Chapter 4

Staggered dimerization in a honeycomb antiferromagnet

The present chapter describes an antiferromagnetic quantum spin-$S$ model on the honeycomb lattice. It consists of two terms – the nearest-neighbor Heisenberg interaction and a multiple spin-exchange term. In the absence of Heisenberg interaction, the model is exactly solvable with a three-fold degenerate dimer ground state. The present study is restricted to spin-$1/2$ case only, although the model is exactly solvable for any $S$. We employ triplon mean-field theory and exact diagonalization approaches in our investigations. Mean-field calculations suggest a continuous quantum phase transition between the staggered dimer order and the Néel order. Within triplon mean-field paradigm, the mean-field exponents for spin-gap and staggered magnetic moment around the criticality are $1/2$ for both. The exact numerical diagonalization studies confirm that there are no other states in the ground state manifold, other than the proposed dimer configurations, at the exactly solvable point, and also seem to indicate the possibility of a continuous phase transition away from the exact case.

4.1 Model Hamiltonian

We study the following quantum spin-$S$ model on the honeycomb lattice.

$$H = J \sum_{\langle ij \rangle} S_i \cdot S_j + \frac{K}{[2S(2S + 1)]^2} \sum_{\bigcirc} [S_{12}^2 S_{34}^2 S_{56}^2 + S_{23}^2 S_{45}^2 S_{61}^2] .$$ (4.1)

The operator $S_{ij}^2$ is the square of total spin on a dimer, i.e., $S_{ij}^2 = (S_i + S_j)^2$. The first term appearing
with exchange interaction $J$ is the conventional nearest-neighbor (nn) Heisenberg interaction. The second term, introduced by Brijesh Kumar, is responsible for the spontaneous dimerization in the ground state. Therefore, at $J = 0$, the model has an exact triply degenerate staggered dimerized ground state. Henceforth, we call this as the exact limit.

The multiple spin interaction is generated by the product of the alternate pairwise total spins on the three nn pairs on each hexagon plaquette of the lattice. For example, see the shaded hexagon plaquette in Fig. 4.1 in which we have two different alternate pairwise products — $(1, 2)(3, 4)(5, 6)$ and $(2, 3)(4, 5)(6, 1)$. 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 $S_{ij}^2$ as $\frac{3}{2} + 2 S_i \cdot S_j$ (for a pair of $S = 1/2$), and regrouping different terms, Eq. (4.1) 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 $H$ 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 $K \in [0, 1]$.

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1 This multiple spin interaction is similar to the interaction considered earlier on the checkerboard lattice [89].
4.2 Exactly solvable ground state

Consider the model at the exactly solvable point, $K = 1$. For spin-1/2 antiferromagnet, Hamiltonian of the model can be written as

$$H_K = \frac{1}{8} \sum \{h_{K1} + h_{K2}\},$$  \hspace{1cm} (4.2)

where $h_{K1} = S_{12}^2 S_{34}^2 S_{56}^2$ and $h_{K2} = S_{23}^2 S_{45}^2 S_{61}^2$. In this case, the nn Heisenberg exchange coming through interaction $J$ 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 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 remain as ‘free’. For example, one such state is $[1,2] \otimes [3,4] \otimes [5,6]$, 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 plaquette, it is straightforward to show that the three dimer ordered configurations shown in Fig. 4.2 form the exact zero energy ground state of $H_K$ on the full lattice. We have cross-checked this analytic assertion by numerically diagonalizing $H$ on 12-site and 18-site spin clusters with periodic boundary conditions. These dimer configurations have also been known to arise in the ground state of the quantum dimer model [90] and a frustrated $SU(2)$ spin model on honeycomb lattice [62]. 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 here.

Let the ground state 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

$$|\phi_1\rangle = \otimes_{(i,j) \in \mathcal{D}} |i, j\rangle,$$  \hspace{1cm} (4.3)

where $\mathcal{D}$ is the set of singlet forming dimers in the state $|\phi_1\rangle$. The other two states, $|\phi_2\rangle$ and $|\phi_3\rangle$,
4.3. Finite size numerical diagonalization

The thick (blue) lines denote the dimer singlets.

are related to \(|\phi_1\rangle\) via the threefold rotation as

\[
|\phi_2\rangle = C_3 |\phi_1\rangle \\
|\phi_3\rangle = C_3^2 |\phi_1\rangle
\]

where \(C_3\) is the clockwise \(2\pi/3\) rotation operator.

