Exact ground and excited states of frustrated antiferromagnets on the CaV_4O_9 lattice

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Abstract

The experimental observation of a spin-gap in the antiferromagnetic spin-$\frac{1}{2}$ compound CaV_4O_9 has been attributed to the formation of a plaquette resonating-valence-bond (PRVB) state in the underlying 1/5-depleted square lattice. We construct a spin-$\frac{1}{2}$ model on the 1/5-depleted lattice for which the PRVB state can be shown to be the exact ground state in a particular parameter regime. In a subspace of this parameter regime, the first excited state can be calculated exactly. In a different parameter regime, the dimer state is shown to be the exact ground state.

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Recent discovery of a spin-gap in CaV_4O_9[1] has given rise to a number of studies[2, 3, 4, 5, 6, 7, 8] to understand the origin of the gap. The magnetic system can be described by the antiferromagnetic (AFM) Heisenberg Hamiltonian for spins of V^{4+} ions ($S = \frac{1}{2}$) on a 1/5-depleted square lattice. The depleted lattice is obtained from the original square lattice by removing $\frac{1}{5}$ of the lattice sites in a regular manner. The depleted lattice is a bipartite lattice which consists of 4-spin plaquettes connected by dimer bonds designated as A and B respectively in Fig. 1.

The minimal model to describe the system is the Heisenberg Hamiltonian

$$H = \sum_{n.n.} J_{nn}\, \vec{S}_i \cdot \vec{S}_j$$

for nearest-neighbour (n.n.) spin-spin interactions. The exchange integral J_{nn} equals to $J_0\,(J_1)$ for plaquette (dimer) bonds. Two limiting cases that

can be considered are: (i) $J_0 \gg J_1$ and (ii) $J_1 \gg J_0$. In the first case, the ground state is the plaquette-resonating-valence-bond (PRVB) state in which the spin configuration in each plaquette is of the RVB type. In the other limit, the ground state consists of singlets (dimers) along the dimer bonds. In both the cases the ground state is a singlet and spin-disordered. Also, the spin-gap has a non-zero value. As pointed out by Ueda et al [2], the wave function of the dimer phase is orthogonal to that of the PRVB phase. The reason is that the two wave functions have different symmetry properties under reflection with respect to the dimer bonds: odd for the dimer state and even for the PRVB state. Thus the two states cannot be continuously connected by changing the parameters J_0 and J_1. An intermediate phase is expected to occur between the dimer and PRVB phases. This phase has been shown[6] to have long range Néel-type order and includes the isotropic coupling case $J_0 = J_1$. The spin-gap defined to be the difference in the energies of the lowest triplet excited state and the ground state, vanishes in the Néel ordered phase. Thus isotropic or near-isotropic n.n. coupling cannot explain the origin of the spin-gap.

Spin-gap has been suggested to occur when $J_0 \neq J_1$ or due to the inclusion of frustrated next-nearest-neighbour (n.n.n.) couplings[2]. It has been further suggested[7] that the spin-Peierls mechanism in the CaV_4O_9 lattice strengthens the plaquette bonds with respect to the inter-plaquette bonds. Taking these suggestions into account, we construct an AFM Heisenberg Hamiltonian on the 1/5-depleted lattice for which the exact ground states in different parameter regimes can be determined. These ground states include both the RVB and dimer states. In addition, a number of excited states are determined exactly.

The Hamiltonian includes the following types of interactions: n.n. plaquette, plaquette diagonal, dimer bonds connecting plaquettes, next-nearest-neighbour (n.n.n.), diagonal bonds connecting plaquettes and Knight's-move-distance-away (KM) interactions of strength J_1, J_2, J_3, J_4, J_5 and J_6 respectively. The pairwise interactions are illustrated in Fig. 2. The n.n.n. bonds connecting the mid-points of the big squares have strength zero. Similarly, the KM bonds which lie outside plaquettes have strength zero. The unequal interaction strengths are possible due to the spin-Peierls mechanism. The

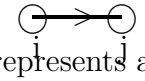
exchange interaction Hamiltonian is given by

$$H_{AF} = \sum_{<ij>} J_{ij}\, \vec{S}_i \cdot \vec{S}_j \tag{1}$$

For the ratio of interaction strengths

$$J_1 : J_2 : J_3 : J_4 : J_5 : J_6 = 1 : \beta : \alpha : \frac{\alpha}{2} : \frac{\alpha}{2} : \frac{\alpha}{2} \tag{2}$$

the PRVB state can be shown to be the exact ground state, when $\beta \leq 1$ and $\alpha \leq .5$.

