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1996 J. Phys.: Condens. Matter 8 7917

(http://iopscience.iop.org/0953-8984/8/42/011)

J. Phys.: Condens. Matter **8** (1996) 7917–7923. Printed in the UK

Two-dimensional $S = 1/2$ antiferromagnet on a plaquette lattice

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Received 17 April 1996, in final form 22 July 1996

Abstract. We consider a simplified model of the magnetic structure of the two-dimensional compound CaV_4O_9 in terms of interacting square plaquettes of spins with two distinct antiferromagnetic exchange constants. We analyse the competition between two types of singlet ground states and the Neel ordered one in terms of, respectively, numerical cluster expansion and nonlinear spin wave theory. The resulting phase diagram agrees well with known quantum Monte Carlo results and suggests a first-order transition between ordered and singlet ground states as a function of the exchange constants.

The recent experimental observation of a spin gap in the layered $S = 1/2$ antiferromagnet CaV_4O_9 [1] has opened a new and interesting perspective in two-dimensional magnetism. We discuss the simplest model of the undoped structure that consists of a square lattice of elementary squares or 'plaquettes', which we will refer to as a CAVO lattice. The magnetic exchange energy within the plaquettes (J_0) and between the plaquettes (J_1) is given by

$$\widehat{H} = J_0 \sum_{\square} \boldsymbol{S}_i \boldsymbol{S}_j + J_1 \sum_{-} \boldsymbol{S}_i \boldsymbol{S}_j . \tag{1}$$

Here ij represent nearest neighbours on edges of a plaquette and between adjacent plaquettes, respectively. Additional (frustrating) couplings are ignored, although they are believed to be necessary for quantitative agreement with experiments.

The nature of the ground state is easy to understand in two limits [2]. In the limit of $J_1 \ll J_0$, the plaquettes form resonating valence-bond-type singlet states, with an energy of $-\frac{1}{2}J_0$ per plaquette, and weak bonds J_1 serve as a perturbation. In the opposite limit of $J_0 \ll J_1$ the interplaquette connections form singlets of energy $-\frac{3}{8}J_1$ per dimer that are weakly interacting via plaquettes. This construction is qualitatively symmetric, but the plaquettes are somewhat 'stronger', so that the critical 'equilibrium' region is centred at $J_1 \simeq \frac{4}{3}J_0$. In this region, in addition to these two quantum singlet phases, the antiferromagnetically ordered Neel state could also be competitive.

A variety of theoretical methods was applied to study the ground states of this model as a function of J_1/J_0 but the results remain contradictory. Our purpose is to compare the energies of three candidates for the ground state of model (1) and estimate the regions of their stability in terms of the ratio J_1/J_0. Unlike other approaches that attempted to treat the system within a unified framework for all J_1/J_0, we choose the most quantitatively reliable

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0953-8984/96/427917+07$19.50

approach for each individual phase. For the two singlet states we develop a numerical perturbation expansion (beyond the second order [2]) in the coupling ratio J_1/J_0 or J_0/J_1, whichever is smaller. The energy of the Neel state is estimated via the nonlinear spin wave approximation, which gives a lower ground state energy than the linear approximation reported in [3]. We also compare our results with ground state energies obtained by direct numerical diagonalization of the Hamiltonian for different finite lattices of up to 24 spins and with frustrating/nonfrustrating boundary conditions.

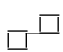
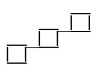
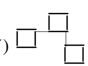
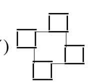
1. Cluster expansion for the ground state

The idea of the cluster expansion is quite general and works for any model with finite range interactions provided the unperturbed Hamiltonian is a sum commuting blocks. For the model under consideration and in both limits $J_1 \ll J_0$ and $J_0 \ll J_1$ the perturbation is a sum of exchange interactions between nearest neighbours, and the zero approximation (the first or second term in (1) respectively) is a sum over independent plaquettes or dimers. Thus in any given order n of the perturbation parameter (the smaller of the coupling ratios) the total correction to the ground state energy of an arbitrary lattice cluster c can be reorganized into a sum over connected graphs that mark the interactions that were used and the unperturbed blocks that were touched:

$$E_c^{(n)} = \sum_g N_{c,g} \varepsilon_g(n) \,. \tag{2}$$

Here $\varepsilon_g(n)$ stands for the contribution of the graph g in the order n and $N_{c,g}$ for the number of ways the graph g can be embedded in the cluster c. The same relation holds for the entire infinite lattice with properly normalized embedding numbers $N_\infty(g)$.

