Spin gap in CaV_4O_9: A large-S approach

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We study the stability of the four-site plaquette order in a model recently introduced by Katoh and Imada to explain the spin gap observed in CaV_4O_9. We show that all relevant types of order (four-site plaquette, dimerized, Néel) can be described within the framework of Schwinger-boson mean-field theory, and we make predictions regarding their relative stability. The results are compared to exact diagonalization of finite clusters.

In a recent paper, Taniguchi *et al.*[1] reported the presence of a spin gap in CaV_4O_9. The structure of the two-dimensional crystal of VO_4 pyramids of CaV_4O_9 is shown on Fig. 1. In the Mott insulating phase, one can consider Vanadium atoms as localized spin-1/2 coupled by superexchange interactions and, as pointed out by Katoh and Imada,[2] a minimal model consists of spin-1/2 antiferromagnetic Heisenberg model including an exchange term J between nearest-neighbor Vanadium atoms. These authors have shown using Monte Carlo simulations that this model has a spin gap. In order to determine the origin of the gap, they introduce a slightly more general Hamiltonian:

$$H=J\sum_{\langle ij\rangle}\vec{S}_i\cdot\vec{S}_j+J'\sum_{\langle ij\rangle'}\vec{S}_i\cdot\vec{S}_j, \tag{1}$$

where $\langle ij\rangle$ (resp. $\langle ij\rangle'$) means full line (resp. dashed line) links on Fig. 1. Using first-order perturbation theory in J'/J, they argue that the system keeps the same type of order as for $J'=0$, i.e., a product of the singlet on each plaquette.

In fact, introducing two links gives us a quantum spin model which might have interesting phase transitions: for $J\gg J'$ the system is gapped and the ground state is the product of the singlet state on each square plaquette, while for $J'\gg J$ the origin of the spin gap is quite different. The ground state is then the product of singlets on each interplaquette bond. The question that naturally arises concerns the way the system is passing from one type of order to the other while increasing J'/J. Another question that can also be asked is whether Néel long-range order can exist for $S=1/2$—the system is not frustrated—and, if this is the case, what is the nature of the transition between this ordered state and the previously mentioned dimerized states.

In this paper, our aim is to answer these questions using a large S approach. A linear spin-wave (LSWT) calculation was done very recently by Ueda *et al.*[3] This theory predicts that the system is Néel ordered for a large region of parameters: $0.25<J'/J<6$. As these authors say, such bounds seem to be incompatible with second-order perturbation theory in J'/J and J/J'. This could be explained by the tendency for LSWT to favor Néel order. Still, the perturbation theory is not expected to be reliable around $J'=J$ and the possibility of stabilizing Néel order in that region remains.

Another method to check the presence of Néel order in the context of a large S approach is to use the Schwinger-boson mean-field theory (SBMFT).[5] One advantage is that it is an improvement over LSWT because it includes higher-order corrections in a self-consistent way. But more importantly, this method is able to describe not only the Néel ordered phase but also the two types of valence bond order which are present in the limits $J\to 0$ and $J'\to 0$. This method starts from a representation of the spin algebra in terms of bosonic operators: $\vec{S}_i=\frac{1}{2}b^\dagger_{i\sigma}\vec{\sigma}_{\sigma\sigma'}b_{i\sigma'}$, the size of the spin being fixed by a constraint on the number of particles on

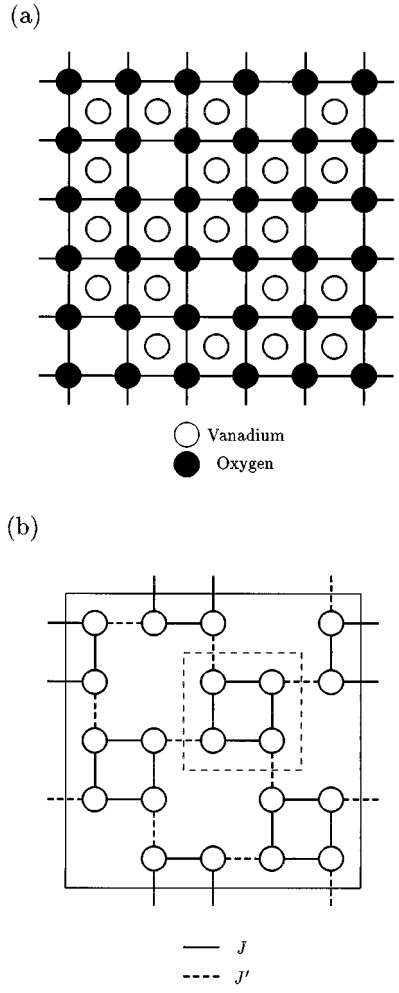

FIG. 1. (a) Schematic structure of the VO_4 pyramids plane. (b) The effective Heisenberg model.

