Honeycomb antiferromagnet with a triply degenerate dimer ground state

Rakesh Kumar,* Dushyant Kumar, and Brijesh Kumar†
School of Physical Sciences, Jawaharlal Nehru University, New Delhi 110067, India
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We present an antiferromagnetic quantum spin-1/2 model on honeycomb lattice. It has two parts, one of which is the usual nearest-neighbor Heisenberg model. The other part is a certain multiple spin interaction term, introduced by us, which is exactly solvable for the ground state. Without the Heisenberg part, the model has an exact threefold degenerate dimer ground state. This exact ground state is also noted to exist for the general spin-*S* case. For the spin-1/2 case, we further carry out the triplon analysis in the ground state to study the competition between the Heisenberg and the multiple spin interactions. This approximate calculation exhibits a continuous quantum phase transition from the dimer order to Néel order.

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I. INTRODUCTION

The low-dimensional quantum spin systems are a subject of great current interest. Much of this research directly attends to the real (quasi-)one-dimensional (1D) and two-dimensional (2D) spin systems studied in the laboratories.[1–4] There is also a formal side to it which is concerned with investigating, at various theoretical levels, the effects of low spatial dimensionality, quantum spin fluctuations, and frustration on the nature of the ground state of a model spin system. Since the antiferromagnetism is sensitive to all of these, the ground state of a quantum antiferromagnet (AF) can choose from a variety of possibilities (known or not yet known).[5–7] For example, the ground state of the spin-1/2 nearest-neighbor (nn) Heisenberg antiferromagnet is a critical spin liquid (with power-law decay of the spin-spin correlations and zero local magnetic moment) in 1D but it has Néel order on two and higher dimensional bipartite lattices. Furthermore, the competing interactions can induce changes in the nature of ground state, say, from being a Néel-ordered state to becoming spontaneously dimerized[8] (or something else). The Majumdar-Ghosh model presents an exactly solvable case of a spontaneously dimerized doubly degenerate singlet ground state in 1D.[9] Similar spin models have also been constructed in 2D.[8,10–13]

Of the spin systems in 2D, the honeycomb lattice comes across as a special case to study. Its nearest-neighbor coordination is three which lies between 1D and the square lattice. Hence, the quantum fluctuations are expected to be stronger on the honeycomb than on the square lattice. Moreover, the honeycomb lattice has two spins per unit cell. Thus, we expect a natural case for spontaneous dimer order, without breaking translational symmetry, in a honeycomb antiferromagnet. For the spin-1/2 nn Heisenberg AF on honeycomb lattice, various calculations[14–16] indeed show larger quantum fluctuations than on the square lattice but the ground state still exhibits Néel order (although weaker than square lattice). Several other studies have shown that under various frustrated conditions the ground state on honeycomb lattice can get disordered.[17–21] Motivated by these observations, one of us (B.K.) constructed a quantum spin model with multiple spin interactions on honeycomb lattice, which has an exact triply degenerate dimer ground state (see Fig. 2). Here, we present this model with an additional nn Heisenberg AF interaction and investigate using triplon mean-field theory (MFT) the transition from the dimer to Néel order in the ground state. We believe this to be an interesting model for two simple reasons. First, a number of antiferromagnetic materials with honeycomb structure have been studied recently.[3,4,22] While most of these exhibit AF order at low temperatures, a few do show spin-gap behavior.[23] It is possible that the dimerized singlet ground state arising in this model may well be realized in a real material. Second, the investigation of deconfined quantum criticality in 2D quantum antiferromagnets has been a subject of active research in recent times.[6,24,25] This model presents an interesting case to study in this context.

This paper is organized as follows. In Sec. II, we discuss the model and its exact ground state. In Sec. III, we do the triplon mean-field theory. In Sec. IV, we discuss the results of the triplon analysis, in particular, the spin gap and staggered magnetization, and the quantum phase diagram. Finally, we conclude with a summary.

II. MODEL

In this paper, we study the following quantum spin-1/2 model on honeycomb lattice (pictorially shown in Fig. 1).

$$H = J\sum_{\langle ij\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + \frac{K}{8}\sum_{\hexagon} [\mathbf{S}_{12}^2\mathbf{S}_{34}^2\mathbf{S}_{56}^2 + \mathbf{S}_{23}^2\mathbf{S}_{45}^2\mathbf{S}_{61}^2]. \quad (1)$$

Here, $\mathbf{S}_{ij}^2=(\mathbf{S}_i+\mathbf{S}_j)^2$. The first term above is the Heisenberg model with nearest-neighbor interaction J. The second term is what we have introduced to realize the dimer ground state (as in Fig. 2). This multiple spin interaction is generated by the product of the pairwise total spins of three nn pairs on each hexagon [in two different ways, (1,2)(3,4)(5,6) or (2,3)(4,5)(6,1)].[26] It involves all the six spins of an hexagon and the summation is taken over all hexagonal plaquettes of the honeycomb lattice with periodic boundary condition. After expanding $\mathbf{S}_{ij}^2$ as $\frac{3}{2}+2\mathbf{S}_i\cdot\mathbf{S}_j$ (for a pair of spin 1/2) and regrouping different terms, Eq. (1) becomes $H=\frac{27}{64}KL+H^{(2)}+H^{(4)}+H^{(6)}$, where L is the total number of lattice sites and $H^{(2)}$, $H^{(4)}$, and $H^{(6)}$ denote the quadratic, quartic, and sextic spin interactions, respectively. Note that H