The detailed analysis is rather tedious (see e.g. similar analysis [4] and references therein), and we only list the resulting sequences of graphs and embedding numbers. To be specific, consider the J_1 expansion about the plaquette state, with four graphs contributing up to the fifth order

$1^{(II)}$  $2^{(IV)}$  $3^{(IV)}$  $4^{(IV)}$  . (3)

We draw the graphs as being embedded in the CAVO lattice, marking the J_0 connections by bold lines. The Roman superscript of a graph denotes the lowest order of perturbation in which it contributes to (2). A connected graph contributes to order n if n interactions can be placed on the links of the graph (one or more per link) such that (i) each block is touched at least twice (otherwise the block cannot return to its singlet ground state), and (ii) no two parts of the graph are connected by only one interaction (it would vanish by spin symmetry). Once the contributions $\varepsilon_g(n)$ of all necessary graphs are known, the nth-order corrections $E_c^{(n)}$ to the ground state energy of an arbitrary lattice cluster c can be calculated using the embedding numbers $N_{c,g}$. One can, however, invert the procedure and recover the contributions of the graphs by solving the system of linear equations (2) for $\varepsilon_g(n)$ in terms of the nth-order corrections $E_c^{(n)}$ of an appropriately selected set of small clusters c.

The most economic or 'optimal' set of clusters is obtained when each cluster embeds some graph in the list of graphs exactly once. Note that the graphs are topological entities, so that the choice of optimal clusters is generally not unique. Once a choice has been made, there is a one-to-one correspondence between the graphs contributing to a given order and the finite clusters carrying the information on their contributions. Therefore we can use the same pictures and labels for optimal clusters as for graphs. For the plaquette expansion the embedding numbers $N_{c,g}$ of all graphs of (3) in all clusters of (3) are

$$N_{c,g}^{pla} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 2 & 1 & 0 & 0 \\ 2 & 0 & 1 & 0 \\ 4 & 0 & 4 & 1 \end{pmatrix}$$

with rows of the matrix corresponding to clusters and columns corresponding to graphs.

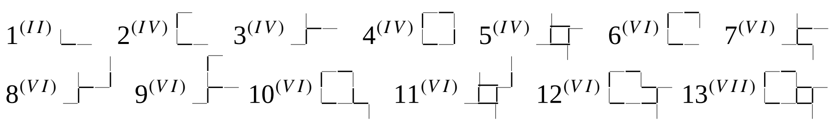
The coefficients $E_c^{(n)}$ are easily extracted from a polynomial fit of high-precision ground state energies of the clusters in the list at several values of the coupling ratio. Having solved for $\varepsilon_g(n)$ from (2) we calculate (to the same fifth order) the ground state energy per site of an infinite lattice with embedding constants $N_{\infty,g}^{pla} = \frac{1}{4}\,(2\,2\,4\,1)$ and obtain

$$E_{pla}(J_0, J_1) = J_0\Bigg[-\frac{1}{2} - \frac{43}{1152}\left(\frac{J_1}{J_0}\right)^2 - 0.00723\left(\frac{J_1}{J_0}\right)^3 - 0.00308\left(\frac{J_1}{J_0}\right)^4 - 0.0022\left(\frac{J_1}{J_0}\right)^5\Bigg] + \dots \qquad (4)$$

So we find five orders of the perturbation series for the *infinite* lattice by diagonalizing only four *finite* and relatively small clusters of up to 16 spins by use of conventional Lanczos algorithms.