The PRVB state has a RVB spin configuration in each plaquette. The RVB configuration is a linear superposition of two valence bond (VB) configurations (Fig. 3). A valence bond is a spin singlet and is represented by a solid line in the Figure. The arrow on the solid line specifies the ordering of spins in the singlet, e.g., 

denotes the spin configuration $\frac{1}{\sqrt{2}}\,(\alpha(i)\,\beta(j) - \beta(i)\,\alpha(j))$; α represents an up-spin and β a down-spin, i and j are the lattice sites. The PRVB state is further an exact eigenstate of H_{AF} as can be easily verified by using any one of the following spin identities

$$\vec{S}_n \cdot (\,\vec{S}_l + \vec{S}_m\,)\,[lm] \equiv 0 \tag{3a}$$

$$\vec{S}_n \cdot (\,\vec{S}_l + \vec{S}_m + \vec{S}_p + \vec{S}_q\,)[lmpq] \equiv 0 \tag{3b}$$

where [lm] denotes the spin singlet associated with a bond lm and [lmpq] is the singlet ground state (Fig. 3) of a plaquette lmpq of four spins. Each plaquette is part of two octahedra as shown in Fig. 4. The sites i, j, k, o, n, p form one octahedron and the sites l, j, k, o, n, m form the other octahedron (the lines of the octahedra are not joined for clarity). When the plaquette is in a RVB state, as shown in Fig. 3, the non-plaquette interactions of the octahedra, corresponding to the bonds ij, ik, in, io, po, pn, pk, pj and lk, lo, ln, lj, mn, mj, mk, mo give zero contribution to the energy because of the spin identities in (3). The PRVB state is thus the exact eigenstate of H_{AF}. The exact energy per plaquette is $-2\,J_1 + \frac{J_2}{2}$. To prove that the exact eigenstate is also the exact ground state, we make use of the method of 'divide and conquer' [10]. Let E_1 and E_g be the exact eigenstate and ground state energies. Thus $E_g \leq E_1$. Let Ψ_g be the exact ground state wave function. The Hamiltonian H_{AF} is divided into sub-Hamiltonians H_i's describing the

octahedra of interactions, $H_{AF} = \sum_i H_i$. Let E_{ig} be the ground state energy of H_i.

For an octahedron, let J'_1, J'_2, J'_3, J'_4, J'_5, and J'_6 be the strengths of the various interactions. The primed symbols denote the same types of interactions as the unprimed symbols do. Let $J'_2 = \beta\, J'_1$ and $J'_3 = J'_4 = J'_5 = J'_6 = \alpha\, J'_1$. When $\beta \leq 1$ and $\alpha \leq \frac{1}{2}$, the exact ground state of the octahedron consists of the RVB state of Fig. 3 in the plaquette and free vertex spins which can be either up or down. The exact ground state energy $E_{ig} = -2\, J'_1 + \frac{J'_2}{2}$. When the sub-Hamiltonians H_i's are added, all the plaquette bonds and the dimer bonds connecting the plaquettes are counted twice. So, $J_1 = 2\, J'_1$, $J_2 = 2\, J'_2$, $J_3 = 2\, J'_3$, $J_4 = J'_4$, $J_5 = J'_5$ and $J_6 = J'_6$. Using the variational theorem,

$$\begin{aligned} E_g &= \langle\, \Psi_g \,|\, H_{AF} \,|\, \Psi_g \rangle = \sum_i \langle\, \Psi_g \,|\, H_i \,|\, \Psi_g \,\rangle \\ &\geq \sum_i E_{ig} \geq N_p\,(\, -4\, J'_1 + J'_2\,) = N_p\,(\, -2\, J_1 + \frac{J_2}{2}\,) \end{aligned}$$

Where N_p is the number of the plaquettes in the lattice. The PRVB state which is the exact eigenstate of the H_{AF} has the energy $E_1 = N_p\,(\, -2\, J_1 + \frac{J_2}{2}\,)$. Thus in $\sum_i E_{ig} \leq E_g \leq E_1$ the lower bound $\sum_i E_{ig}$ has the same value as the upper bound E_1. In other words, the exact PRVB eigenstate of H_{AF} is the exact ground state with the ratio of interaction strengths $J_1 : J_2 : J_3 : J_4 : J_5 : J_6 = 2\, J'_1 : 2\,\beta\, J'_1 : 2\,\alpha\, J'_1 : \alpha\, J'_1 : \alpha\, J'_1 : \alpha\, J'_1$ which reduces to the ratio given in (2).