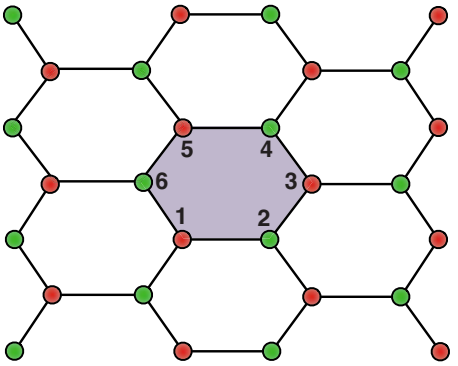

FIG. 1. (Color online) Pictorial representation of the model [Eq. 1]. The connecting lines denote the nearest-neighbor antiferromagnetic interaction, J. The shaded hexagon denotes the multiple spin interaction, K, present on every hexagonal plaquette.

respects the lattice translation and point-group symmetries and is also SU(2) invariant. We take J, $K>0$, and $J+K=1$ sets the unit of energy. Thus, $J=1-K$, where $K\in[0,1]$.

A. Exact ground state

Consider the model for $K=1$. It can be written as

$$H_K=\frac{1}{8}\sum_{\hexagon}[h_{K1}+h_{K2}], \tag{2}$$

where $h_{K1}=\mathbf{S}_{12}^2\mathbf{S}_{34}^2\mathbf{S}_{56}^2$ and $h_{K2}=\mathbf{S}_{23}^2\mathbf{S}_{45}^2\mathbf{S}_{61}^2$. In this case, the nn Heisenberg exchange is absent. We only have the multiple spin interactions. Clearly, h_{K1} and h_{K2} have positive eigenvalues, with zero as the minimum. Hence, the ground-state energy of H_K is bounded below by zero. The operator h_{K1} gives zero when at least one of the three concerned spin pairs, that is, (1,2), (3,4), or (5,6), forms a singlet. Similarly for h_{K2}. Therefore, on a single hexagon, a zero energy eigenstate of $h_{K1}+h_{K2}$ can be obtained by simultaneously forming singlets on the opposite edges. It leaves the remaining two spins as "free." For example, one such state is $[1,2]\otimes|m_3\rangle\otimes[4,5]\otimes|m_6\rangle$, where $[i,j]=(|\uparrow_i\downarrow_j\rangle-|\downarrow_i\uparrow_j\rangle)/\sqrt{2}$ is the singlet formed by i and j spins, and $m_k=\uparrow$ or $\downarrow$. Moreover, there are three ways of choosing such dimer forming spin pairs. After knowing these dimer states for a single hexagon, it is straightforward to show that the three dimer-ordered configurations shown in Fig. 2 form the exact zero energy ground state of H_K on the full lattice. We have also cross checked this analytic assertion by numerically diagonalizing H on 12-site and 18-site spin clusters with periodic boundary conditions. The results for the 18-site cluster are presented in the following section. These dimer configurations have also been known to arise in the ground state of the quantum dimer model[27] and a frustrated SU(2) spin-1/2 model on honeycomb lattice.[19] Ours is an example of a quantum spin model on honeycomb lattice where this triply degenerate dimer ground state is realized *exactly*. Moreover, it is valid for arbitrary spins (discussed below), although we have presented only spin-1/2 case in this paper.

Let the ground-state dimer configurations be denoted as $|\phi_1\rangle$, $|\phi_2\rangle$, and $|\phi_3\rangle$. These dimer states do not break the translational symmetry of the lattice and are obviously SU(2) invariant. The point-group rotational symmetry is broken, however. The wave function of the dimer state, $|\phi_1\rangle$, can be explicitly written as

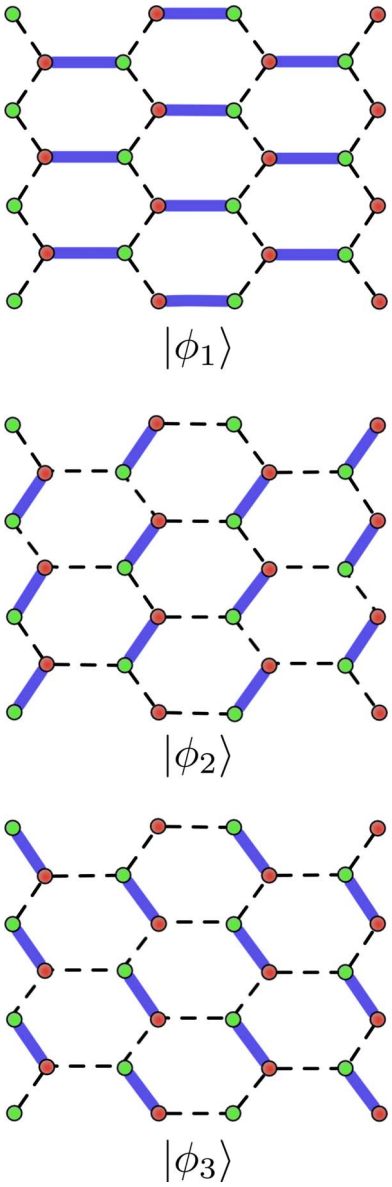

FIG. 2. (Color online) The dimer states $|\phi_1\rangle$, $|\phi_2\rangle$, and $|\phi_3\rangle$ form the ground state of the Hamiltonian, H_K. The thick (blue) lines denote the dimer singlets.