In the case of the dimer expansion in the small parameter J_0/J_1 the unperturbed blocks are smaller (two sites), so that we can reach seventh order of perturbation with clusters not exceeding 12 sites. The list of graphs/clusters contains 13 entries

$1^{(II)}$ $2^{(IV)}$ $3^{(IV)}$ $4^{(IV)}$ $5^{(IV)}$ $6^{(VI)}$ $7^{(VI)}$ $8^{(VI)}$ $9^{(VI)}$ $10^{(VI)}$ $11^{(VI)}$ $12^{(VI)}$ $13^{(VII)}$



(5)

in the same notations as (3), with the following matrix of embedding numbers

$$N_{c,g}^{dim} = \begin{pmatrix} 1&0&0&0&0&0&0&0&0&0&0&0&0 \\ 2&1&0&0&0&0&0&0&0&0&0&0&0 \\ 2&0&1&0&0&0&0&0&0&0&0&0&0 \\ 4&4&0&1&0&4&0&0&0&0&0&0&0 \\ 4&0&4&0&1&0&4&0&0&0&0&0&0 \\ 3&2&0&0&0&1&0&0&0&0&0&0&0 \\ 3&0&2&0&0&0&1&0&0&0&0&0&0 \\ 3&1&1&0&0&0&0&1&0&0&0&0&0 \\ 3&2&1&0&0&0&0&0&1&0&0&0&0 \\ 5&5&1&1&0&5&0&1&1&1&0&0&0 \\ 5&2&4&0&1&0&4&2&1&0&1&0&0 \\ 6&4&2&0&0&3&1&2&0&0&0&1&0 \\ 7&6&4&1&1&6&4&4&2&2&2&1&1 \end{pmatrix}.$$

The embedding constants for the infinite lattice are $N_{\infty,g}^{dim} = \frac{1}{4}\,(4\,8\,4\,1\,1\,16\,4\,16\,8\,8\,8\,4\,4)$, and the ground state energy per site in the dimer expansion is

$$E_{dim}(J_0, J_1) = J_1\Bigg[-\frac{3}{8} - \frac{3}{32}\left(\frac{J_0}{J_1}\right)^2 - \frac{3}{128}\left(\frac{J_0}{J_1}\right)^3 - 0.02295\left(\frac{J_0}{J_1}\right)^4 - 0.0213\left(\frac{J_0}{J_1}\right)^5 - 0.0240\left(\frac{J_0}{J_1}\right)^6 - 0.0205\left(\frac{J_0}{J_1}\right)^7\Bigg] + \dots \qquad (6)$$

2. Nonlinear spin waves

We apply the conventional Holstein–Primakoff formalism, that parametrizes spins in terms of harmonic oscillators. This approach provides results of a remarkable precision for the $s = 1/2$ Heisenberg antiferromagnet on a square lattice (see [5] for review). The CAVO lattice is treated as a square lattice with nodes containing four spins. In the approximation of noninteracting spin waves, we find

$$E^{linear}_{Neel}(J_0, J_1) = -\left(J_0 + \frac{1}{2}J_1\right)\left[s(s+1) - \frac{s}{4}\int \mathrm{Tr}\left\{\sqrt{1-\gamma^2(\boldsymbol{p}, J_0, J_1)}\right\}\frac{\mathrm{d}^2 p}{(2\pi)^2}\right] \qquad (7)$$

where the integral in $\boldsymbol{p} = (p_1, p_2)$ is over the square $2\pi \times 2\pi$ Brillouin zone. This is analogous to the result for the simple square lattice except that $\gamma(\boldsymbol{p}, J_0, J_1)$ is now a 4×4 matrix with two exchange parameters

$$\gamma(\boldsymbol{p}, J_0, J_1) = \frac{1}{2J_0 + J_1}\begin{pmatrix} 0 & J_0 & J_1\mathrm{e}^{\mathrm{i}p_1} & J_0 \\ J_0 & 0 & J_0 & J_1\mathrm{e}^{\mathrm{i}p_2} \\ J_1\mathrm{e}^{-\mathrm{i}p_1} & J_0 & 0 & J_0 \\ J_0 & J_1\mathrm{e}^{-\mathrm{i}p_2} & J_0 & 0 \end{pmatrix}.$$

