Chapter 5

Degenerate Shastry-Sutherland state on square lattice

In a recent work [Phys. Rev. B 77, 014419 (2008)], a quantum spin-1/2 model having an exact fourfold degenerate Shastry-Sutherland ground state was constructed, and studied using finite-size numerical exact diagonalization. There, a schematic quantum phase diagram suggesting many competing phases was also proposed. In the present chapter, we consider an extended version of this model and investigated using the dimer and plaquette triplon mean-field theories. The corresponding quantum phase diagram (in the thermodynamic limit) is found to have many interesting phases: commensurate and incommensurate magnetically ordered phases, and columnar dimer, Shastry-Sutherland, sublattice columnar dimer (sublattice-CDS), and plaquette spin-gapped phases. Among themselves, these phases undergo a continuous or a level-crossing transition.

5.1 The Model

We consider the following spin-1/2 antiferromagnet on the square lattice:

\[
\mathcal{H} = J_1 \sum_{(i,j)} S_i \cdot S_j + J_2 \sum_{\langle(i,j)\rangle} S_i \cdot S_j + J_3 \sum_{\langle\langle(i,j)\rangle\rangle} S_i \cdot S_j \\
+ K \sum_{[i,j,k,l,m,n]} \mathcal{P}_{3/2}(i,j,k) \mathcal{P}_{3/2}(l,m,n). \tag{5.1}
\]

The couplings \(J_1\), \(J_2\), and \(J_3\) are respectively first, second, and third nearest-neighbor antiferromagnetic Heisenberg interactions. The sum over \([i,j,k,l,m,n]\) denotes as the sum on all horizontal
5.1.1 The Model

and vertical plaquettes. On each plaquette, we have two oppositely oriented triangles with vertices \(i, j, k\) and \(l, m, n\). The vertices of each triangle separately belong to same sublattice of the square lattice (see Fig. 5.1). By construction, the interaction \(K\) is made out of second and third nearest neighbor interactions, so the only competing terms with it are \(J_2\) and \(J_3\). For this reason, we also included the \(J_3\) interaction in the Hamiltonian. In original model (see Ref. [32]), the explicit \(J_3\) interaction was not present.

The multiple spin-exchange interaction, \(K\), projects four SS dimer states (see Fig. 5.2). This interaction represents interaction between two projection operators of quartet states. The projection operators are on oppositely oriented triangular plaquettes. When the total spin on a triangle form doublet, these projector operators washed out those states. Alternatively, this means that on acting a doublet state, these operators give zero eigenvalue. A projection operator on the quartet state is defined as

\[
P_{3/2}(i, j, k) = \frac{1}{3} \left[ (S_i + S_j + S_k)^2 - \frac{3}{4} \right] = \frac{1}{2} + \frac{2}{3} (S_i \cdot S_j + S_j \cdot S_k + S_k \cdot S_i).
\] (5.2)

The advantage of the projector operator \(P_{3/2}\) is that either it gives the same state with eigenvalue unity or simply annihilates the state on which it is acting. So the product of two projector operators will definitely be positive. Thus, the minima of \(K\) interaction will be zero. This happens when any one triangle forms a doublet state by forming one single dimer on it. This is an essential part of making exact fourfold degenerate SS dimer state in the present Hamiltonian.

Using the definition of projection operator given by Eq. (5.2), the Hamiltonian Eq. (5.1) can be simplified as

\[
\mathcal{H} = \frac{1}{2} KL + J_1 \sum_{(i,j)} S_i \cdot S_j + \tilde{J}_2 \sum_{(i,j)} S_i \cdot S_j + \tilde{J}_3 \sum_{(i,j)} S_i \cdot S_j + \frac{4}{9} K \sum_{[i,j,k,l,m,n]} h(i, j, k) h(l, m, n)
\] (5.3)

where \(L\) is the number of lattice sites, \(\tilde{J}_2 = J_2 + \frac{4}{3} K\), \(\tilde{J}_3 = J_3 + \frac{2}{3} K\), and \(h(i, j, k) = S_i \cdot S_j + S_j \cdot S_k + S_k \cdot S_i\).
5.1. The Model

Figure 5.1: This figure shows the pictorial representation of the Hamiltonian Eq. (5.1). Filled circles and squares denote the spins on two sublattices. Broken lines represent $J_1$ interactions whereas solid lines $J_2$ interactions. Similarly, $J_3$ interactions can be understood (not shown here). The shaded triangles with opposite orientations represent the multiple spin-exchange interaction $K$.