$$|\phi_1\rangle=\otimes\prod_{(i,j)\in\mathcal{D}}[i,j], \tag{3}$$

where $\mathcal{D}$ is the set of singlet forming bonds in the state $|\phi_1\rangle$. The other two states, $|\phi_2\rangle$ and $|\phi_3\rangle$, are related to $|\phi_1\rangle$ via the threefold rotation as

$$|\phi_2\rangle=\mathcal{C}_3|\phi_1\rangle, \tag{4a}$$

$$|\phi_3\rangle=\mathcal{C}_3^2|\phi_1\rangle, \tag{4b}$$

where $\mathcal{C}_3$ is the clockwise $2\pi/3$ rotation operator.

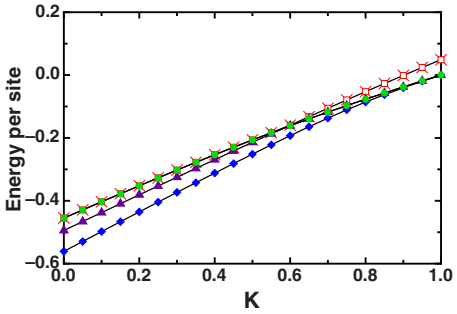

FIG. 3. (Color online) Eigenvalues from the exact diagonalization of H on the 18-site honeycomb cluster (total $S_z=0$).

To this end, we would like to mention that this exact ground state of H_K is also valid for a general spin-S system. For the spin-S case, a dimer would denote a singlet state formed by a pair of spin S. Everything else (that is, the dimer pattern, the degeneracy, and the ground-state energy) is the same. Since the maximum total spin of a pair of spin S is $2S$, we can rescale the coupling $\frac{K}{8}$ to $\frac{(K/2)}{[2S(2S+1)]^2}$. This just makes the energy contribution of the multiple spin interaction comparable (in powers of S) to that of the Heisenberg part.

B. Finite size numerical diagonalization

We have done exact numerical diagonalization of H on an 18-site honeycomb cluster with periodic boundary conditions (see Fig. 4). This is just to numerically verify the exact ground state on a small cluster. We only use total magnetization and spin-inversion symmetries in the coding. The exact diagonalization (ED) results clearly show that the ground state for the exactly solvable case ($K=1$) is indeed triply degenerate with zero energy (see Fig. 3). Away from the exact case, the ground-state energy decreases smoothly without level crossing. Moreover, the degeneracy seems to survive, for K close to 1, even on such a small lattice. We therefore expect the ground state to remain triply degenerate over a finite range of K values for large enough systems.

In order to ascertain the nature of $K=1$ ground state, we compute the spin-spin correlation, $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$, and the dimer-dimer correlation,

$$D_{(i,j,k,l)} = \langle (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l) \rangle - \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle \langle \mathbf{S}_k \cdot \mathbf{S}_l \rangle. \quad (5)$$

The latter helps in identifying the dimer order. The $D_{(i,j,k,l)}$ is positive when the two dimers are singlet correlated and negative otherwise. We compute these correlations first in the numerically generated ground state and then compare them with those calculated in the exactly known dimer ground state. On the infinite lattice, the nn spin-spin correlation is equal to $-1/4$ (further neighbor spin correlations are identically zero in the states of Fig. 2) and the dimer-dimer correlation is 1/8 when two dimers are pure singlets.

For $K=1$, the numerical ground-state wave functions would be some orthogonal linear combinations of $|\phi_1\rangle$, $|\phi_2\rangle$, and $|\phi_3\rangle$. The choices of the linear combination are not unique, however. As we have not implemented translation and point-group symmetries in our computational scheme, this ambiguity in the degenerate output states of our (less sophisticated) program remains. Therefore, we use the zero-temperature thermal density operator to correctly compute the ground-state properties. For an operator $\hat{\mathcal{O}}$, its thermal average is given by $\langle \hat{\mathcal{O}} \rangle = \mathrm{Tr}(\hat{\rho}\hat{\mathcal{O}})$, where $\hat{\rho} = Z^{-1} e^{-\beta \mathcal{H}}$ is the thermal density operator ($Z = \mathrm{Tr}\, e^{-\beta \mathcal{H}}$). In the zero-temperature limit, the density operator reduces to

$$\hat{\rho} = \frac{1}{N_g} \sum_{\nu=1}^{N_g} |\Psi_\nu\rangle\langle\Psi_\nu|, \quad (6)$$

where N_g is the degeneracy of the ground state and $|\Psi_\nu\rangle$ are the orthonormalized ground-state eigenvectors. In the present calculation, $N_g=3$.