The next correction of order $O(s^0)$ is the average of quartic terms in the boson Hamiltonian in the linearly reconstructed ground state. Its direct evaluation is rather tedious (see [6]), but we have found a method to simplify the calculation. For bipartite lattices with two equivalent sublattices one can prove

$$\langle \boldsymbol{S}_a \boldsymbol{S}_b \rangle = (s + c_{ab})^2$$

for any pair of nearest-neighbour spins with c_{ab} being a constant of order s^0. In other words, every $\langle \boldsymbol{S}_a \boldsymbol{S}_b \rangle$ is a full square in the nonlinear spin wave approximation. Therefore, the nonlinear spin wave result may be found by (i) separating the contributions from different types of nearest neighbours, (ii) completing the square for each of them and (iii) summing up the results. Thus for the model (1) we arrive at

$$E_{Neel}(J_0, J_1) = -\left[J_0\left(s + \frac{1}{2} - C_0\right)^2 + \frac{1}{2}J_1\left(s + \frac{1}{2} - C_1\right)^2\right]$$

$$C_\alpha = \frac{1}{8}\int \mathrm{Tr}\left\{\frac{1 - \gamma(\boldsymbol{p}, J_0, J_1)\gamma_\alpha(\boldsymbol{p})}{\sqrt{1-\gamma^2(\boldsymbol{p}, J_0, J_1)}}\right\}\frac{\mathrm{d}^2 p}{(2\pi)^2} \qquad (8)$$

where $\gamma_\alpha(\boldsymbol{p})$ denotes $\gamma_0(\boldsymbol{p}) = \gamma(\boldsymbol{p}, 1, 0)$ or $\gamma_1(\boldsymbol{p}) = \gamma(\boldsymbol{p}, 0, 1)$. For the model under consideration $s = 1/2$ should be substituted.

3. Discussion

The results of our estimates for the infinite CAVO lattice are collected together in figure 1. In these figures we used the energy of the classical Neel state $E_{classical}(J_0, J_1) = -(2J_0 + J_1)/8$ as a unit and we plot the rescaled ground state energy $\widetilde{E} = E(J_0, J_1)/E_{classical}(J_0, J_1)$ as a function of the reduced coupling $\widetilde{J} = J_1/(J_0 + J_1)$ using notations that are similar to those adopted in [3]).

For each of the two perturbative expansion we plot the energy in all computed orders starting from the second. A tentative Padé extrapolation indicates closest singularities at respectively $J_1/J_0 \sim 1.4$ for the plaquette expansion and $J_1/J_0 \sim 1.05$ for the dimer one, but the orders we reached are not sufficient to reveal the analytical structure of the expansions.

