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Spontaneous trimerization in two-dimensional antiferromagnets

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Abstract

In this paper, we propose an exotic quantum paramagnetic state in two-dimensional antiferromagnets—the spontaneous trimer state—which is the direct product state of the trimers of spins. Each trimer is a singlet state formed by three neighboring spins with $SU(3)$ symmetry. A frustrated spin-1 Heisenberg model in the kagome lattice is investigated. By analogy to the pseudo-potential approach in the fractional quantum Hall effect (FQHE), we find that the trimer state provides a good description for the exact ground state of this model. Other interesting properties, such as the local excitations as well as magnetization plateaus have also been investigated.

(Some figures in this article are in colour only in the electronic version)

The nature of quantum paramagnetic phases of two-dimensional (2D) antiferromagnetic systems has attracted considerable attention in the past 20 years because of its potential association with the physics of the cuprate superconductors [1]. Because of the absence of long-range order in the quantum paramagnetic phase, a central problem is the classification of the phases and critical points. According to their symmetries, we could classify these phases into two classes. The first kind is known as the 'spin liquid state', which restores the $SU(2)$ symmetry of the Néel state and does not break any symmetry of the original Hamiltonian. One of the well-known state in this class is the resonating valence-bond (RVB) state [2], or the 'spin liquid' state [3].

On the other hand, Read and Sachdev have presented another possibility [4] based on the Schwinger boson analysis of the $SU(N)$ quantum antiferromagnets. Their key point is that the condensation of the instantons with the Berry phase leads to the spontaneous breaking of the spatial symmetry (translational or rotational symmetry) of the original Hamiltonian. Similar to the solid, this state possesses a short-range order and thus is nominated as the valence-bond solid (VBS) state. The structure of the VBS state depends on $2S$ (mod 4), where S is the spin of the $SU(N)$ model. For the usual spin $1/2$ systems, the corresponding VBS state is known as the 'dimer state', where two nearest spins form a singlet or a dimer: $(\uparrow\downarrow - \downarrow\uparrow)/\sqrt{2}$ and the overall state is the direct product state of all those dimers.

The dimer state was first proposed as the exact ground state for the Majumdar–Ghosh (MG) model [5], which is an antiferromagnetic spin chain with nearest and next nearest neighbor interactions. For a particular ratio of those exchange interactions, the model is exactly solvable and has a two-foldly degenerate dimer ground state. Inspired by the exact solution of the MG model, Shastry and Sutherland proposed a 2D model [6] known as the Shastry–Sutherland (SS) model, whose ground state is the exact dimer state in two dimensions. It has been used in understanding the physical properties of $SrCu_3(BO_3)_2$ [7], which is topologically equivalent to the SS model.

Most of the above research focused on the spin-$1/2$ systems, however, as to the spin-1 strongly correlated systems, more and more interesting novel quantum paramagnetic phases have been discovered. One of the well-known examples is the Affleck–Kennedy–Lieb–Tasaki (AKLT) state [8], which breaks no symmetry of the Hamiltonian and is believed to provide a good description for the ground state of the pure 1D spin-1 Heisenberg chain [9, 10]. As to the 2D case, the situation is apparently more complex, the VBS state in the spin-1 quantum paramagnetic phase could be either the dimer state [11] that spontaneously breaks the translational symmetry, or a generalized AKLT state [10] that only breaks the rotational symmetry.

In this paper, we present an exotic VBS state called the 'trimer state' for the spin-1 system, which is an analogue to its counterpart in a spin-$1/2$ system: the dimer state. Such a state is a direct product state of trimer singlets and each spin singlet is formed by three spins with the total spin zero. Inspired by