Since the numerical eigenstates are orthonormal, we use them directly to compute the correlations in the ground state, as prescribed above. These data are shown in the second column of Tables I and II. The exact wave functions, $|\phi_1\rangle$, etc., are not orthogonal. Therefore, we first orthogonalize them using Gram-Schmidt procedure (on the same cluster as used for numerical diagonalization; see Fig. 4), then apply the density operator averaging to compute the correlations. These are given in the third column of the two tables. Clearly, the spin correlations are nearest-neighbor type and the dimer correlation matches nicely with dimer order in the exact ground state (compare Fig. 2 with Fig. 4). The numbers from the two calculations are in excellent agreement.

TABLE I. Spin-spin correlation, $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$, on 18-site cluster.

(i,j)	Numerical diagonalization	Exact
(10, 1)	0.00012025	0.00012025
(10, 2)	0.00000000	0.00000000
(10, 3)	0.00012025	0.00012025
(10, 4)	−0.00012025	−0.00012025
(10, 5)	0.00000000	0.00000000
(10, 6)	−0.00012025	−0.00012025
(10, 7)	0.00012025	0.00012025
(10, 8)	−0.00012025	−0.00012025
(10, 9)	−0.24987975	−0.24987975
(10, 11)	−0.24987975	−0.24987975
(10, 12)	−0.00012025	−0.00012025
(10, 13)	0.00000000	0.00000000
(10, 14)	−0.00012025	−0.00012025
(10, 15)	−0.24987975	−0.24987975
(10, 16)	−0.00012025	−0.00012025
(10, 17)	0.00000000	0.00000000
(10, 18)	0.00000000	0.00000000

III. TRIPLON MEAN-FIELD THEORY

While at $K=1$, the ground state of H has an exact dimer order but it is known to be a Néel-ordered AF state when $K=0$. It would be interesting, therefore, to make some investigation of the transition from the dimer to Néel-ordered ground state, as K is varied. Here, we present an approximate

TABLE II. Dimer-dimer correlation on 18-site cluster.

Dimer-dimer	Numerical diagonalization	Exact
(10, 15) (2, 7)	0.12496993	0.12496992
(10, 15) (4, 9)	0.12496993	0.12496992
(10, 15) (6, 11)	0.12496993	0.12496992
(10, 15) (8, 13)	0.12493986	0.12495990
(10, 15) (12, 17)	0.12493986	0.12495990
(10, 15) (14, 1)	0.12496993	0.12496992
(10, 15) (16, 3)	0.12496993	0.12496992
(10, 15) (18, 5)	0.12496993	0.12496992
(10, 15) (2, 3)	−0.06253008	−0.06251004
(10, 15) (4, 5)	−0.06253008	−0.06251004
(10, 15) (6, 1)	−0.06253008	−0.06251004
(10, 15) (8, 9)	−0.06253008	−0.06251004
(10, 15) (12, 7)	−0.06253008	−0.06251004
(10, 15) (16, 17)	−0.06253008	−0.06251004
(10, 15) (18, 13)	−0.06253008	−0.06251004
(10, 15) (2, 1)	−0.06253008	−0.06251004
(10, 15) (4, 3)	−0.06253008	−0.06251004
(10, 15) (6, 5)	−0.06253008	−0.06251004
(10, 15) (8, 7)	−0.06253008	−0.06251004
(10, 15) (12, 11)	−0.06253008	−0.06251004
(10, 15) (14, 13)	−0.06253008	−0.06251004
(10, 15) (18, 17)	−0.06253008	−0.06251004

study of this quantum phase transition by doing triplon analysis with respect to the dimerized singlet ground state. A *triplon is a triplet excitation residing on a dimer and dispersing due to the interactions present in the system. While a nonzero gap in the triplon dispersion implies a stable dimer phase, the gaplessness corresponds to having AF order in the ground state*.[8,28]

The triplon analysis is conveniently carried out in the bond-operator representation in which the singlet and three triplet states of a pair of spin 1/2 (a bond) are described as

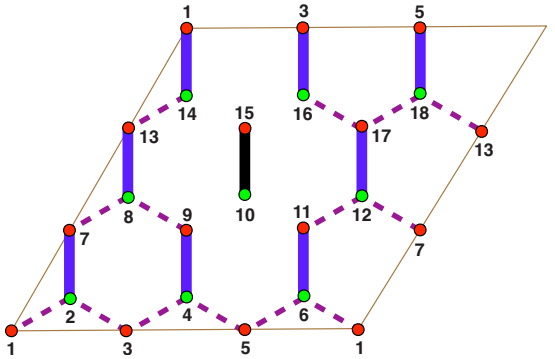

FIG. 4. (Color online) The dimer-dimer correlations are calculated by taking the dimer (10,15) as reference. Thickness of the dimers is proportional to their dimer correlation values. The thick blue dimer denotes the positive correlation (i.e., a singlet), whereas the purple (dashed) ones represent negative correlation (i.e., no singlet).

