Spin gap in CaV$_4$O$_9$: a large S approach

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Abstract

We study the stability of the 4-site plaquette order in a model recently introduced by Katoh and Imada to explain the spin gap observed in CaV$_4$O$_9$. We show that all relevant types of order (4-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 CaV₄O₉. The structure of the two dimensional crystal of VO₄ pyramids of CaV₄O₉ is shown on figure 1. In the Mott insulating phase, one can consider Vanadium atoms as localized spin one half 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 neighbors 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:

\[ H = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j + J' \sum_{\langle ij \rangle'} \vec{S}_i \cdot \vec{S}_j \]  

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

In fact, introducing two links gives us a new 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 inter-plaquette bonds. 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 Rapid Communication, our aim is to answer these questions using a large S approach. A linear spin wave (LSWT) calculation was done very recently by Ueda at al. [3]. This theory predicts that the system is Néel ordered for a large region of parameters: \( .25 < J'/J < 6 \). As these authors say, such the bounds seem to be incompatible with second order perturbation theory in \( J'/J \) and \( J/J' \). This could be explained by the tendency 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 a improvement over LSWT because it includes higher order corrections in a self-consistent way. But more importantly, this method is able to described 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: \( \vec{S}_i = \frac{1}{2} b_{i \sigma}^\dagger \vec{\sigma}_{\sigma \sigma'} b_{i \sigma'} \), the size of the spin being fixed by a constraint on the number of particles on each site: \( b_{i \uparrow}^\dagger b_{i \uparrow} + b_{i \downarrow}^\dagger b_{i \downarrow} = 2S \). Defining operators that are quadratic in terms of the bosonic operators by \( 2B_{ij}^\dagger = b_{i \uparrow}^\dagger b_{j \uparrow} + b_{i \downarrow}^\dagger b_{j \downarrow} \) and \( 2A_{ij}^\dagger = b_{i \uparrow}^\dagger b_{j \downarrow} - b_{i \downarrow}^\dagger b_{j \uparrow} \), the Hamiltonian can be written:

\[
H = \sum_{(i,j)} J_{ij} (B_{ij}^\dagger B_{ij} - A_{ij}^\dagger A_{ij}) \tag{2}
\]

At the mean-field level, one introduces the following order parameters: \( < A_{ij}^\dagger > = 2\alpha_{ij} \) and \( < B_{ij}^\dagger > = 2\beta_{ij} \) and the Hamiltonian is replaced with:

\[
H_{MF} = \sum_{(i,j)} J_{ij} (\beta_{ij} (B_{ij} + B_{ij}^\dagger) - \alpha_{ij} (A_{ij} + A_{ij}^\dagger) - \beta_{ij}^2 + \alpha_{ij}^2) \tag{3}
\]

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 \( \mu \sum_i \left( b_{i \uparrow}^\dagger b_{i \uparrow} + b_{i \downarrow}^\dagger b_{i \downarrow} - 2S \right) \), where the chemical potential \( \mu \) plays the role of a Lagrange parameter. Because there are only antiferromagnetic like correlations in this model, we have: \( < B_{ij}^\dagger > = 0 \) for all pairs of spin \((i, j)\).

These equations have two types of solution: In the first one, the dispersion relation is vanishing 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

\[
\lim_{|i-j| \to \infty} < \vec{S}_i, \vec{S}_j > = \pm S^*^2 \tag{4}
\]
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 as the same physical properties as the dimerized one.

The phase diagram of the model under investigation is represented on figure 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 $-0.5J$: 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. At $J'/J = 2.4$ a gap opens again and the order corresponds to dimers on the $J'$ bonds. The order parameters $<\alpha_{ij}>$ 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 exact diagonalisation 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) dose not have the symmetry of the square lattice. For these two clusters, we have calculated the ground state energy, the spin gap $\Delta_N$ and the staggered magnetization defined by $M^* = \langle \phi | (1/N \sum_i \epsilon_i S^z_i) | \phi \rangle$ 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]:

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

$$\Delta_N = \Delta_0 + \frac{C_2}{N}$$
\[ M_N^* = M_0^* + \frac{C_3}{N^{1/2}} \]  

(7)

The results are displayed on figures 2, 3 and 4. Taken literally, the results are inconsistent because the magnetization and the gap cannot be non zero at the same time. But given the sizes we could look at, very small values of \( M^* \) or \( \Delta \) are not accessible and only sizable values should considered as meaningful. With this proviso, we find a qualitative agreement between exact diagonalisation 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 \( 0.04J \) at \( J'/J = 1.25 \). This two facts suggests that the SBMFT calculation 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 \( \text{CaV}_4\text{O}_9 \) can be found in the context of \( 1/S \) approach provided one goes beyond LSW 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 \( 0.1J \) 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 then what SBMFT predicts, and in particular it 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 \( \text{CaV}_4\text{O}_9 \) is concerned, this might be of minor importance. As pointed out recently
by Sano et al. [8], the plaquette order can be stabilized by introducing some frustration into the system.

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FIGURES

FIG. 1. (a) Schematic structure of the VO$_4$ pyramids plane. (b) 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. 3. Magnetization versus $J'/J$ 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 $N = \infty$ (●)