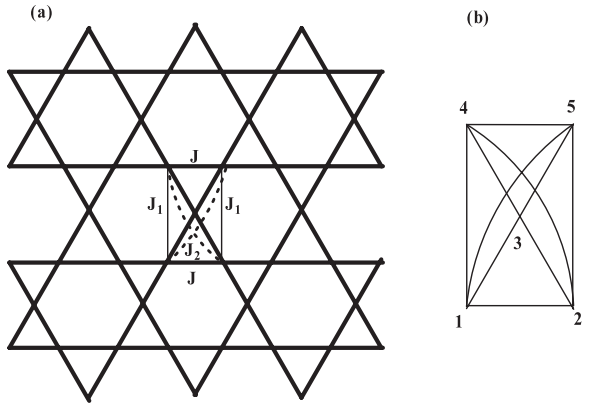

Figure 1. (a) The structure of the frustrated Heisenberg model in the kagome lattice, with the nearest coupling J, the second nearest coupling J_1, and the third nearest coupling J_2. (b) One block consists of five sites and each one equally couples with the others.

the seminal work of [9], we construct a frustrated Heisenberg model in a kagome lattice, and find that at a particular ratio of the different couplings, the 'trimer state' provides a very good approximation for the exact ground state of this model, which means that the wavefunction overlap between the trimer state and the exact ground state of this model is very close to 1. Similar states have been discussed in an $SU(3)$ spin tetrahedron chain [12–15] and in the kagome lattice with distorted coupling [16]. To our knowledge, we provide the first example in an isotropic 2D $SU(2)$ antiferromagnet with uniform coupling, where the spins are spontaneously trimerized. Then we investigate the excited properties and the magnetization plateaus in the applied magnetic field. It is believed that this state represents a class of the VBS state: N neighboring spins cluster together to form a singlet with $SU(N)$ symmetry and spontaneously break the lattice symmetry (translational or rotational). Within a cluster, each spin is maximally entangled with other spins. We call this kind of state a 'cluster state' while the usual dimer state is the simplest case.

The ground state of the quantum antiferromagnetic Heisenberg model on the kagome lattice has been intensively investigated by various methods, but is still far from being totally understood even for the spin-1/2 case. Recently, Laws *et al* found a Ni^{2+}-based material: $Ni_3V_2O_8$ [17], which is topologically equivalent to a frustrated spin-1 Heisenberg model in a kagome lattice. The spin-1 model on the kagome lattice has been investigated in several works [18–21], but most of them focus on the uniaxial anisotropic case. Here we focus on the $SU(2)$ case and introduce the second nearest $\langle\langle \mathbf{ij} \rangle\rangle$ and the third nearest $[\mathbf{ij}]$ couplings J_1 and J_2. Explicitly, the Hamiltonian for this model is given by

$$H = J \sum_{\langle \mathbf{i},\mathbf{j} \rangle} \mathbf{S_i} \cdot \mathbf{S_j} + J_1 \sum_{\langle\langle \mathbf{i},\mathbf{j} \rangle\rangle} \mathbf{S_i} \cdot \mathbf{S_j} + J_2 \sum_{[\mathbf{i},\mathbf{j}]} \mathbf{S_i} \cdot \mathbf{S_j}, \qquad (1)$$

with the structure schematically shown in figure 1.

In this paper we focus on the point $J = 3J_1 = 3J_2$ and investigate the ground state at this point by the 'pseudo-potential approach', which was first used to get the famous Laughlin wavefunction in the FQHE [22]. The analogy between the Heisenberg model and the FQHE was first introduced by Arovas *et al* [9] in the 1D case, where the Hamiltonian of a spin-1 antiferromagnetic Heisenberg model is decomposed into the summation of projection operators

$$H_1 = \sum_i \mathbf{S_i} \cdot \mathbf{S_{i+1}} = \sum_i [3P^2(i, i+1) + P^1(i, i+1) - 2]. \quad (2)$$

Observing that the AKLT state [8] is the exact ground state of the first part of the summation of projection operators $\sum_i 3P^2(i, i+1)$, where $P^S(i, i+1)$ projects the spin state of the bond $(i, i+1)$ onto the subspace with total spin S. They took it as a trial ground state of the spin-1 Heisenberg chain and consider the second part $\sum_i P^1(i, i+1)$ as a perturbation. It turns out that the AKLT state is a very good approximation of the exact ground state of equation (2) and numerical results show that the difference between the ground state energies of these two states is within 5%. Recently, we generalized this method to the 2D spin-1 $J_1 - J_2$ antiferromagnetic model [10], and found that at the maximal frustrated point $(J_1 = 2J_2)$, the ground state could be described by a two-fold 2D generalized AKLT state, which completely agrees with the general prediction from field theory [4], and was verified by the 2D Density Matrix Renormalization Group (DMRG) result [23].