bosons (called bond operators).[28] The bosonic creation operators, $s^\dagger$ and $t_\alpha^\dagger$ ($\alpha=x,y,z$), respectively, create singlet and triplet states on a bond, subjected to the physical constraint, $s^\dagger s+t_\alpha^\dagger t_\alpha=1$ (repeated Greek indices summed over). The two spin operators on a dimer are represented as

$$S_{1\alpha}=\frac{1}{2}(s^\dagger t_\alpha+t_\alpha^\dagger s-i\epsilon_{\alpha\beta\gamma}t_\beta^\dagger t_\gamma), \tag{7a}$$

$$S_{2\alpha}=\frac{1}{2}(-s^\dagger t_\alpha-t_\alpha^\dagger s-i\epsilon_{\alpha\beta\gamma}t_\beta^\dagger t_\gamma), \tag{7b}$$

where subscripts 1 and 2 denote the two spins of the dimer and $\epsilon_{\alpha\beta\gamma}$ is the totally antisymmetric tensor. In the simplest triplon analysis, the singlet background is treated as a mean field ($s^\dagger\equiv\bar{s}$) and the triplon dispersion is calculated by keeping only bilinear terms in the corresponding triplon Hamiltonian (that is, by ignoring the interaction between triplons; please consult Ref. 8 for an in-detail description of the triplon analysis; we follow the same strategy as therein).

Although the exact dimer ground state is triply degenerate, we can only take one of these as the reference state to carry out the triplon analysis. This is of course a limitation when there are degenerate dimer configurations to choose from (such as in the present case) and one would wish not to pick one over others. However, we adopt this limited approach to get some idea of the effect of the nn Heisenberg interaction on the exact dimer ground state. We rewrite H in terms of the bond operators, taking $|\phi_1\rangle$ as the reference background, where the bond-operator representation is used for the spins on the singlet forming dimers. The Heisenberg exchange interaction on such a dimer can now be written as

$$\mathbf{S}_1(\mathbf{R})\cdot\mathbf{S}_2(\mathbf{R})=-\frac{3}{4}\bar{s}^2+\frac{1}{4}t_\alpha^\dagger t_\alpha, \tag{8}$$

where $\mathbf{R}$ is the position vector of the dimer. For the spins coming from different dimers, we have

$$\mathbf{S}_l(\mathbf{R})\cdot\mathbf{S}_m(\mathbf{R}+\underline{\delta})\approx(-1)^{l+m}\frac{\bar{s}^2}{4}[t_\alpha^\dagger(\mathbf{R})t_\alpha(\mathbf{R}+\underline{\delta})+t_\alpha(\mathbf{R})t_\alpha(\mathbf{R}+\underline{\delta})+\text{H.c.}]\quad(l,m=1,2). \tag{9}$$

By using Eqs. (8) and (9), and applying the constraint on bond operators globally (with μ as Lagrange multiplier), we get the following mean-field Hamiltonian for H,

$$H_{mf}=E_0+\frac{1}{2}\sum_{\mathbf{k}}\{(\lambda-\bar{s}^2\xi_{\mathbf{k}})[t_{\mathbf{k}\alpha}^\dagger t_{\mathbf{k}\alpha}+t_{-\mathbf{k}\alpha}t_{-\mathbf{k}\alpha}^\dagger]-\bar{s}^2\xi_{\mathbf{k}}[t_{\mathbf{k}\alpha}^\dagger t_{-\mathbf{k}\alpha}^\dagger+t_{-\mathbf{k}\alpha}t_{\mathbf{k}\alpha}]\}. \tag{10}$$

Here, the triplon operators have been Fourier transformed from the dimer lattice to the corresponding reciprocal lattice, with $\mathbf{k}$ vectors lying in its first Brillouin zone. Moreover,

$$E_0=\frac{L}{2}\left[\frac{J}{4}+\frac{9}{8}K-\left(J+\frac{9}{8}K\right)\bar{s}^2-\frac{5}{2}\lambda+\lambda\bar{s}^2\right], \tag{11}$$

$$\lambda = \frac{1}{4}\left(J + \frac{9}{8}K\right) - \mu, \tag{12}$$

and

$$\xi_{\mathbf{k}} = \left[J + \frac{9}{8}K(1-\bar{s}^2)\right]\cos\left(\frac{3}{2}k_x\right)\cos\left(\frac{\sqrt{3}}{2}k_y\right), \tag{13}$$

where λ is the effective chemical potential. Equation (10) is diagonalized by the Bogoliubov transformation. The diagonal mean-field Hamiltonian can be written as

$$H_{mf} = E_0 + \sum_{\mathbf{k}} E_{\mathbf{k}}\left(\gamma^{\dagger}_{\mathbf{k}\alpha}\gamma_{\mathbf{k}\alpha} + \frac{3}{2}\right), \tag{14}$$

where $\gamma_{\mathbf{k}\alpha}$'s are Bogoliubov bosons and the triplon dispersion, $E_k = \sqrt{\lambda(\lambda - 2\bar{s}^2\xi_k)} \geq 0$. The ground-state energy per site is given by

$$e_g[\lambda, \bar{s}^2] = e_0 + \frac{3}{2L}\sum_{\mathbf{k}} E_{\mathbf{k}}, \tag{15}$$

where $e_0 = E_0/L$. By minimizing the ground-state energy with respect to λ and $\bar{s}^2$, we get the self-consistent equations, whose solution gives the mean-field results.