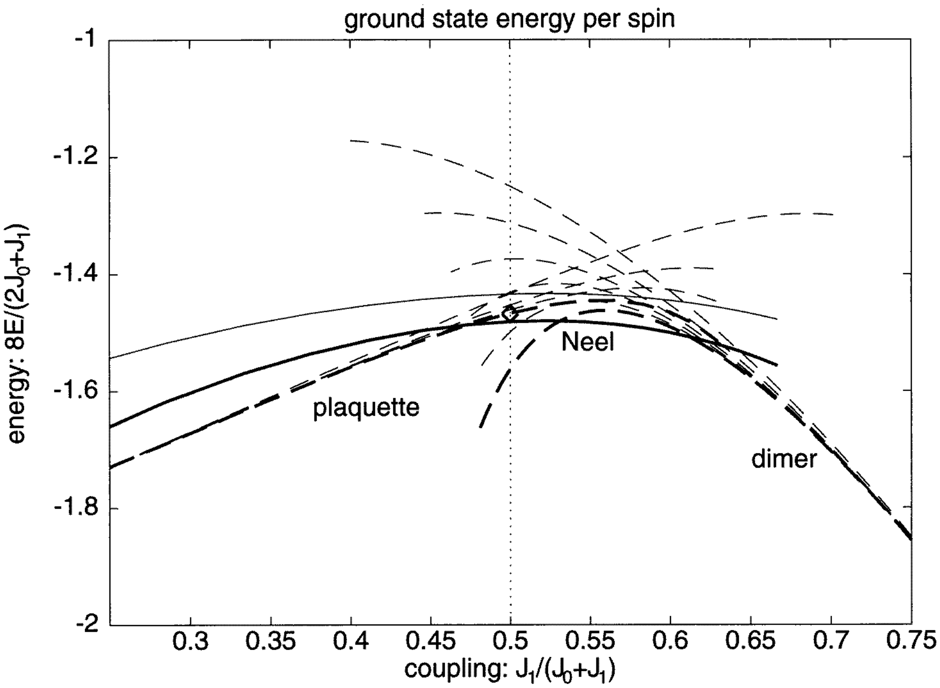


Figure 1. The rescaled ground state energy per spin $\widetilde{E}$ versus the reduced coupling $\widetilde{J}$. Thin and solid lines represent, respectively, the linear and nonlinear spin wave approximations (7) and (8). Dashed ascending and descending lines represent, respectively, the perturbation expansions for the plaquette (4) and dimer singlets (6), starting from the second order. The highest orders (fifth for plaquettes and seventh for dimers) are the bold dashes. The two (hardly distinguishable) diamonds are variational Monte Carlo results [7] and [8].

However, both expansions appear to converge well in their dominant region, and we believe that the highest order of perturbation provides a good estimate for the ground state energy. Another fact to be noted is that the ground state energy per site $E_{rvb}(J_0, J_0) = -0.5499J_0$ following from (4) at the symmetric point $J_0 = J_1$ agrees perfectly with variational Monte Carlo calculations of the singlet state that give $-0.5510J_0$ [7] $-0.5495J_0$ [8].

The precision of the $1/s$ expansion is generally less controlled than that of ordinary perturbation theories, as the small parameter is not obvious (see [5] for discussion). However, we see that at the symmetric point ($J_1 = J_0$) the first correction of order s (linear spin waves) gives $\simeq$ 45% of the classical Neel energy and the second, nonlinear correction, is $\sim$ 10% of the first one. Such a convergence is only a little worse than in the case of the square lattice where these ratios are 32% and 8%, respectively, and we expect negligible further corrections.

From figure 1 we see that each of the three states minimizes the energy in some region of J_1/J_0. Namely, the Neel state is stable in the interval $0.90 < J_1/J_0 < 1.6$, and the plaquette and dimer singlets are stable correspondingly below and above this region. This is in a perfect quantitative agreement with the Monte Carlo simulations [9], but not with the approximate treatments of [10] and [11], where the Neel interval is considerably overestimated.

Inspecting the convergence of the perturbation series in figure 1 we find it rather poor