To this end, we would like to mention that this exact ground state of \(H_K\) is also valid for the general spin-\(S\) system. The model for general-\(S\) has been studies using spin-\(S\) bond-operator mean-field theory [91]. 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 rescaled the coupling \(K\) to \(K(2S(2S+1))^{-1}\). This just makes the energy contribution of the multiple spin interaction comparable (in powers of \(S\)) to that of the Heisenberg part.

4.3 Finite size numerical diagonalization

We have done exact numerical diagonalization of \(H\) on 12-site and 18-site honeycomb clusters with periodic boundary conditions (see Fig. 4.4). This is just to numerically verify the exact ground state on small clusters. We only use total magnetization and spin-inversion symmetries in programming codes. The exact diagonalization results clearly show that the ground state for the exactly solvable
Finite size numerical diagonalization

Figure 4.3: Eigenvalues from the exact diagonalization of $H$ in $S_z = 0$ sector. The results on 12-site honeycomb cluster is shown in left figure whereas the figure at right is for 18-site cluster.

case ($K = 1$) is indeed threefold degenerate (see Fig. 4.3). Away from the exact case, the ground state energy decreases from zero smoothly without level crossing. Moreover, the degeneracy seems to survive, for $K$ close to 1, even on a small cluster. We therefore expect the ground state to remain triply degenerate over a finite range of $K$ on thermodynamic large systems.

In order to ascertain the nature of $K = 1$ ground state, we compute the spin-spin correlation, $\langle S_i \cdot S_j \rangle$, and the dimer-dimer correlation,

$$D_{i,j,k,l} = \langle (S_i \cdot S_j)(S_k \cdot S_l) \rangle - \langle S_i \cdot S_j \rangle \langle S_k \cdot S_l \rangle.$$

The latter helps in identifying the dimer order. According to this definition, the $D_{i,j,k,l}$ is positive when the two dimers are correlated, and negative if the dimers are uncorrelated. 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. 4.2), and the dimer-dimer correlation is 1/8 when two dimers are perfect singlets.

For $K = 1$, the numerical ground state wavefunctions 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 $\hat{O}$, its thermal average is given by $\langle \hat{O} \rangle = \text{Tr}(\hat{\rho} \hat{O})$, where $\hat{\rho} = Z^{-1} e^{-\beta \hat{H}}$ is the thermal density operator ($Z = \text{Tr} e^{-\beta \hat{H}}$). In the zero temperature limit, the density operator
Table 4.2: Dimer-dimer correlation on 12-site cluster.

<table>
<thead>
<tr>
<th>Dimer</th>
<th>Exact diagonalization</th>
<th>Exact</th>
</tr>
</thead>
<tbody>
<tr>
<td>(2,1)</td>
<td>-0.244768</td>
<td>-0.244768</td>
</tr>
<tr>
<td>(2,3)</td>
<td>-0.245889</td>
<td>-0.245889</td>
</tr>
<tr>
<td>(2,4)</td>
<td>-0.008221</td>
<td>-0.008221</td>
</tr>
<tr>
<td>(2,5)</td>
<td>-0.248879</td>
<td>-0.248879</td>
</tr>
<tr>
<td>(2,6)</td>
<td>-0.002242</td>
<td>-0.002242</td>
</tr>
<tr>
<td>(2,7)</td>
<td>0.000373</td>
<td>0.000373</td>
</tr>
<tr>
<td>(2,8)</td>
<td>-0.000373</td>
<td>-0.000373</td>
</tr>
<tr>
<td>(2,9)</td>
<td>0.002242</td>
<td>0.002242</td>
</tr>
<tr>
<td>(2,10)</td>
<td>-0.002242</td>
<td>-0.002242</td>
</tr>
<tr>
<td>(2,11)</td>
<td>0.000373</td>
<td>0.000373</td>
</tr>
<tr>
<td>(2,12)</td>
<td>-0.000373</td>
<td>-0.000373</td>
</tr>
</tbody>
</table>

Table 4.1: Spin-spin correlation on 12-site cluster.