A RVB state similar to that shown in Fig. 3 is obtained by replacing the '+' sign by a '−' sign. We designate this new RVB state as Ψ_{RVB1} with the original RVB state labeled as Ψ_{RVB}. We construct a new state PRVB1 (total spin $S = 0$) for the 1/5-depleted lattice in which all the plaquettes, except one, have spin configuration corresponding to Ψ_{RVB}. In one plaquette, the spin configuration corresponds to Ψ_{RVB1}. For the ratio of interaction strengths given in (2) and when

$$\beta \geq \alpha + 0.5 \qquad (4)$$

the PRVB1 state can be shown to be exact first excited state of H_{AF}.

As in the case of the ground state, the proof can be obtained by means of the spin identity in (3) and using the method of 'divide and conquer'.

Exact diagonalisation of an octahedron corresponding to a sub-Hamiltonian H_i, shows that Ψ_{RVB} is the ground state and Ψ_{RVB1} the first excited state when $\beta \leq 1$, $\alpha \leq \frac{1}{2}$ and the restriction in (4) is satisfied. The rest of the proof is similar to that for the ground state. The energy of the first excited state of H_{AF} is $E_f = -\frac{3\,J_2}{2} + (N_p - 1)(-2\,J_1 + \frac{J_2}{2})$. The energy of the ground state is $E_g = N_p\,(-2\,J_1 + \frac{J_2}{2})$. Then the energy difference D is

$$D = E_f - E_g = 2\,J_1 - 2\,J_2 \qquad (5)$$

Evidence of a $S = 0$ first excited state has been obtained earlier in the numerical calculations of[11].

The PRVB1 state is N_p-fold degenerate as the plaquette, in which the spin configuration is Ψ_{RVB1}, can be any one of the N_p plaquettes. Other excited states can be constructed in which r ($r = 2, 3, 4, ...$ etc.) plaquettes have the spin configuration Ψ_{RVB1}. In all the other $(N_p - r)$ plaquettes the spin configuration is Ψ_{RVB}.

We now consider the parameter regime in which the dimer state is the exact ground state. The ground state consists of singlets along the dimer bonds. It can be shown that the dimer state is the exact ground state when the ratio of interaction strengths is

$$J_1 : J_2 : J_3 : J_4 : J_5 : J_6 = \frac{\gamma}{3} : \frac{\gamma}{3} : 1 : \frac{\gamma}{6} : \frac{\gamma}{6} : \frac{\gamma}{6} \qquad (6)$$

where $\gamma \leq 1$. The method of proof is the same as in the case when the PRVB state is the exact ground state. The dimer state can be shown to be an exact eigenstate by making use of the spin identity (3).

In the dimer ground state, the energy associated with each dimer bond is $-\frac{3\,J_3}{4}$. While using the method of 'divide and conquer', the Hamiltonian H_{AF} is sub-divided into Hamiltonian H_i's, each H_i describing a triangle of spins. The triangle consists of a dimer bond (strength J_3') and two other bonds (strength $\gamma\,J_3'$). For $\gamma \leq 1$, the ground state of the triangle is a singlet along the dimer bond. Each dimer bond connects two plaquettes and each plaquette contributes three sub-Hamiltonians which include the dimer bond in question. For example, in Fig. 4, the plaquette jkon contributes three sub-Hamiltonians corresponding to the triangle jmn, kmn and omn, all of which include the dimer bond mn. When adding all the H_i's, each dimer bond is counted six times, the plaquette bonds are counted twice and

the other bonds are counted once. This leads to the ratio of interaction strengths given in (6).