Next we use a similar method to investigate the ground state of equation (1). Notice that at the point $J = 3J_1 = 3J_2$, equation (1) could be rewritten as the sum of identical blocks $\mathfrak{B}_\alpha$, as shown in figure 1(b). Each block is constructed of five spins and every spin is coupled to the other four spins identically, thus we have

$$H/J_1 = \sum_\alpha \mathfrak{B}_\alpha \qquad \text{with } \mathfrak{B}_\alpha = \sum_{\mathbf{i},\mathbf{j} \in \alpha} \mathbf{S_i} \cdot \mathbf{S_j} = \tfrac{1}{2}\mathbf{S}_\alpha^2 - 5 \quad (3)$$

where α denotes the αth block, and $\mathbf{S}_\alpha = \sum_{i \in \alpha} \mathbf{S_i}$ is the total spin operator of the five spins within the αth block. Then we expand this block Hamiltonian by the projection operators of the total spin in a block in terms of the projection operators:

$$\mathfrak{B}_\alpha = \sum_{S=0}^{5} C_S \mathbf{P}_\alpha^{\mathbf{S}} - 5 \qquad \text{with } C_s = S(S+1)/2. \qquad (4)$$

The operator $\mathbf{P}_\alpha^S$ projects the spin state of the αth block onto the subspace with total spin S: $\mathbf{P}_\alpha^S |S'\rangle = \delta_{S,S'}$. $\mathbf{P}_\alpha^S$ is defined as:

$$\mathbf{P}_\alpha^S = \prod_{i \neq S} \frac{\mathbf{S}^2 - i(i+1)}{S(S+1) - i(i+1)}. \qquad (5)$$

To make progress, we further decompose it into two parts

$$\mathfrak{B}_\alpha = \mathfrak{B}_\alpha^0 + \mathfrak{B}_\alpha^1,$$

with $\mathfrak{B}_\alpha^0 = 15\mathbf{P}_\alpha^5 + 10\mathbf{P}_\alpha^4 + 6\mathbf{P}_\alpha^3 - 5$ and $\mathfrak{B}_\alpha^1 = 3\mathbf{P}_\alpha^2 + \mathbf{P}_\alpha^1$. Dividing the original Hamiltonian into two parts is inspired by the success of Haldane's pseudo-potential method in FQHE [22] and the model of the spin-1 chain [9]. Returning to our spin model, now it is clear why we divided the Hamiltonian like this; notice that the coefficient decreases

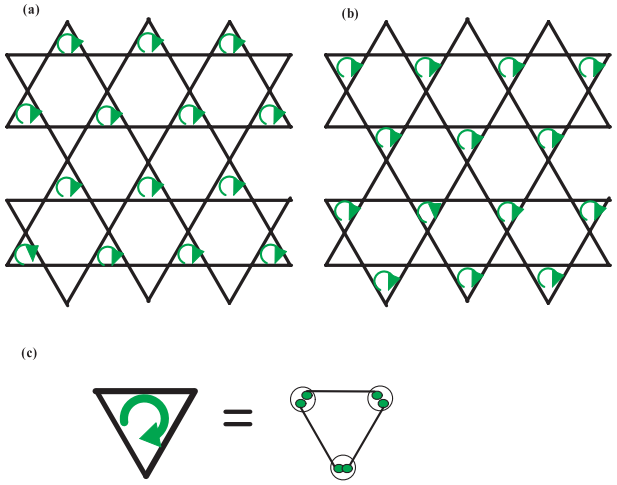

Figure 2. (a) and (b) The two-fold degenerate ground states: $|\Psi_a\rangle$ and $|\Psi_b\rangle$. (c) The structure of one trimer: an AKLT chain with length $L = 3$ and periodical boundary condition.

rapidly as S descends, thus if we can find the exact ground state of $\mathfrak{B}_0 = \sum_\alpha \mathfrak{B}^0_\alpha$, we can treat the left part $\mathfrak{B}_1 = \sum_\alpha \mathfrak{B}^1_\alpha$ and investigate the properties of the ground state.