Table 4.3: Spin-singlet correlation on 12-site cluster.

Table 4.4: Dimer-singlet correlation on 12-site cluster.

\[ \hat{\rho} = \frac{1}{N_g} \sum_{\nu=1}^{N_g} |\Psi_\nu\rangle \langle \Psi_\nu| \]  

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 the Tables 4.1, 4.2, 4.3 and 4.4. The exact wavefunctions, \( |\phi_1\rangle \) etc. are not orthogonal. Therefore, we first orthogonalize them using Gram-Schmidt procedure (on the same cluster as used for numerical diagonalization; see Fig. 4.4), 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 with dimer order in the exact ground state (compare Fig. 4.2 with Fig. 4.4). The numbers from the two calculations are in excellent agreement.
### Table 4.3: Spin-spin correlation on 18-site cluster.

<table>
<thead>
<tr>
<th>Dimer</th>
<th>ED</th>
<th>Exact</th>
</tr>
</thead>
<tbody>
<tr>
<td>(10, 1)</td>
<td>0.00012025</td>
<td>0.00012025</td>
</tr>
<tr>
<td>(10, 2)</td>
<td>0.00000000</td>
<td>0.00000000</td>
</tr>
<tr>
<td>(10, 3)</td>
<td>0.00012025</td>
<td>0.00012025</td>
</tr>
<tr>
<td>(10, 4)</td>
<td>-0.00012025</td>
<td>-0.00012025</td>
</tr>
<tr>
<td>(10, 5)</td>
<td>0.00000000</td>
<td>0.00000000</td>
</tr>
<tr>
<td>(10, 6)</td>
<td>-0.00012025</td>
<td>-0.00012025</td>
</tr>
<tr>
<td>(10, 7)</td>
<td>0.00012025</td>
<td>0.00012025</td>
</tr>
<tr>
<td>(10, 8)</td>
<td>-0.00012025</td>
<td>-0.00012025</td>
</tr>
<tr>
<td>(10, 9)</td>
<td>-0.24987975</td>
<td>-0.24987975</td>
</tr>
<tr>
<td>(10,10)</td>
<td>-0.24987975</td>
<td>-0.24987975</td>
</tr>
<tr>
<td>(10,11)</td>
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<td>-0.00012025</td>
</tr>
<tr>
<td>(10,12)</td>
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</tr>
<tr>
<td>(10,13)</td>
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<td>-0.00012025</td>
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<tr>
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<td>(10,15)</td>
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</tr>
<tr>
<td>(10,16)</td>
<td>-0.00012025</td>
<td>-0.00012025</td>
</tr>
<tr>
<td>(10,17)</td>
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<td>0.00000000</td>
</tr>
<tr>
<td>(10,18)</td>
<td>0.00000000</td>
<td>0.00000000</td>
</tr>
</tbody>
</table>

