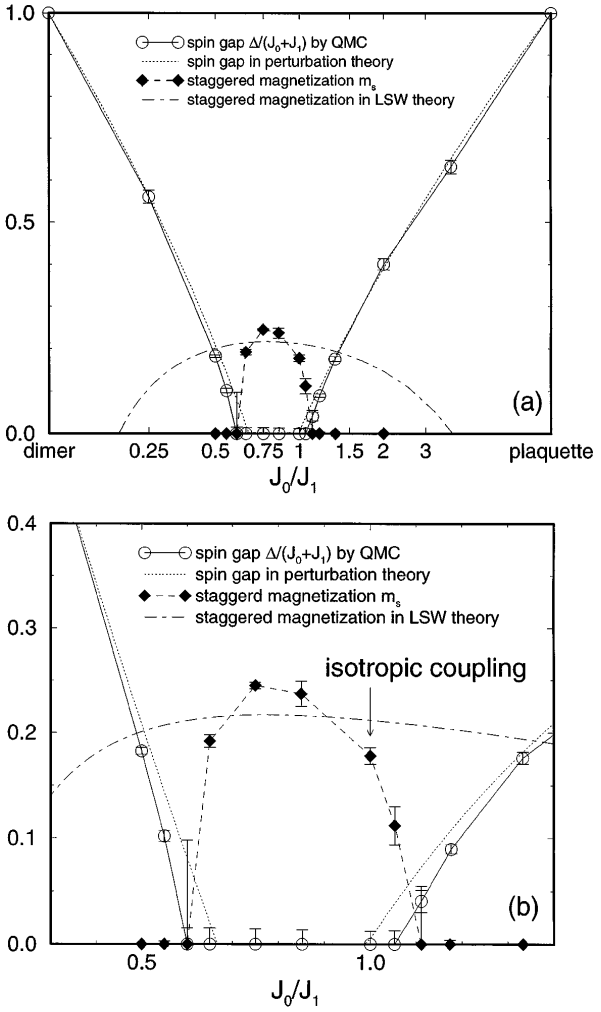

FIG. 2. Phase diagram as a function of the ratio J_0/J_1. (a) The whole range of couplings. The leftmost point corresponds to the dimer limit $J_0 = 0$ and the rightmost point to the plaquette limit $J_1 = 0$. (b) A detail of the phase diagram around the isotropic point plotted as a function of J_0/J_1. 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 bounds, as described in more detail in the text. As a reference we have included the perturbation theory estimates for the gap [2] and the linear spin wave theory (LSW) estimates for the staggered moment.

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 $0.55 < \alpha_c^l < 0.65$ and the upper boundary between $1.05 < \alpha_c^u < 1.1$. At isotropic coupling LRO thus survives the depletion of the lattice. The critical point α_c^u 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 Δ can be obtained from the low temperature behavior of the uniform susceptibility χ. 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 Δ from a fit of the low temperature behavior of the uniform susceptibility χ by 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 [2,3]. 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

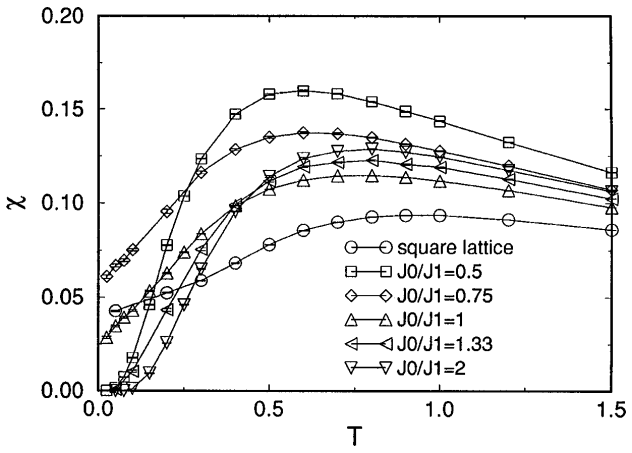

FIG. 3. Temperature dependence of the uniform susceptibility χ for different ratios of the couplings J_0/J_1. For each temperature the system size was taken large enough to see the value for the infinite system. The lowest temperatures were calculated on an $N = 512$ spin lattice. As a reference we have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