A. Dimer phase

When the minimum of the triplon dispersion is nonzero, the dimer phase is stable against triplet excitations. Therefore, the gapped triplon phase corresponds to having dimer ground state. The self-consistent equations in this case are

$$\bar{s}^2 = \frac{5}{2} - \frac{3}{L}\sum_{\mathbf{k}} \frac{\lambda - \bar{s}^2\xi_{\mathbf{k}}}{E_{\mathbf{k}}}, \tag{16a}$$

$$\lambda = J + \frac{9}{8}K + \frac{3\lambda}{L}\sum_{\mathbf{k}} \frac{\eta_{\mathbf{k}}}{E_{\mathbf{k}}}, \tag{16b}$$

where $\eta_{\mathbf{k}} = \xi_{\mathbf{k}} - \frac{9}{8}K\bar{s}^2\cos(\frac{3}{2}k_x)\cos(\frac{\sqrt{3}}{2}k_y)$. These equations are obtained by minimizing the ground-state energy, i.e., $\partial e_g/\partial\lambda = 0$ and $\partial e_g/\partial\bar{s}^2 = 0$.

The weight of having singlet state on a dimer is measured by $\bar{s}^2$. If all the dimers form perfect singlets (like in the exact case), then $\bar{s}^2 = 1$. Otherwise, we get $\bar{s}^2 < 1$, due to triplon fluctuations in the ground state.

B. Néel phase

As K is gradually decreased away from $K=1$, at some point we find that the triplon gap vanishes (see Fig. 5). The triplon dispersion now touches zero at $\mathbf{k}=\mathbf{Q}$, where $\mathbf{Q}=(0,0)$. It means the triplon occupancy at wave vector $\mathbf{Q}$ becomes singular, which implies the Bose condensation of triplons at $\mathbf{Q}$. Thus, we need to introduce a third quantity (in addition to λ and $\bar{s}^2$), the triplon condensate density, n_c, which is notionally given by

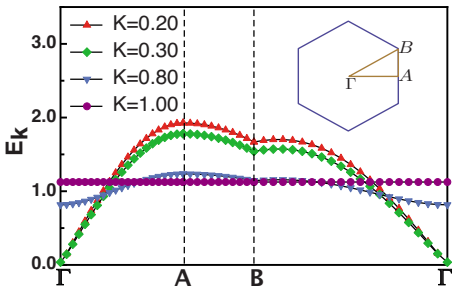

FIG. 5. (Color online) Triplon dispersion E_k for different values of K. The Γ, A, and B denote the wave vectors $(0,0)$, $(2\pi/3,0)$, and $(2\pi/3,2\pi/3\sqrt{3})$ in the Brillouin zone.

$$n_c = \frac{2}{L}\langle t^{\dagger}_{\mathbf{Q}\alpha} t_{\mathbf{Q}\alpha}\rangle \equiv \frac{3}{L}\left(\frac{\lambda - \bar{s}^2\xi_{\mathbf{Q}}}{E_{\mathbf{Q}}}\right). \tag{17}$$

Now, the revised set of self-consistent equations is

$$\lambda = 2\bar{s}^2\xi_{\mathbf{Q}}, \tag{18a}$$

$$n_c = \frac{1}{2\eta_{\mathbf{Q}}}\left[\lambda - \frac{3\lambda}{L}\sum_{\mathbf{k}\neq\mathbf{Q}} \frac{\eta_{\mathbf{k}}}{E_{\mathbf{k}}} - \left(J + \frac{9}{8}K\right)\right], \tag{18b}$$

$$\bar{s}^2 = \frac{5}{2} - n_c - \frac{3}{L}\sum_{\mathbf{k}\neq\mathbf{Q}} \frac{\lambda - \bar{s}^2\xi_{\mathbf{k}}}{E_{\mathbf{k}}}. \tag{18c}$$

Physically, the nonzero n_c corresponds to having an AF order in the ground state, which in the present case comes out to be the Néel order. The staggered magnetic moment in the Néel phase is given by $M_s = \bar{s}\sqrt{n_c}$.

IV. RESULTS AND DISCUSSION

Now, we present the results obtained by the triplon mean-field calculation. The self-consistent Eqs. (16a) and (16b) of the gapped phase are solved for λ and $\bar{s}^2$, for different values of K. Interestingly, for $K=1$, it gives $\bar{s}^2 = 1$, same as the exact answer (see Fig. 6). Moreover, the mean-field and exact ground-state energies are both zero, as shown in Fig. 9. At the exact point, the triplon dispersion, $E_{\mathbf{k}}$ (plotted in Fig. 5), is flat with an energy gap of 1.125.