We now discuss the relevance of our model for understanding the properties of the experimental system CaV_4O_9. Earlier theoretical studies [2, 3, 4, 5, 6, 7] have explained the experimentally-observed spin gap by assuming CaV_4O_9 to be in a PRVB phase. Our model AFM Hamiltonian shows that in a particular parameter regime the PRVB state is the exact ground state on the 1/5-depleted square lattice. Our model includes only two more further-neighour interactions (n.n.n. and KM) than those considered in the earlier papers. We have assigned different interaction strengths to the intra-plaquette and inter-plaquette bonds. This is not unrealistic in view of the possibility that the spin-Peierls mechanism is at work in CaV_4O_9 [7, 9]. The ratio of interaction strengths for which the exact ground and first excited states can be determined corresponds to a region, rather than a single point, in parameter space. More experiments are, however, needed to settle the question of how many exchange parameters are needed to give a realistic description of CaV_4O_9.

As already mentioned, Albrecht et al[11] find the existence of singlet states in the spin gap, from the results of exact diagonalisation of finite systems. In the parameter regime given in Eq.(4), the PRVB1 state which is a singlet (S=0) has energy lower than that of the first triplet excited state. This state thus constitutes an in-gap state. Experimental signature of this state can be obtained from Raman scattering experiments. The fact that the singlet excitation is the lowest excited state is due to the presence of further-neighbour interactions. In this case, there should be an asymmetry

$$\Delta(k_x,\, k_y) \neq \Delta(k_y,\, k_x)$$

in the spin-gap at the wave vector $\vec{k} = (k_x,\, k_y)$. This asymmetry can be detected in neutron scattering experiments. There is so far no theory which can give a good fit to the experimental data of the susceptibility over the whole temperature range of 20 K to 700 K. Two possible reasons that have been suggested are: (i) inclusion of more terms in the Hamiltonian besides n.n. and diagonal and (ii) the dependence of the exchange constants on temperature due to spin-phonon coupling. Our model Hamiltonian includes more interaction terms than in earlier models. A theoretical calculation[12] on the temperature dependence of susceptibility is in progress and the results will be published elswhere.

In summary, we have constructed a $S = \frac{1}{2}$ AFM Heisenberg model on the 1/5-depleted lattice for which the PRVB and dimer states are the exact ground states in different parameter regimes. Both these states are singlets and spin-disordered. In the parameter regime in which the PRVB state is the exact ground state, an added restriction on the parameter values, makes it possible to obtain the exact first excited state. The relevance of the model to understand the physical properties of CaV_4O_9 is not well-established. The model, however, can serve as a toy model to study quantum phase transition from the PRVB to the dimer phase and also can be a starting point for studies of more realistic model Hamiltonians.

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Figure Captions

Fig. 1 The 1/5-depleted lattice of CaV_4O_9. A and B represent four-spin plaquettes and dimer bonds connecting plaquettes, respectively.

Fig. 2 The n.n. plaquette, plaquette diagonal, dimer bonds connecting plaquettes, next-nearest-neighbour (n.n.n.), diagonal bonds connecting plaquettes and Knight's-move-distance-away (KM) interactions of strengths J_1, J_2, J_3, J_4, J_5 and J_6 respectively. For clarity, the non-nearest-neighbour bonds are drawn singly.

Fig. 3 The RVB spin configuration in each four-spin plaquette in the PRVB state. The solid line denotes a singlet. The arrow sign indicates the ordering of spins in a singlet; i, j, k and l are the lattice sites.

Fig. 4 The plaquette okjn as part of two octahedra. The vertices i, l, p and m are connected to all the sites of the plaquette. Many of these connections are not shown in the Figure for clarity.