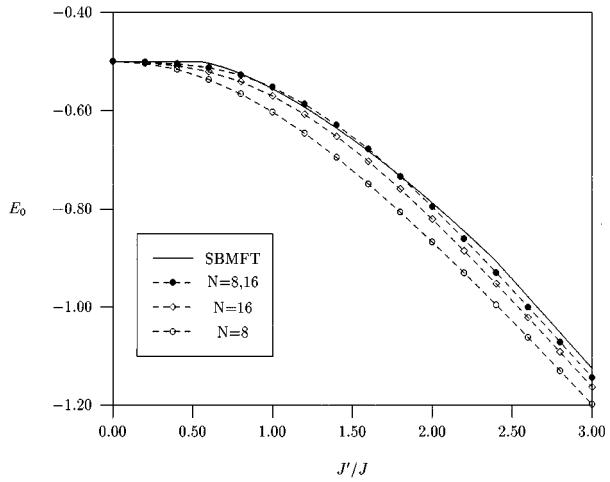

FIG. 2. Ground-state energy J'/J for $N=8$ (○) and $N=16$ (◇) clusters and values extrapolated for $N=\infty$ (●) together with SBMFT energy (full line).

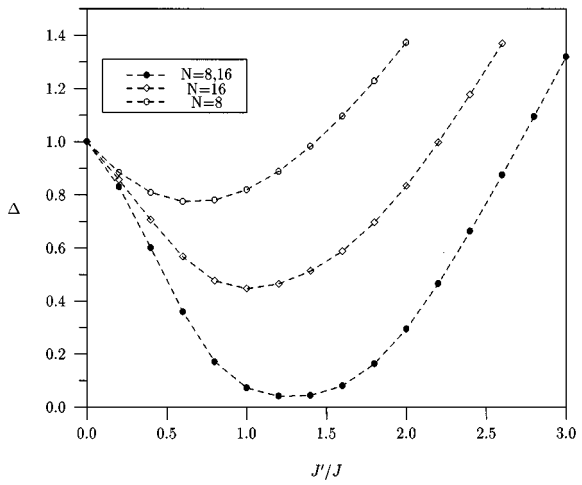

FIG. 4. Spin gap versus J'/J for $N=8$ (○) and $N=16$ (◇) clusters and values extrapolated for $N=\infty$ (●).

each site: $b^\dagger_{i\uparrow}b_{i\uparrow}+b^\dagger_{i\downarrow}b_{i\downarrow}=2S$. Defining operators that are quadratic in terms of the bosonic operators by $2B^\dagger_{ij}=b^\dagger_{i\uparrow}b_{j\uparrow}+b^\dagger_{i\downarrow}b_{j\downarrow}$ and $2A^\dagger_{ij}=b^\dagger_{i\uparrow}b^\dagger_{j\downarrow}-b^\dagger_{i\downarrow}b^\dagger_{j\uparrow}$, the Hamiltonian can be written

$$H=\sum_{(i,j)} J_{ij}(:B^\dagger_{ij}\,B_{ij}:-A^\dagger_{ij}\,A_{ij}). \tag{2}$$

At the mean-field level, one introduces the following order parameters: $\langle A^\dagger_{ij}\rangle=2\alpha_{ij}$ and $\langle B^\dagger_{ij}\rangle=2\beta_{ij}$ and the Hamiltonian is replaced with

$$H_{\mathrm{MF}}=\sum_{(i,j)} J_{ij}[\beta_{ij}(B_{ij}+B^\dagger_{ij})-\alpha_{ij}(A_{ij}+A^\dagger_{ij})-\beta^2_{ij}+\alpha^2_{ij}]. \tag{3}$$

Finally, the local constraint is replaced by a global one and is enforced only on the average through the addition to the Hamiltonian of a term $\mu\Sigma_i(b^\dagger_{i\uparrow}b_{i\uparrow}+b^\dagger_{i\downarrow}b_{i\downarrow}-2S)$, where the chemical potential μ plays the role of a Lagrange parameter. Because there are only antiferromagnetic-like correlations in this model, we have: $\langle B_{ij}\rangle=0$ for all pairs of spin (i,j).

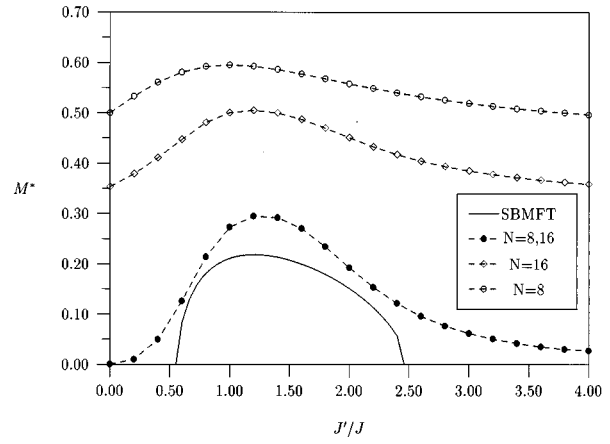

FIG. 3. Magnetization versus J'/J for $N=8$ (○) and $N=16$ (◇) clusters and values extrapolated for $N=\infty$ (●) together with SBMFT energy (full line).