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,

$$m_s^2 = \langle\psi|\left[\frac{1}{N}\sum_{\mathbf{r}}\mathbf{S}_{\mathbf{r}}(-1)^{||\mathbf{r}||}\right]^2|\psi\rangle. \quad (1)$$

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 [14],

$$m_s(N) = m_s(\infty) + O\left(\frac{1}{\sqrt{N}}\right). \quad (2)$$

Figure 4 shows the system size dependence of m_s. Let us first discuss couplings in the spin gap regime. There the finite cluster results 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 $1/\sqrt{N}$ behavior of the gapped phase. A rough estimate shows an exponent of the order 0.5, as expected from the mapping to the nonlinear σ 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. 2) 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 $m_s = 0.3074(4)$ [16]. 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 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 $0.55 < \alpha_c^l < 0.65$ 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

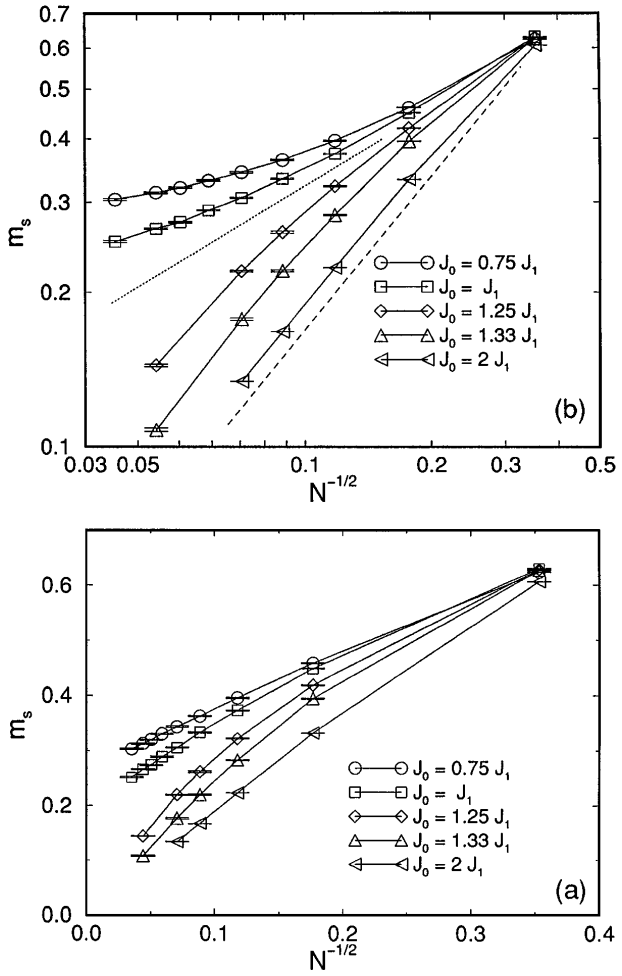

FIG. 4. System size dependence of the staggered magnetization m_s for different ratios of the couplings J_0/J_1. For each system size the temperature was chosen low enough to see the ground state properties. The largest systems contain $N = 800$ spins. (a) m_s plotted as a function of $N^{-1/2}$. A linear 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.

LRO sets in at $1.05 < \alpha_c^u < 1.11$. 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 [2], 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 [17].

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 [18]. 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 nonlinear σ model [15].

In comparison to experimental results on CaV_4O_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 stability of this plaquette RVB phase and the gap are greatly enhanced by a frustrating next nearest neighbor coupling. Thus we expect that already a quite small frustration will destroy LRO and can explain the substantial gap observed in CaV_4O_9.

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