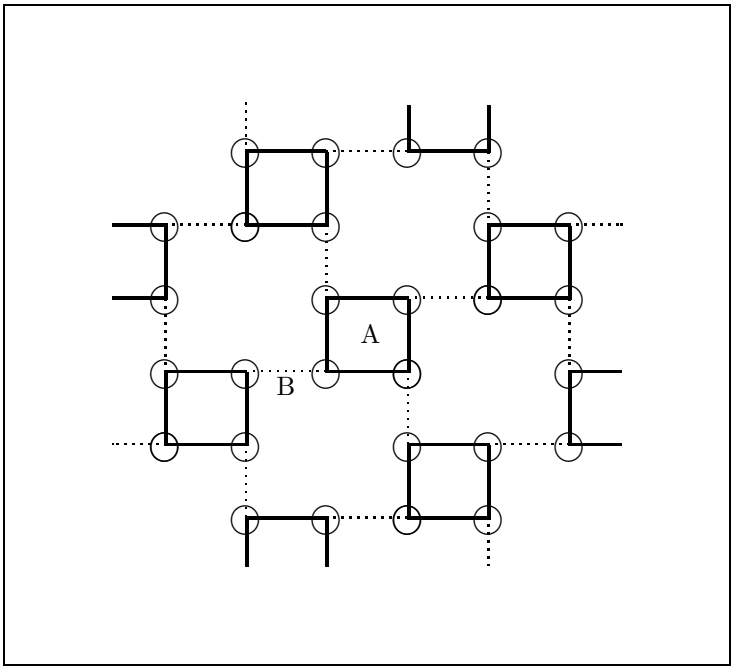

Fig. 1

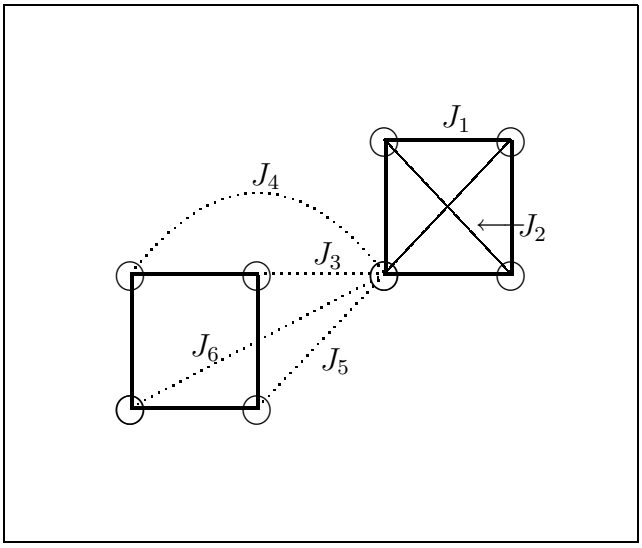

Fig. 2

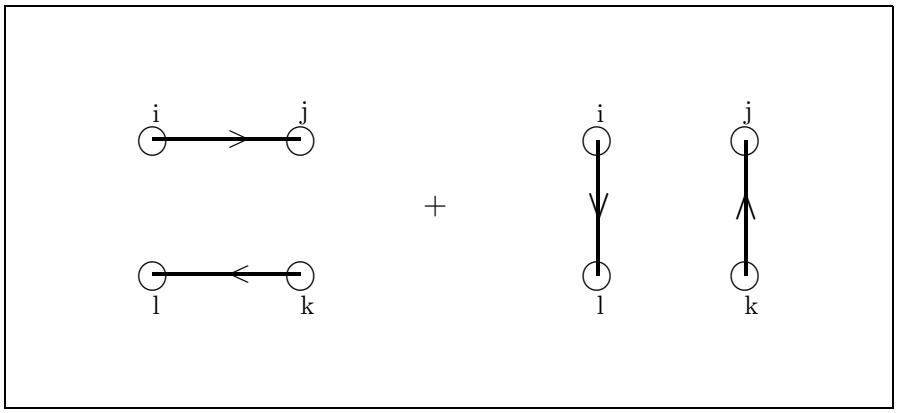

Fig. 3

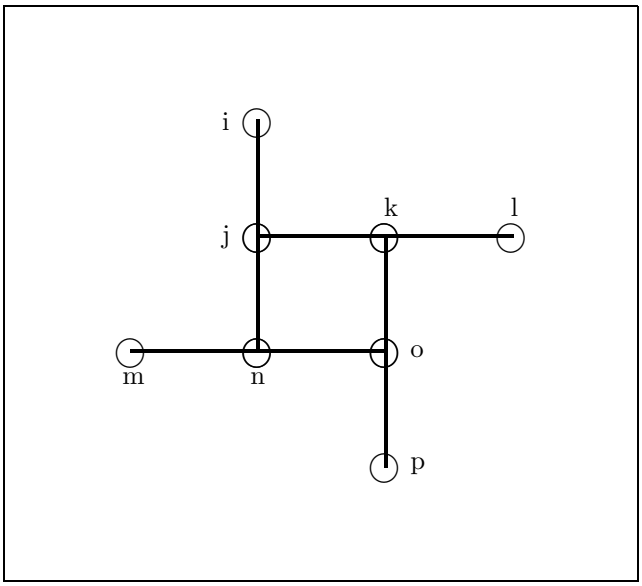

Fig. 4