This value of gap at $K=1$ is in agreement with an estimate of 9/8, which is calculated as: $\Delta_{(K=1)} = \langle\tau|H_K|\tau\rangle/\langle\tau|\tau\rangle$, where

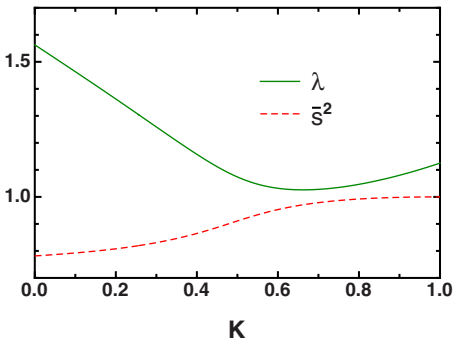

FIG. 6. (Color online) The singlet weight, $\bar{s}^2$, and the effective chemical potential λ.

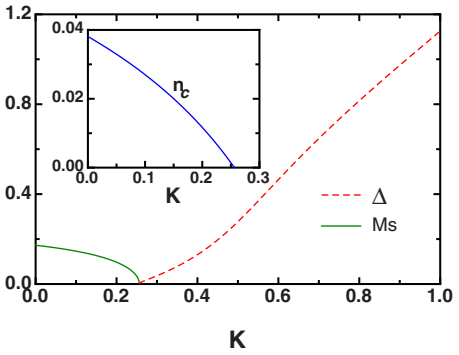

FIG. 7. (Color online) The spin gap, $\Delta = E_{\mathbf{Q}}$, and the staggered moment M_s. Inset: the triplon condensate density, n_c.

$|\tau\rangle$ is a "trial" localized triplon state as defined below

$$|\tau\rangle = \left(\otimes \prod_{(i,j)\in\mathcal{D}'} [i,j]\right) \otimes \{k,l\}. \tag{19}$$

Here, $\mathcal{D}'$ is the set of all dimers in $\mathcal{D}$ except (k,l) and $\{k,l\}$ denotes a triplet state on the bond (k,l). The set $\mathcal{D}$ is the set of all singlet forming dimers in $|\phi_1\rangle$ [see Eq. (3)].

For $K<1$, $E_{\mathbf{k}}$ acquires a finite width, with minimum at the Γ point [that is, $\mathbf{Q}$]. Eventually, it touches zero at $\mathbf{Q}$ and remains so below $K^*=0.256$. For $K<K^*$, we solve Eq. (18) self-consistently. As shown in Fig. 7, now the staggered magnetization, M_s, acquires a nonzero value while the gap remains zero. Our value of M_s at $K=0$ (pure Heisenberg model) is around 0.173, as compared to its value of 0.242 from spin-wave analysis,[14] 0.266 from series expansion,[16] and 0.22 from Monte Carlo simulations.[15] Our calculation gives a smaller value of the staggered magnetization because it is built around the singlet phase which obviously has more quantum fluctuations in it.

The triplon mean-field theory thus predicts a continuous transition from the dimer to Néel-ordered phase in the ground state of H for spin 1/2. The phase diagram is just a line presented in Fig. 8. As pointed out earlier, the $K=1$ model has the same exact ground state for higher spins also. Therefore, in some future studies, it would be interesting to extend this quantum phase diagram to include a spin axis, with $S=1/2,1,3/2,\ldots$ to $S\to\infty$ (the classical limit). The classical case can be discussed right away. Let the spins be classical vectors. The H_K will now have an infinitude of spin configurations in the ground state (not related via global spin rotation) because the spins on each dimer (of Fig. 2), separately, must cancel. Hence, H_K itself is a frustrated model. But the classical nn Heisenberg interaction does not need to compete against H_K for winning the ground state as the infinite set of pairwise spin-canceled configurations also includes the Néel states. Therefore, in the classical limit, K^*

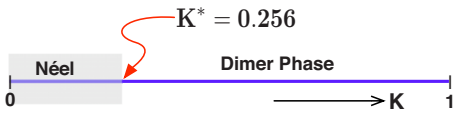

FIG. 8. (Color online) The mean-field quantum phase diagram of H for spin 1/2.

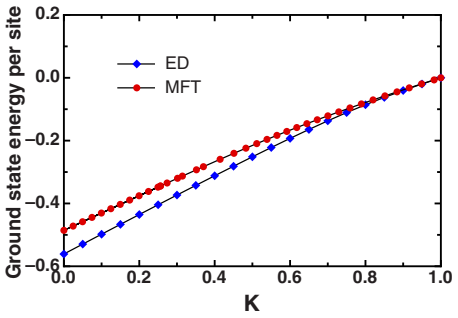

FIG. 9. (Color online) The ground-state energy from the MFT and the ED (18-site cluster). The two compare well close to $K=1$.

$=1$. This discussion reveals an important feature of H, which is that the multiple spin and Heisenberg interactions do not compete against (or frustrate) each other. Instead, the quantum mechanics acts better when K is sufficiently large. Hence, the Néel to dimer order transition in the ground state of H is driven purely by the quantum fluctuations.