### Table 4.4: Dimer-dimer correlation on 18-site cluster.

<table>
<thead>
<tr>
<th>Dimer-Dimer</th>
<th>ED</th>
<th>Exact</th>
</tr>
</thead>
<tbody>
<tr>
<td>(10,15) (2,7)</td>
<td>0.12496993</td>
<td>0.12496992</td>
</tr>
<tr>
<td>(10,15) (4,9)</td>
<td>0.12496993</td>
<td>0.12496992</td>
</tr>
<tr>
<td>(10,15) (6,11)</td>
<td>0.12496993</td>
<td>0.12496992</td>
</tr>
<tr>
<td>(10,15) (8,13)</td>
<td>0.12493986</td>
<td>0.12495990</td>
</tr>
<tr>
<td>(10,15) (12,17)</td>
<td>0.12493986</td>
<td>0.12495990</td>
</tr>
<tr>
<td>(10,15) (14,1)</td>
<td>0.12496993</td>
<td>0.12496992</td>
</tr>
<tr>
<td>(10,15) (16,3)</td>
<td>0.12496993</td>
<td>0.12496992</td>
</tr>
<tr>
<td>(10,15) (18,5)</td>
<td>0.12496993</td>
<td>0.12496992</td>
</tr>
<tr>
<td>(10,15) (2,3)</td>
<td>-0.06253008</td>
<td>-0.06251004</td>
</tr>
<tr>
<td>(10,15) (4,5)</td>
<td>-0.06253008</td>
<td>-0.06251004</td>
</tr>
<tr>
<td>(10,15) (6,1)</td>
<td>-0.06253008</td>
<td>-0.06251004</td>
</tr>
<tr>
<td>(10,15) (8,9)</td>
<td>-0.06253008</td>
<td>-0.06251004</td>
</tr>
<tr>
<td>(10,15) (12,7)</td>
<td>-0.06253008</td>
<td>-0.06251004</td>
</tr>
<tr>
<td>(10,15) (16,17)</td>
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<td>-0.06251004</td>
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<tr>
<td>(10,15) (18,13)</td>
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<tr>
<td>(10,15) (8,7)</td>
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<td>-0.06251004</td>
</tr>
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<td>(10,15) (12,11)</td>
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</tbody>
</table>
4.4 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 study of this quantum phase transition by doing triplon analysis with respect to the dimerized singlet ground state. Although the exact dimer ground ground state is threefold degenerate, we can take one of these as the reference state to carry out the triplon mean-field analysis. This is of course a limitation when there are degenerate dimer configurations and we want to take some appropriate orthonormalized linear combination of those. However, we adopt this limited approach to get some idea of the effect of nearest-neighbor Heisenberg interaction on the exact dimer ground state.

We consider the dimer state $|\phi_1\rangle$ (see Fig. 4.5) as the reference state for the triplon mean-field analysis. Then, the Hamiltonian Eq. (4.1) for spin-1/2 reduces in the following form

$$H = H_J + H_K$$  \hspace{1cm} (4.7)

with

$$H_J = J \sum_R \left[ S_1(R) \cdot S_2(R) + S_1(R) \cdot S_2(R - \frac{3}{2} a\hat{x} + \frac{\sqrt{3}}{2} a\hat{y}) + S_2(R) \cdot S_1(R + \frac{3}{2} a\hat{x} + \frac{\sqrt{3}}{2} a\hat{y}) \right]$$
4.4. Triplon mean-field theory

Now re-writing the Hamiltonian Eq. (4.7) in terms of bond operators and taking mean-field approximations as discussed in Sec. 2.1.2, we obtain the following mean-field Hamiltonian in momentum space.

\[ H_{mf} = E_0 + \frac{1}{2} \sum \sum \left\{ (\lambda - s^2 \xi_k) \left[ t^{\dagger}_\alpha(k) t_\alpha(k) + t_\alpha(-k) t^{\dagger}_\alpha(-k) \right] \right. \\
\left. - s^2 \xi_k \left[ t^{\dagger}_\alpha(k) t_\alpha(-k) + t_\alpha(-k) t^{\dagger}_\alpha(k) \right] \right\} \]  \hspace{1cm}(4.8)

In the above equation, k-summation is on all discrete k-points lying inside the first Brillouin zone.
of the lattice and constant $E_0$, effective chemical potential $\lambda$, and $\xi$ have the following forms.

$$E_0 = \frac{L}{2} \left[ \frac{J}{4} + \frac{9}{8} K - \left( J + \frac{9}{8} K \right) \bar{s}^2 - \frac{5}{2} \lambda + \lambda \bar{s}^2 \right], \quad (4.9)$$

$$\lambda = \frac{1}{4} \left( J + \frac{9}{8} K \right) - \mu, \quad (4.10)$$

$$\xi_k = \left[ J + \frac{9}{8} K (1 - \bar{s}^2) \right] \cos \left( \frac{3}{2} k_x \right) \cos \left( \frac{\sqrt{3}}{2} k_y \right). \quad (4.11)$$

We diagonalize it by Bogoliubov transformation as done in Sec. 3.2, we obtain the following diagonalized mean-field Hamiltonian,

$$H_{mf} = E_0 + \sum_k \sum_{\alpha} E_k \left( \gamma_{\alpha}^\dagger(k) \gamma_{\alpha}(k) + \frac{1}{2} \right) \quad (4.12)$$

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

$$e_g [\lambda, \bar{s}^2] = e_0 + \frac{3}{2L} \sum_k E_k \quad (4.13)$$

where $e_0 = E_0/L$. Minimization of the ground state energy with respect to unknown mean-field parameters $\lambda$ and $\bar{s}^2$ gives coupled self-consistent equations. Solution of these equations leads to mean-field results.