First we focus on the exact ground state of the model

$$\mathfrak{B}_0 = \sum_\alpha [15\mathbf{P}^5_\alpha + 10\mathbf{P}^4_\alpha + 6\mathbf{P}^3_\alpha - 5]. \tag{6}$$

Since $\mathfrak{B}^0_\alpha$ is positive semidefinite, any state with the total spin of each block $S^{\mathrm{T}}_\alpha \leqslant 2$ is the exact ground state of equation (5). We can find that the only possibilities are the two-fold degenerate states: $|\Psi_a\rangle$ and $|\Psi_b\rangle$, as shown in figures 2(a) and (b), where each block possesses a trimer singlet. We can classify all the triangles in the kagome lattice into two classes: A and B. $|\Psi_a\rangle$ ($|\Psi_b\rangle$) is the direct product state of the trimer singlets within the A(B) triangles:

$$\begin{aligned} |\Psi_{a(b)}\rangle &= \prod_{i\in \mathrm{A(B)}} |S\rangle_i \\ |S\rangle_i &= \frac{1}{\sqrt{6}}(|1,-1,0\rangle - |1,0,-1\rangle + |0,1,-1\rangle \\ &\quad - |0,-1,1\rangle + |-1,0,1\rangle - |-1,1,0\rangle) \end{aligned} \tag{7}$$

the index i represents the dual lattice site and is located in the center of the triangles of the kagome lattice. $|S\rangle_i$ is a singlet composed of three spins on neighboring sites within the ith triangle. Because there exists a trimer singlet in each block, the total spin of each spin block could not be larger than 2. Therefore it is straightforward that we have found the exact ground states of $\mathfrak{B}_0$, or in other words, the trial ground state of our original Hamiltonian equation (3).

Now we will discuss the effect of the perturbation part. Since the trial ground states are two-fold degenerate, there seems to be a possibility that the perturbation would resonate these two trial ground states $|\Psi_a\rangle$ and $|\Psi_b\rangle$ to form a linear superposition and further lower the energy of the ground state. However, we would show this is not the case, at least in the thermodynamic limit. Let us calculate the nondiagonal term $\langle\Psi_a|H|\Psi_b\rangle$, it is not difficult to find that this nondiagonal term vanishes in the thermodynamic limit ($N \to \infty$), just like the 1D MG model. So the perturbation would not shift the two-fold degeneracy of the trial ground states and we can safely conclude that $|\Psi_a\rangle$ and $|\Psi_b\rangle$ provide a good description for the Hamiltonian equation (3). It is straightforward to get the ground energy of this variational ground state at the point $J = 3J_1 = 3J_2$

$$\langle\Psi_a|H|\Psi_a\rangle = -\tfrac{3}{2}JN, \tag{8}$$

where N is the number of triangles in the lattice.

Here we would briefly discuss the validity of the perturbation method we used above. The success of our perturbation method depends on the difference between the coefficients of the projection operator in the unperturbed Hamiltonian and that in the perturbation part. In our model, the rapidly decreasing coefficients of the projection operator as S descends make sure that the perturbation method works very well (actually, we have applied a similar method to the frustrated spin-1 Heisenberg model in the square lattice [10], and obtained a result which is consistent with the numerical result [23]). Usually, when we consider a certain variational wavefunction as a good approximation for the ground state of a Hamiltonian, there are two-fold meanings: (i) the variational energy is as low as possible; (ii) the variational wavefunction is close to that of the real ground state, which means the overlap between these two wavefunctions is close to 1. Indeed, it is possible to find other variational wavefunctions which provide the same or even lower variational energy for Hamiltonian (1). However, without the perturbation method, condition (ii) cannot be satisfied. Take our model for example, we can construct another state $|\Psi_c\rangle$ [24], an arbitrary covering of the kagome lattice with nearest neighbor dimers ($\frac{1}{\sqrt{3}}|1,-1\rangle - |0,0\rangle + |-1,1\rangle$). It is well known that such a covering necessarily contains $N/4$ triangles for which no bond is occupied (defaults) and $3N/4$ triangles with a dimer. It is easy to verify that the variational energy of Ψ_c is exactly the same as Ψ_a or Ψ_b. However, this does not mean that Ψ_c also provides a good approximation of the real ground state, because it does not satisfy condition (ii).