Figure 5.2: The four SS dimer states. These states spontaneously break the lattice symmetry and connected by lattice translations in $x$ and $y$ directions.
5.2 Triplon mean-field theory

The model is exactly solvable at the limit \( J_1 = J_2 = J_3 = 0 \). At this exact known point, we have fourfold degenerate SS dimer state. The triplon mean-field theory [44] is a very useful theoretical technique in a situation where bonded spins form frozen singlet dimers spontaneously. The present Hamiltonian is also fulfilling this requirement. Thus, we formulate the triplon mean-field theory for this Hamiltonian by taking one reference dimer state. We also noticed that, in some particular region of quantum phase diagram, the plaquette crystal phase is energetically favorable than CDS states. This predictions comes from another version of triplon mean-field theory — plaquette triplon mean-field theory [46]. With a brief description of these two approaches, we present the mean-field formulations of the Hamiltonian on a chosen reference dimer state.

5.2.1 Dimer triplon mean-field theory

5.2.1.1 On Shastry-Sutherland dimer state

![Figure 5.3](image-url)

Figure 5.3: Figure (a) shows the Shastry-Sutherland dimer state, figure (b) is its unit cell. The corresponding Brillouin zone is shown in figure (c).

We choose the reference SS dimer state shown in Fig. 5.3. In principle, we can choose any one SS dimer state out of four. For the sake of generality, we assume different singlet condensation on each dimer within the unit cell. Lets say the singlet condensation amplitudes on the dimers (1)
and (2) are some \(c\)-numbers \(s_1\) and \(s_2\) respectively. We also used distinct type of triplet operators on these dimers, say \(t_1^{(1)}\) and \(t_2^{(2)}\). The global constraints are imposed by introducing chemical potentials \(\mu^{(1)}\) and \(\mu^{(2)}\) on the dimers of the unit cell:

\[
- \sum_{R} \left\{ \mu_1 \left[ s_1^2 + \sum_{\alpha} t_1^{(1)\dagger}(R) t_1^{(1)}(R) - 1 \right] + \mu_2 \left[ s_2^2 + \sum_{\alpha} t_2^{(2)\dagger}(R) t_2^{(2)}(R) - 1 \right] \right\}.
\]

These chemical potentials fix the number of bosons. We also ignored the triplet-triplet interactions as they change the results very minutely. This leads to quadratic form in triplet operators. Using these simplifications and approximations, the mean-filed Hamiltonian can be written as

\[
H_{mf} = E_0 + H_{mf}^k, \tag{5.5}
\]

where,

\[
E_0 = \frac{1}{4} L \left[ 2K + \frac{1}{2} \tilde{J} - \frac{5}{2} \left( \lambda^{(1)} + \lambda^{(2)} \right) - \tilde{J} \left( s_1^2 + s_2^2 \right) + \lambda^{(1)} s_1^2 + \lambda^{(2)} s_2^2 \right] \tag{5.6}
\]

\[
H_{mf}^k = \frac{1}{2} \sum_{k, \alpha} \sum_{i=1,2} \left[ \left( \lambda(i) - \epsilon_{k}^{(i)} \right) \left( t_{k\alpha}^{(i)\dagger} t_{-k\alpha}^{(i)} + t_{-k\alpha}^{(i)\dagger} t_{k\alpha}^{(i)} \right) \right.
\]

\[
- \epsilon_{k}^{(i)} \left( t_{k\alpha}^{(i)\dagger} t_{-k\alpha}^{(i)\dagger} + t_{-k\alpha}^{(i)\dagger} t_{k\alpha}^{(i)} \right). \tag{5.7}
\]