### 4.4.1 Staggered dimer phase

When the minima of the triplon dispersion is nonzero, the dimer phase is stable against triplon excitations. Therefore, gapped triplon phase corresponds to dimer ground state. The saddle point equations $\partial_\lambda e_g = 0$ and $\partial_{\bar{s}^2} e_g = 0$ give the following self-consistent equations

$$\bar{s}^2 = \frac{5}{2} - \frac{3}{L} \sum_k \frac{\lambda - \bar{s}^2 \xi_k}{E_k}, \quad (4.14)$$

$$\lambda = J + \frac{9}{8} K + \frac{3\lambda}{L} \sum_k \frac{\eta_k}{E_k}, \quad (4.15)$$

where

$$\eta_k = \xi_k - \frac{9}{8} K \bar{s}^2 \cos \left( \frac{3}{2} k_x \right) \cos \left( \frac{\sqrt{3}}{2} k_y \right). \quad (4.16)$$

The parameter $\bar{s}$ measures the average amplitude of the singlet condensation on a dimer. If all the dimers form perfect singlets as in our exactly solvable limit of the model, we get $\bar{s}^2 = 1$. The average amplitude of singlet condensation will reduce however, when triplons fluctuate in the ground state.
4.4. Triplon mean-field theory

Figure 4.6: Triplon dispersion $E_k$ for different values of $K$. The $\Gamma$, $A$, 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.

4.4.2 Néel ordered phase

In the magnetically ordered phase, dispersion touches zero at some $k$-points, denoted as $Q$. For the present case, the ordering wave vector at which dispersion vanishes is $Q = (0,0)$ (see Fig. 4.6). It means the triplon occupancy at the wave vector $Q$ becomes singular, which implies the Bose condensation of triplons at $Q$. Thus, we need to introduce a third mean-field parameter, in addition to $\lambda$ and $\tilde{s}^2$, the triplon condensate density, $n_c$, which is defined by

$$n_c = \frac{2}{L} \sum_{\alpha} \langle t_{\alpha}^\dagger(Q)t_{\alpha}(Q) \rangle = \frac{3}{L} \left( \frac{\lambda - \tilde{s}^2 \xi_Q}{E_Q} \right).$$  \hfill (4.17)

Now, we get the following self-consistent equations

$$\lambda = 2 \tilde{s}^2 \xi_Q, \hfill (4.18)$$

$$n_c = \frac{1}{2 \eta_Q} \left[ \lambda - \frac{3 \lambda}{L} \sum_{k\neq Q} \frac{\eta_k}{E_k} \left( J + \frac{9}{8} K \right) \right], \hfill (4.19)$$

$$\tilde{s}^2 = \frac{5}{2} - n_c - \frac{3}{L} \sum_{k\neq Q} \frac{\lambda - \tilde{s}^2 \xi_k}{E_k}. \hfill (4.20)$$

The nonzero triplon density physically corresponds to an antiferromagnetic 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 $M_s = \tilde{s} \sqrt{n_c}$. 
4.5 Results and Discussion

In this section, we present the results obtained by the triplon mean-field calculations. The self-consistent Eqs. (4.14) and (4.15) of the gapped dimer phase are solved for $\lambda$ and $s^2$, for different values of $K$. Interestingly, for $K = 1$, it gives $s^2 = 1$, same as the exact value (see Fig. 4.7). Moreover, the mean-field and exact ground state energies are both zero, as shown in Fig. 4.8. At the exact limit, the triplon dispersion, $E_k$ (plotted in Fig. 4.6), 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:

$$\Delta_{(K=1)} = \frac{\langle \tau | H_K | \tau \rangle}{\langle \tau | \tau \rangle}, \quad (4.21)$$

where $| \tau \rangle$ is a “trial” localized triplon state as defined below

$$| \tau \rangle = \bigotimes_{(i,j) \in D'} [i,j] \otimes \{k,l\}. \quad (4.22)$$

Here, $D'$ is the set of all dimers in $D$ except $(k,l)$ and $\{k,l\}$ denotes a triplet state on the bond $(k,l)$. The set $D$ is the set of all singlets forming dimers in $\phi_1$ [see Eq. (4.3)].