It is also interesting to study the properties of the excited states as well as the magnetization process of this model. It is well known that for the 2D dimer model, there are magnetization plateaus in the magnetization curve because of the localization of the single triplet excitation [7, 25, 26]. It is natural to ask whether this happens in our model. Notice that for a single trimer, the lowest excitation state is the triplet state, which is the direct product of a dimer singlet composed of two spins and a single spin-1. The triplet state is shown in figure 3. For convenience, we use $|m\rangle$ to denote the trimer singlet in the mth triangle being excited to a triplet:

$$\begin{aligned} |m\rangle &= \prod_{i\neq m} |S\rangle_i \bigotimes |T\rangle_m \\ |T\rangle_m &= \frac{1}{\sqrt{3}}(|1,-1\rangle + |-1,1\rangle - |00\rangle) \bigotimes |\alpha\rangle \end{aligned} \tag{9}$$

where $\alpha = 1, 0$, or -1 represent the spin state of the single spin-1. To study the low energy excitation of this model. We

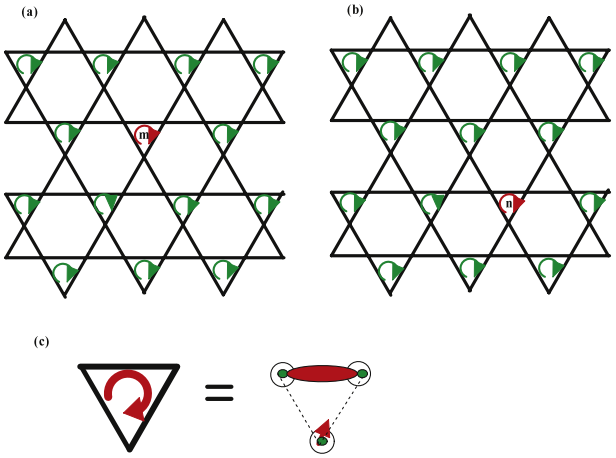

Figure 3. The triplet excited states: a triplet located at m (a) and n (b), m and n are nearest neighbor triplets. A triplet is formed by a dimer of two spin-1 and a single spin-1 (c).

find that

$$\langle m|H|n\rangle = 0 \tag{10}$$

for $m \neq n$ and

$$\langle m|H|m\rangle = -J - \tfrac{3}{2}J(N-1) \tag{11}$$

where H is the Hamiltonian (3), and $|m\rangle$ and $|n\rangle$ are shown in figures 3(a) and (b). The energy spectrum can be calculated by the single mode approximation (SMA) [9]. A momentum eigenstate is thus defined as:

$$|\mathbf{k}\rangle = \sum_{\mathbf{r}_m} \mathrm{e}^{\mathrm{i}\mathbf{r}_m\mathbf{k}}|m\rangle. \tag{12}$$

Using equations (10) and (11), we can get:

$$\langle \mathbf{k}|H|\mathbf{k}\rangle = N(-J - \tfrac{3}{2}J(N-1)). \tag{13}$$

The energy spectrum is:

$$\omega_{\mathbf{k}} = \frac{\langle \mathbf{k}|H|\mathbf{k}\rangle}{\langle \mathbf{k}|\mathbf{k}\rangle} - E_0 = \frac{J}{2} \tag{14}$$

where E_0 is the ground state energy defined in equation (8). Notice that under the single mode approximation, the energy spectrum $\omega_{\mathbf{k}}$ is independent of $\mathbf{k}$, which means that the single triplet excitation is almost dispersionless and thus localized, at least to the order we considered. Equation (14) also means that it is gapped, with a gap of $\frac{1}{2}J$. This dispersionless triplet excitation in our model is different from its one-dimensional analogue [27, 28], and it would localize or form a bound pair with other single triplets, just as in the dimer state [29, 30]. Because of the localization of the single triplet excitation, there are magnetization plateaus in the magnetization curve in our model. A simple calculation will show that at least one magnetization plateau appears at $m/m_{\mathrm{sat}} = 1/2$ corresponding to the phase (c) in figure 3. It is known that a similar magnetization process and magnetization plateaus have been studied in other spin-1 systems, such as the $S = 1$ spin chain [28] or the $S = 1$ Heisenberg model