The renormalized effective chemical potentials \(\lambda(i)\) and \(\epsilon_{k}^{(i)}\) are give by following equations

\[
\lambda(i) = \frac{1}{4} \tilde{J} - \mu^{(i)} \tag{5.8}
\]

\[
\epsilon_{k}^{(i)} = \frac{1}{2} \tilde{J} \left( \cos 2k_x + \cos 2k_y + \cos 2k_x \cos 2k_y \right)
\]

\[
- \frac{2}{3} K s_i^2 \cos 2k_x \cos 2k_y . \tag{5.9}
\]

A close inspection of Eqs. (5.7) tells us that the mean-field Hamiltonian decoupled into independent triplet operators that is no mixing of triplet operators \(t_1^{(1)}\) and \(t_1^{(2)}\). So, we can further simplify Eqs. (5.6), (5.7), (5.8), and (5.9) by taking \(s_1 = s_2 = s\), \(t^{(1)} = t^{(2)} = t\), and \(\mu^{(1)} = \mu^{(2)} = \mu\). We diagonalize the resultant mean-field Hamiltonian by unitary Bogoliubov transformation:

\[
t_{k\alpha} = \gamma_{k\alpha} \cos \theta_k + \gamma_{-k\alpha}^\dagger \sinh \theta_k, \quad \gamma_{k\alpha} \text{ and } \gamma_{-k\alpha} \text{ are Bogoliubov bosons.}
\]

The diagonalized mean-field Hamiltonian thus obtain is following

\[
H_{mf} = E_0 + 2 \sum_{k, \alpha} E_k \left( \gamma_{k\alpha}^{\dagger} \gamma_{k\alpha} + \frac{1}{2} \right). \tag{5.10}
\]

In the above equation, \(E_k = \sqrt{\lambda (\lambda - 2 s^2 \epsilon_k^i)} \geq 0\) is triplon quasiparticle dispersion. The energy of the mean-field Hamiltonian Eq. (5.10), in the absence of quasi particles, gives ground state
5.2. Triplon mean-field theory

5.2.1.2 On columnar dimer state

The triplon mean-field theory predicts columnar dimer state on the square lattice in the range $0.19 \lesssim J_2/J_1 \lesssim 0.67$ (see Ref. [44]). The present Hamiltonian also has these two interactions, $J_1$ and $J_2$. In this regard, we formulate triplon mean-field theory on the reference columnar dimer state shown in Fig. 5.4. This dimer state has single dimer per unit cell. We assume singlet Bose condensation on the singlet dimers (shown thick line segments in Fig. 5.4) always. Let it be $\bar{s}$.

We ignore triplon interactions and the constraint, $\bar{s}^2 + t_\alpha^\dagger t_\alpha = 1$, is applied globally by choosing chemical potential $\mu$ on each bonded spins uniformly: $\mu \sum_R [\bar{s}^2 + \sum_\alpha t_\alpha^\dagger(R)t_\alpha(R) - 1]$, where $R$ denotes position of a dimer. The triplon mean-field Hamiltonian of the model on the CDS state is now can written as

$$\mathcal{H}_{mf} = E_0 + H_{mf}^k,$$

(5.11)
5.2. Triplon mean-field theory

where

\[
E_0 = \frac{1}{2} L \left[ K + \frac{1}{4} J_1 - J_1 \bar{s}^2 - \frac{5}{2} \lambda + \lambda \bar{s}^2 \right]
\]  
\( (5.12) \)

\[
H_{mf}^k = \frac{1}{2} \sum_{k, \alpha} \left[ (\lambda - \bar{s}^2 \xi_k) \left( t_{\alpha}^{\dagger} t_{\alpha} + t_{-\alpha}^{\dagger} t_{-\alpha} \right) - \bar{s}^2 \xi_k \left( t_{\alpha}^{\dagger} t_{-\alpha}^{\dagger} + t_{-\alpha} t_{\alpha} \right) \right].
\]  
\( (5.13) \)