For $K < 1$, dispersion curve, $E_k$, acquires a finite width, with minimum at the $\Gamma$ point [that is, $Q$]. Eventually, it touches zero at $Q$ and remains so below $K^* = 0.256$. For $K < K^*$, we solve self-consistent Eqs. (4.18), (4.19), and (4.20). As shown in Fig. 4.9, now the staggered
Figure 4.8: The ground state energy from the triplon mean-field theory and the exact diagonalization (12-site and 18-site clusters). The two compare well close to $K = 1$ as system size for exact diagonalization is 18-site.

Figure 4.9: The spin gap, $\Delta = E_Q$, and the staggered moment $M_s$. Inset: the triplon condensate density.
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 [57], 0.266 from series expansion [59], and 0.22 from Monte-Carlo simulations [58]. 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.

$$K^* = 0.256$$

Figure 4.10: The mean-field quantum phase diagram of $H$ for spin-1/2.

The triplon mean-field theory thus predicts a continuous transition from the dimer to Néel-ordered ground state of $H$ for spin-1/2. The phase diagram is just a line presented in Fig. 4.10. As discussed 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 $S \rightarrow \infty$ (the classical limit). Recently, the model for general-$S$ has been studied using spin-$S$ bond-operator mean-field theory [91]. If we do classical study by considering spins as classical vectors or a semi-classical analysis by linear spin-wave theory, we get always get ordered phase, that is, Néel in the entire range of $K$. Both interactions, $H_J$ and $H_K$, contain only nearest spin exchange terms. The structure of the Hamiltonian permits only oppositely polarized spins on a nearest neighbor bond to have minimum classical energy. Of course, not a unique spin configuration holds this condition. Since there are infinite ways the spins on a nearest neighbor bond can be in antiparallel. So classically infinitely large degeneracy occurs in the ground state spin configuration. In linear spin-wave theory, because of Hamiltonian structure, no competing terms appear to destabilize the ordered Néel state. Hence, we always get critical point at $K^* = 1$. The order and disorder transition in our case is purely happening because of the quantum mechanical effects which play very crucial roles in stabilizing singlet dimer order against a Néel order at sufficient large coupling strength $K$. 
Critical Exponents

Here we compute the critical exponents of the spin-gap and staggered magnetic moment within the triplon mean-field theory. Near to critical point that is $K^*$, the critical exponents for spin-gap and staggered magnetic moment are defined as

$$\text{Spin-gap: } \Delta \sim |K - K^*|^{\nu z}, \quad (4.23)$$

$$\text{Staggered magnetic moment: } M_s \sim |K - K^*|^{\beta}, \quad (4.24)$$

where $z$ is the dynamical exponent and $\nu$ is the correlation length exponent. For Lorentz invariant systems, the dynamical exponent $z = 1$. A Lorentz invariant system is the one where both space and time directions are equivalent and most systems occurring in nature generally are Lorentz invariant.

Close to the quantum critical point, we have almost linear variations in $\lambda$ and $s^2$ with respect to $K$. We linearly expand these quantities and find the relations in critical exponents as given below.

4.5.0.1 Spin-gap

Infinitesimally away from the quantum critical point $K^*$, in the dimerized singlet phase, we do the following replacements

$$K \rightarrow K^* + \epsilon, \quad (4.25)$$

$$\lambda \rightarrow \lambda^* - |g_\lambda| \epsilon, \quad (4.26)$$

$$s^2 \rightarrow s^*^2 + g_s \epsilon, \quad (4.27)$$

where $g_\lambda$ and $g_s$ are gradients of $\lambda$ and $s^2$ respectively in Fig. 4.7. Quantities with asterisks are their respective values at the quantum critical point.