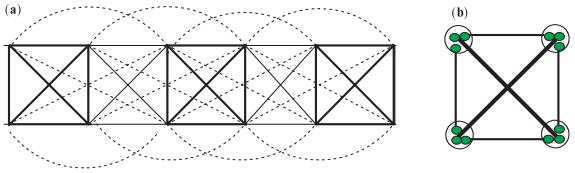

Figure 4. (a) The cluster state ($N = 4$) in a spin-3/2 ladder. (b) The structure of an $SU(4)$ cluster singlet. A 3/2 spin can be considered as a symmetrized state of three 1/2 spins. The bold lines represent a singlet for two 1/2 spins.

(only including the nearest coupling) in uniform and distorted kagome lattices [31].

However, we have not considered the effect of the interactions between the triplets, which may lead to new plateaus. Furthermore, it is possible that the higher order interaction would result in a pair of neighboring triplets, similar to the dimer phase [29, 30].

Now we will discuss the stability of our trimer state. Up to now, all of our analyses are based on a special point of our coupling parameters. At this point, the trimer state actually provides a good approximation of the exact ground state, a slight perturbation should not change the nature of this ground state because it is a gapped VBS state which cannot be changed by a small perturbation unless it can overcome the energy gap. Just as in the one-dimensional AKLT state, introducing a small next nearest coupling cannot change the nature of the 1D VBS state [32].

Usually there are two kinds of perturbation, the first kind is deviation from the special point of the coupling parameters $J = 3J_1 = 3J_2$, as analyzed above, where slight deviation will not change the nature of the ground state. However, there is another important point: $J_1 = J_2 = 0$, which corresponds to the $S = 1$ antiferromagnetic Heisenberg model with only nearest coupling. Hida first studied the ground state at this point by means of exact diagonalization and the cluster expansion [16]. It is shown that the ground state in this case is the hexagonal singlet solid (HSS) state, rather than the trimer state in our case. The second possible perturbation for this kind of spin-1 system is the spin biquadratic term, which is necessary to construct the 1D AKLT model [8]. However, as shown in [9], the exact ground state of the AKLT Hamiltonian ($3P^2(i, i+1)$ in equation (2)) which includes the biquadratic term, actually provides a good variational ground state for the pure $S = 1$ Heisenberg Hamiltonian without the biquadratic term. Therefore in our case, the perturbation of including the small spin biquadratic term would not change the nature of our trimer state.

The trimer state is not the only example of our cluster state, another example is the spin-3/2 $SU(4)$ spin ladder [33]. At a particular value of the coupling, the ground state of this model is the exact spontaneous plaquette ground state, in which four 3/2 spins form an $SU(4)$ spin singlet plaquette and spontaneously break the translational symmetry, as shown in figure 4. This model is a generalization of the MG model to the spin ladder systems and its ground state belongs to our cluster state with $N = 4$.

In conclusion, we have discussed the spontaneous trimerization in a frustrated spin-1 antiferromagnet in a kagome lattice by the pseudo-potential approach. Other interesting properties such as the magnetization plateaus appearing in the magnetization curve in our model are also discussed. We believe that this exotic trimer state could be verified numerically by the 2D DMRG method.

Acknowledgments

After completing this work, we learnt that in two recent papers [34, 35], similar topics about the exotic 2D VBS state have been discussed. ZC thanks Flavio S Nogueira for helpful discussions. This work is supported by the NSFC, the Knowledge Innovation Project of CAS, the National Program for Basic Research of MOST and programs of the Chinese Academy of Sciences and Deutscher Akademischer Austausch Dienst (DAAD).

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