In the above equations, \( \lambda = \frac{1}{2} J_1 - \mu \) is effective chemical potential and \( \xi_k \) is given by

\[
\xi_k = \frac{1}{2} J_1 (\cos 2k_x - 2 \cos k_y) + \tilde{J}_2 (\cos 2k_x \cos k_y + \cos k_y)
- \tilde{J}_3 (\cos 2k_x + \cos 2k_y).
\]  
\( (5.14) \)

The mean-field Hamiltonian is diagonalized by unitary Bogoliubov canonical transformation: \( t_{\alpha} = \gamma_{\alpha} \cosh \theta_k + \gamma_{-\alpha}^{\dagger} \sinh \theta_k \). The diagonalized mean-field Hamiltonian can now be written as

\[
H_{mf} = E_0 + \sum_{k, \alpha} E_k \left( \gamma_{\alpha}^{\dagger} \gamma_{\alpha} + \frac{1}{2} \right).
\]  
\( (5.15) \)

The dispersion relation is given by \( E_k = \sqrt{\lambda (\lambda - 2 \bar{s}^2 \xi_k)} \). Here, \( \gamma \) are Bogoliubov quasi-excitations. The minimization of the ground state (i.e., energy of the Hamiltonian in the absence of quasi-excitations) with respect to \( \bar{s} \) and \( \lambda \) gives the coupled self-consistent equations. The solution of which gives mean-field results.

5.2.1.3 On sublattice columnar dimer state

For \( J_1 = 0 \) case, we formulated the dimer triplon mean-field theory on the columnar dimer state which is formed by spins of the same sublattice (see Fig. 5.5). This appears to be a natural choice in the present case. To show it, consider \( J_2-J_3 \) Heisenberg antiferromagnetic model on square lattice. In this case, the Hamiltonian decouples into two independent parts each of which corresponds to one sublattice. The independency comes from the fact that the spins of two sublattices are not interacting with each other. Furthermore, the Hamiltonian on each sublattice is like the \( J_1-J_2 \) model on the square lattice. Moreover, we know that the \( J_1-J_2 \) model has a columnar dimer state in the intermediate region [44]. So, the sublattice-CDS as shown in Fig. 5.5 would be a good dimer state in the case of \( J_2-J_3 \) Heisenberg antiferromagnetic model on the square lattice. We also know that the \( K \)-part of the Hamiltonian Eq. (5.1) has fourfold degenerate SS dimer state. So we study \( J_2-J_3-K \) model to see the competition among sublattice-CDS, SS, and magnetically ordered phases.
5.2. Triplon mean-field theory

Figure 5.5: This figure shows a sub-lattice columnar dimer state. We choose $x$ and $y$ axes as depicted in the figure. There are two dimers per unit cell and shown in shaded region.

Since there are two dimers in the unit cell, we choose two different singlet condensation amplitudes (say, $s$ and $p$) and triplets ($t_\alpha$ and $q_\alpha$). The mean-field Hamiltonian can now be written as

$$H_{mf} = E_0 + \sum_{k, \alpha} \left[ \Lambda_s \left( t_{k\alpha}^* t_{k\alpha} + t_{-k\alpha}^* t_{-k\alpha}^* \right) + \Delta_s \left( t_{k\alpha}^* t_{-k\alpha} + t_{-k\alpha}^* t_{k\alpha} \right) \right]$$
$$+ \sum_{k, \alpha} \left[ \Lambda_p \left( q_{k\alpha}^* q_{k\alpha} + q_{-k\alpha}^* q_{-k\alpha}^* \right) + \Delta_p \left( q_{k\alpha}^* q_{-k\alpha} + q_{-k\alpha}^* q_{k\alpha} \right) \right], \quad (5.16)$$