Below, we present the critical behavior of the spin gap and staggered magnetization within this mean-field triplon analysis. The critical exponents for both the quantities are derived by analyzing the equations for the gap and n_c in the small neighborhood of K^*,

$$\Delta \approx 0.12(K-K^*)^{1/2}, \tag{20a}$$

$$M_s \approx 0.44(K^*-K)^{1/2}. \tag{20b}$$

Figure 10 shows an enlarged plot of the mean-field data around K^*, together with the estimated critical behavior. In the standard notation, $M_s \sim |K-K^*|^{\beta}$ and $\Delta \sim |K-K^*|^{z\nu}$, where z is the dynamical exponent and ν is the correlation length exponent. The critical exponent, $\beta=1/2$, from our calculation is same as the exponent from the quantum $O(N)$ nonlinear sigma model theory of a 2D quantum antiferromagnet in the limit $N\to\infty$.[29] It is more than the numerically computed value of 0.34 for a planar antiferromagnet.[30] Taking $z=1$, as it is normally, we get $\nu=1/2$, as compared to the value of 1 from the large N sigma model theory[29] and the numerically computed values of 0.69 for a quantum antiferromagnet on CaV_4O_9 structure[30] and 0.63 for the staggered dimer model on square lattice.[31]

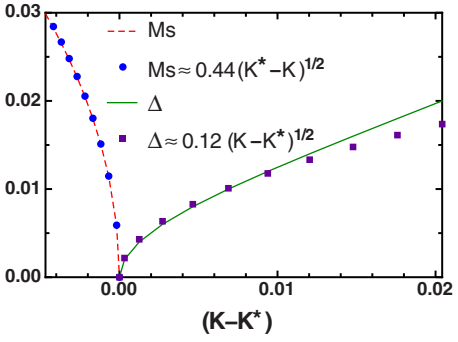

FIG. 10. (Color online) The spin gap and staggered magnetization near the critical point. The estimated results of Eq. (20) are also plotted.

TABLE III. Comparison of the dimer-dimer correlation in the ground state of the $K=1$ model without six-spin interaction with that in the exact dimer state (on the 18-site cluster).

Dimer-dimer	$(H^{(6)}=0)$ [a]	Exact dimer state
(10, 15) (2, 7)	0.12418097	0.12496992
(10, 15) (4, 9)	0.11922044	0.12496992
(10, 15) (6, 11)	0.11922044	0.12496992
(10, 15) (8, 13)	0.11916810	0.12495990
(10, 15) (12, 17)	0.11916810	0.12495990
(10, 15) (14, 1)	0.11922044	0.12496992
(10, 15) (16, 3)	0.11922044	0.12496992
(10, 15) (18, 5)	0.12418097	0.12496992
(10, 15) (2, 3)	−0.05880858	−0.06251004
(10, 15) (4, 5)	−0.05880858	−0.06251004
(10, 15) (6, 1)	−0.05923653	−0.06251004
(10, 15) (8, 9)	−0.06645716	−0.06251004
(10, 15) (12, 7)	−0.05880858	−0.06251004
(10, 15) (16, 17)	−0.06645716	−0.06251004
(10, 15) (18, 13)	−0.05880858	−0.06251004
(10, 15) (2, 1)	−0.05880858	−0.06251004
(10, 15) (4, 3)	−0.05923653	−0.06251004
(10, 15) (6, 5)	−0.05880858	−0.06251004
(10, 15) (8, 7)	−0.05880858	−0.06251004
(10, 15) (12, 11)	−0.06645716	−0.06251004
(10, 15) (14, 13)	−0.06645716	−0.06251004
(10, 15) (18, 17)	−0.05880858	−0.06251004

[a]Ground-state energy=−0.19361445.

V. SUMMARY

We have constructed and studied a quantum spin-1/2 model on honeycomb lattice. In one limit of the interaction parameter ($K=1$), the model has an exact threefold degenerate dimer ground state. Away from the exact case ($0 \leq K < 1$), we study the evolution of the ground state using triplon mean-field theory. The mean-field theory is exact at $K=1$ and it shows a continuous quantum phase transition from the dimer-ordered to Néel-ordered ground state at $K^*=0.256$. Within this mean-field theory, the critical exponents for the spin gap and the staggered magnetization are found to be 1/2. We also have performed the exact diagonalization on an 18-site honeycomb cluster. For $K=1$, it gives the triply degenerate ground state with the same spin-spin and dimer-dimer correlations as in the exact dimer ground state.

Finally, we would also like to note that while the present model contains two-, four-, and six-spin interactions, it seems okay to ignore the six-spin terms as far as the dimer order in the ground state is concerned. The data presented in Table III shows that, although the dimer order in the ground state of K=1 model *without* six-spin terms (that is, $H^{(6)}=0$) has weakened as compared to that in the exact dimer states, the positions of singlet-correlated bonds remains unchanged (note the sign of the dimer-dimer correlation). In fact, the triplon MFT anyway ignores the six-spin interactions, and still agrees with the exact results at K=1. This simplistic observation presents a realistic hope of realizing such ground states in some honeycomb material because a combination of two-spin and four-spin interactions with suitable strengths is not completely unlikely to occur in a real system.

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*rakesh.phys@gmail.com
†bkumar@mail.jnu.ac.in

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