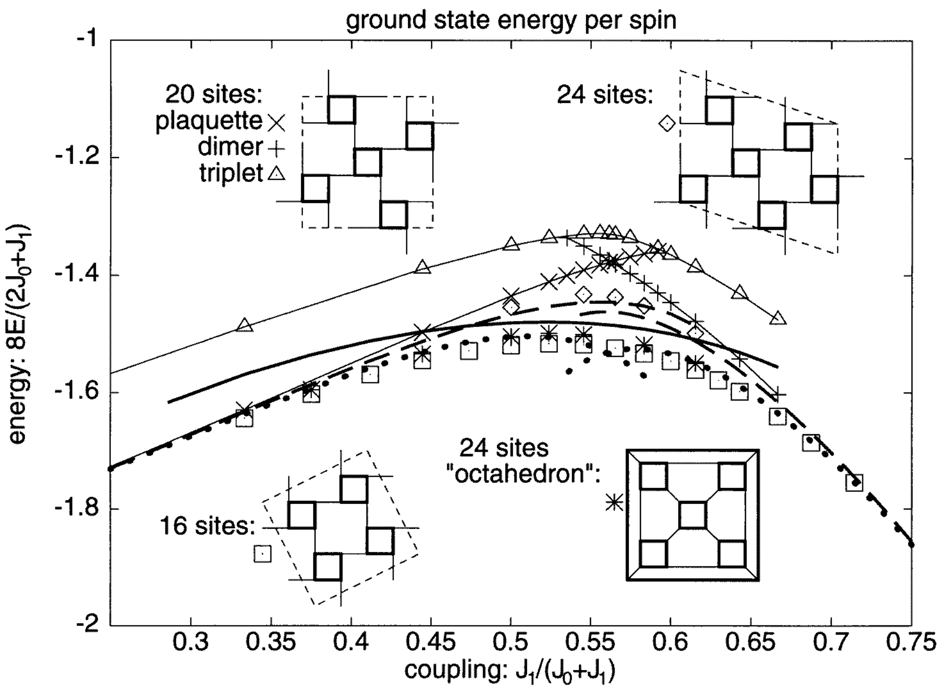


Figure 2. Energies of different finite clusters cut out from the CAVO lattice and that of an octahedron made up of plaquettes. Bold solid and dashed lines show the nonlinear spin wave and perturbative results on the infinite lattice. Dotted lines represent the perturbation expansion for the 24-site octahedron in the plaquette and dimer phase up to, respectively, fifth and sixth order.

in the intermediate region $J_1/J_0 \sim 1.2$, but quite reasonable near the visible borders of the Neel state. Neither of our three curves for the ground state energy shows an anomaly in the vicinity of the intersection point. Thus we conjecture the occurrence of discontinuous (first-order) transitions as a function of J_1/J_0, as a result of a direct competition in energy. A somewhat similar transition has been reported for a square lattice with additional frustrating couplings [12]. In fact, presently available results obtained by finite-temperature simulations or mean-field type approximations cannot exclude this scenario. It assumes that the gap does not vanish on the border of the singlet regions, in agreement with recent perturbative estimates [13] extrapolated to the point $J_1/J_0 = 0.9$.

On the other hand, the fact that we use entirely different approaches for each region does not allow us to insist on the first-order character of the transitions. The situation might be sensitive to minor quantitative changes due to higher-order correction, so that the intersecting curves could turn out to be tangents.

We found it interesting to complete the picture by direct Lanczos-type diagonalization of finite clusters. The results for four clusters and their configurations are presented in figure 2. Three clusters are cut out from the infinite lattice with periodic boundary conditions imposed. The cluster of four plaquettes is nonfrustrated, whereas those of five and six plaquettes are both frustrated. We have also considered another nonfrustrated cluster of six plaquettes designed ‘artificially’ as an octahedron with vertices decorated by plaquettes. The energies are rather scattered due to small cluster sizes, but we observe that frustrated clusters tend to follow the singlet perturbation curves, while nonfrustrated ones follow the energy of the Neel state. To verify our cluster expansion we have computed the perturbation expansion for the six plaquette octahedron (to fifth order in the plaquette phase and to sixth order in the dimer one). The results shown in figure 2 suggest that our perturbation series are sufficiently precise in the regions of interest.

The energy spectrum of the frustrated five plaquette cluster is rather unusual. Due to a special symmetry, two singlet states belonging to different representations intersect near $J_1 \approx 1.3 \cdot J_0$, while the lowest triplet state is continuous in J_1/J_0. Although this curious example may be quite special, we consider it as confirming the possibility of first-order transitions on the infinite CAVO lattice.

Acknowledgments

We are indebted to Laurent Levy (SNCI, Grenoble) for acquainting us with the CaV_4O_9 problem. SVM thanks Laboratoire de Physique Mathématique, Université Montpellier II for hospitality and access to computational facilities.

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