Now from the definition of spin-gap $\Delta = \sqrt{\lambda (\lambda - 2s^2 \xi_Q)}$, we need $\lambda$, $s^2$, and $\xi_Q$. For this, we have

$$\xi_Q = J + \frac{9}{8}K (1 - \bar{s}^2) \quad (4.28)$$

$$= 1 + \frac{1}{8}K - \frac{9}{8} \bar{s}^2 \quad (\because J + K = 1) \quad (4.29)$$

$$= 1 + \frac{1}{8} (K^* + \epsilon) - \frac{9}{8} (K^* + \epsilon) (s^*^2 + g_s \epsilon) \quad (4.30)$$

$$\approx \xi_Q^* + \frac{1}{8} \epsilon \left[ 1 - 9 \left( g_s K^* + \bar{s}^*^2 \right) \right] \quad (4.31)$$
4.5. Results and Discussion

where $\xi_Q^* = 1 + \frac{1}{8} K^* - \frac{9}{8} K^* \bar{s}^2$. In the last step of above equation, we ignored the terms of higher order $O(\epsilon^2)$ as $\epsilon$ is very small. With this approximation, we get

$$\Delta = \sqrt{(\lambda^* - |g_\lambda| \epsilon) \left\{ (\lambda^* - |g_\lambda| \epsilon) - 2 (\bar{s}^2 + g_s \epsilon) \left[ \xi_Q^* + \frac{1}{8} \epsilon [1 - 9 (g_s K^* + \bar{s}^2)] \right] \right\} } \quad (4.32)$$

$$\approx \left\{ -\lambda^* \left[ g_\lambda + 2 g_s \xi_Q^* + \frac{1}{4} \bar{s}^2 - \frac{9}{4} g_s K^* \bar{s}^2 - \frac{9}{4} \bar{s}^2 \right] \right\}^{1/2} \epsilon^{1/2} \quad (4.33)$$

$$= 0.12 \epsilon^{1/2} \quad (4.34)$$

$$= 0.12 (K - K^*)^{1/2} \quad (4.35)$$

In the Eq. (4.33), we used condition $\lambda^* = 2 \bar{s}^2 \xi_Q^* \text{[which is Eq. (4.18) at the quantum critical point]}$ and ignored higher order terms $O(\epsilon^2)$.

4.5.0.2 Staggered magnetic moment

Moving away from the quantum critical point towards the magnetically ordered phase (Neel), close to $K^*$, we do the following substitutions.

$$K \rightarrow K^* - \epsilon \quad , \quad (4.36)$$

$$\lambda_Q \rightarrow \lambda_Q^* + g_\lambda \epsilon \quad , \quad (4.37)$$

$$\bar{s}^2 \rightarrow \bar{s}^2 - |g_s| \epsilon \quad . \quad (4.38)$$

From Eq. (4.19), we have

$$n_c = \frac{\lambda_Q - (1 + \frac{1}{2} K)}{2 \eta_Q} - \frac{3}{2 L} \sum_{k \neq Q} \frac{\cos \left( \frac{3}{2} k_x \right) \cos \left( \frac{\sqrt{3}}{2} k_y \right)}{\sqrt{1 - \cos \left( \frac{3}{2} k_x \right) \cos \left( \frac{\sqrt{3}}{2} k_y \right)}} \quad . \quad (4.39)$$

In the above equation, we used the following relations

$$\eta_k = \eta_Q \cos \left( \frac{3}{2} k_x \right) \cos \left( \frac{\sqrt{3}}{2} k_y \right) \quad , \quad (4.40)$$

$$\xi_k = \xi_Q \cos \left( \frac{3}{2} k_x \right) \cos \left( \frac{\sqrt{3}}{2} k_y \right) \quad , \quad (4.41)$$

$$E_Q = \sqrt{\lambda_Q (\lambda_Q - 2 \bar{s}^2 \xi_k)} \quad , \quad (4.42)$$

$$\lambda_Q = 2 \bar{s}^2 \xi_Q \quad . \quad (4.43)$$
At the quantum critical point, we have $n_c = 0$ and so this leads to the following condition