These equations have two types of solutions: In the first one, the dispersion relation vanishes at three points in the Brillouin zone. This means that the bosons are condensed with a macroscopic expectation S^* for the occupation number of the zero-energy modes with the consequence that

$$\lim_{|i-j|\to\infty} (\vec{S}_i\cdot\vec{S}_j)=\pm S^{*2}. \tag{4}$$

The second kind of solution has a gap in the excitation spectra. There is then no long-range order. The mean-field equations have two remarkable solutions of this type: one has the same energy and the same correlation functions as the plaquette-singlet one, while the other has the same physical properties as the dimerized one.

The phase diagram of the model under investigation is represented in Fig. 3. At the SBMFT approximation, there is only one solution for a given value of J'/J. For $J'/J<0.6$ we find a gapped solution with an energy per site $-0.5\ J$: this solution has the same correlation functions as the plaquette singlet. At the critical value $J'/J\approx 0.6$ the gap closes. For $0.6<J'/J<2.4$ the system exhibits Néel long-range order (LRO). At $J'/J=2.4$ a gap opens again and the order corresponds to dimers on the J' bonds. The order parameters $\langle\alpha_{ij}\rangle$ are continuous through both transitions which shows they are second order. Note that this result is meaningful and not a mere consequence of the approximation used. For another model, using SBMFT, we found a first-order transition between LRO and valence bond order.[4] We have also done an exact diagonalization of small clusters in order to check our results and to have an estimation of the gap in the spin excitations. As there are four atoms per unit cell, one can only study square clusters having $4N$ atoms. Besides, if N is odd, periodic boundary conditions are frustrating. So the only tractable clusters have 8 and 16 spins. The next cluster (24 sites) does not have the symmetry of the square lattice. For these two clusters, we have calculated the ground-state energy, the spin gap Δ_N, and the staggered magnetization defined by $M^*=\langle\phi|(1/N\Sigma_i\epsilon_i S^z_i)^2|\phi\rangle$, where

$|\phi\rangle$ is the ground state of the system and $\epsilon_i = \pm 1$ depending on the orientation of the corresponding classical spin in the Néel state.

The different quantities can be scaled to their thermodynamic limit:[6]

$$\frac{E_N}{N} = E_0 + \frac{C_1}{N^{3/2}}, \tag{5}$$

$$\Delta_N = \Delta_0 + \frac{C_2}{N}, \tag{6}$$

$$M_N^* = M_0^* + \frac{C_3}{N^{1/2}}. \tag{7}$$

The results are displayed in Figs. 2–4. Taken literally, the results are inconsistent because the magnetization and the gap cannot be nonzero at the same time. But given the sizes we could look at, very small values of M^* or Δ are not accessible and only sizable values should be considered as meaningful. With this proviso, we find a qualitative agreement between exact diagonalization and SBMFT. The staggered magnetization we deduced from finite size extrapolation is rather large for $0.6 < J'/J < 2.4$ and comparable to the value calculated within SBMFT. In that region of parameters, the value of the extrapolated gap is small and reaches a minimum of 0.04 J at $J'/J = 1.25$. These two facts suggests that the SBMFT calculations are reliable, and that the system under investigation sustains Néel long-range order for $J'/J \approx 1.25$

To summarize, the plaquette singlet order proposed by Katoh and Imada to explain the magnetic properties of CaV_4O_9 can be found in the context of the $1/S$ approach (provided one goes beyond LSWT theory by using SBMFT[7]). This method enables one to study the stability of this type of order with respect to the switching on of a coupling J' between the plaquettes. The result is that Néel order is recovered in a sizable region around $J'/J \approx 1.25$, to disappear again when J'/J is large enough in favor of a dimerized order. It would be very interesting to see if Monte Carlo calculations of the gap sustain the presence of a gapless region.

At a more quantitative level, let us note that the predictions of SBMFT disagree with the results of Katoh and Imada who reported a gap of about 0.1 J for $J' = J$, while the SBMFT predicts the system to be already gapless for $J'/J \approx 0.6$. A possible explanation is that there is indeed an ordered phase but it is smaller than what SBMFT predicts and, in particular, does not include the point $J' = J$. Note, however, that contrary to Katoh and Imada's results, very recent Monte Carlo simulations by Troyer *et al.*[9] suggest that Néel order is present for the case $J' = J$. More work is needed to check that point. In any case, as far as CaV_4O_9 is concerned, this might be of minor importance. As pointed out by Sano and Takano[8] and Ueda *et al.*,[3] the plaquette order can be stabilized by introducing some frustration into the system.

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