$$
\frac{3}{2L} \sum_{k \neq Q} \frac{\cos \left( \frac{3}{2} k_x \right) \cos \left( \frac{\sqrt{3}}{2} k_y \right)}{\sqrt{1 - \cos \left( \frac{3}{2} k_x \right) \cos \left( \frac{\sqrt{3}}{2} k_y \right)}} = \frac{\lambda^*_Q - \left(1 + \frac{1}{8} K^* \right)}{2 \eta_Q^*}.
$$

Using the condition (4.44), from Eq. (4.39) we obtain

$$
\eta_c = \frac{\lambda_Q - (1 + \frac{1}{8} K)}{2 \eta_Q} - \frac{\lambda^*_Q - (1 + \frac{1}{8} K^*)}{2 \eta_Q^*}
$$

$$
= \frac{1}{2 \eta_Q \eta_Q^*} \left\{ \left[ \lambda_Q - \left(1 + \frac{1}{8} K \right) \right] \eta_Q^* - \left[ \lambda^*_Q - \left(1 + \frac{1}{8} K^* \right) \right] \eta_Q \right\}
$$

$$
\approx \frac{1}{2 \eta_Q^2} \left\{ \left[ \lambda_Q - \left(1 + \frac{1}{8} K \right) \right] \eta_Q^* - \left[ \lambda^*_Q - \left(1 + \frac{1}{8} K^* \right) \right] \eta_Q \right\}
$$

$$
\approx \frac{1}{2 \eta_Q^2} \left\{ \left[ \frac{1}{8} + g_\lambda \right] \eta_Q^* - \left[ \lambda^*_Q - \left(1 + \frac{1}{8} K^* \right) \right] \left[ -\frac{1}{8} \right] + \frac{9}{4} \left| g_\lambda \right| K^* + \tilde{s}^2 \right\}.
$$

Hence, staggered magnetic moment near to criticality is given by

$$
M_s \approx \frac{\tilde{s}^* \epsilon^2}{\sqrt{2} \eta_Q} \left\{ \left[ \frac{1}{8} + g_\lambda \right] \eta_Q^* - \left[ \lambda^*_Q - \left(1 + \frac{1}{8} K^* \right) \right] \left[ -\frac{1}{8} \right] \left[ \frac{9}{4} \left| g_\lambda \right| K^* + \tilde{s}^2 \right] \right\}^{\frac{1}{2}}
$$

$$
= 0.44 \epsilon^2
$$

$$
= 0.44 \left( K^* - K \right)^{\frac{1}{2}}.
$$

We plotted the estimated results and mean-field data around the criticality in Fig. 4.11. 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 $N \to \infty$ [92]. It is more than the numerically computed value of 0.34 for a planar antiferromagnet [93]. Also, for spin-gap we get $\nu = 1/2$, as compared to the value of 1 from the large $N$ sigma model theory [92] and the numerically computed values of 0.69 for a quantum antiferromagnet on CaV$_4$O$_9$ structure [93] and 0.63 for the staggered dimer model on square lattice [94].

4.6 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 12-site and 18-site honeycomb clusters. 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.

The model as discussed in Sec. 4.1 contains two-spin, four-spin, and six-spin interactions. If we ignore the six-spin terms at \( K = 1 \), it seems okay as far as the dimer order in the ground state is concerned. The data presented in Appendix 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 the singlet-correlated bonds remains unchanged (note the sign of the dimer-dimer correlation). In fact, the triplon mean-field anyways ignores the six-spin interactions, still agrees with the exact results at \( K = 1 \). This simplistic observation presents a realistic hope for realizing such ground states in some honeycomb materials because a combination of two-spin and four-spin interactions with suitable strengths is not completely unlikely to occur in a real system. Very recently, numerous other studies on honeycomb antiferromagnets also predict the staggered dimer order [95, 96, 97].

Figure 4.11: The spin-gap and staggered magnetization near the critical point. The estimate results of Eqs. (4.35) and (4.51) are also plotted.