where,

$$E_0 / N = 2K - 3 \left( \frac{1}{4} s^2 + \frac{1}{4} p^2 \right) j_2 + (1 - s^2) \mu_s + (1 - p^2 ) \mu_p + \frac{1}{2} K s^2 p^2$$
$$- \frac{3}{2} \left( \frac{1}{2} j_2 - \frac{1}{6} K s^2 - \frac{1}{6} K p^2 - \mu_s - \mu_p \right), \quad (5.17)$$

$$\Lambda_s = \frac{1}{2} \left( \frac{1}{4} j_2 - \mu_s - \frac{1}{6} K p^2 \right) + \Delta_s, \quad (5.18)$$

$$\Lambda_p = \frac{1}{2} \left( \frac{1}{4} j_2 - \mu_p - \frac{1}{6} K s^2 \right) + \Delta_p. \quad (5.19)$$
5.2. Triplon mean-field theory

\[ \Delta_s = \frac{1}{4} \tilde{J}_2 \tilde{s}^2 (2 \cos k_y - \cos 2k_x) - \frac{1}{2} \tilde{J}_3 \tilde{s}^2 (1 + \cos 2k_x) \cos k_y \]
\[ + \frac{1}{6} K \tilde{s}^2 \tilde{p}^2 [(1 + \cos k_y) \cos 2k_x - \cos k_y], \quad (5.20) \]
\[ \Delta_p = \frac{1}{4} \tilde{J}_2 \tilde{p}^2 (2 \cos k_y - \cos 2k_x) - \frac{1}{2} \tilde{J}_3 \tilde{p}^2 (1 + \cos 2k_x) \cos k_y \]
\[ + \frac{1}{6} K \tilde{s}^2 \tilde{p}^2 [(1 + \cos k_y) \cos 2k_x - \cos k_y]. \quad (5.21) \]

Now, the diagonalized Hamiltonian can be written as the following form

\[ H_{mf} = E_0 + 2 \sum_{k,\alpha} E_k^\alpha \left( \gamma_{k\alpha} \gamma_{\kappa\alpha} + \frac{1}{2} \right) + 2 \sum_{k,\alpha} E_k^\eta \left( \eta_{k\alpha} \eta_{\kappa\alpha} + \frac{1}{2} \right), \quad (5.22) \]

where \( \gamma \) and \( \eta \) are Bogoliubov bosons, and \( E_k^\alpha = \sqrt{\Lambda^2_k - \Delta^2_s} \geq 0 \) and \( E_k^\eta = \sqrt{\Lambda^2_p - \Delta^2_p} \geq 0 \) are dispersion relations.

5.2.2 Plaquette triplon mean-field theory

\[ \mathcal{H}_p = J_1 (S_1 \cdot S_2 + S_2 \cdot S_3 + S_3 \cdot S_4 + S_4 \cdot S_1) + J_2 (S_1 \cdot S_3 + S_2 \cdot S_4). \quad (5.23) \]
5.2. Triplon mean-field theory

Let the total spin on bonds (13) and (24) are \( S_{13} \) and \( S_{24} \) respectively. Then, the total spin of the plaquette will be

\[
S = S_{13} + S_{24} ; \quad (S_{13} = S_1 + S_3 \quad \text{and} \quad S_{24} = S_2 + S_4).
\]

(5.24)

Using Eq. (5.24), we can rewrite Eq. (5.23) as follows

\[
\mathcal{H}_p = \frac{J_1}{2}(S^2 - S^2_{12} - S^2_{34}) + J_2(S_1 \cdot S_3 + S_2 \cdot S_4).
\]

(5.25)

The obtained energy spectrum of the Hamiltonian \( \mathcal{H}_p \) is given in Table (5.1). If \( J_1 \) is greater than \( J_2 \), we can restrict ourselves in the restricted subspace of the singlet state with energy \(-2J_1 + \frac{1}{2}J_2 \) and the three degenerate triplet states having energy \(-J_1 + \frac{1}{2}J_2 \). These states can viewed as the states emerging out of vacuum by the boson creation operators as

\[
|s\rangle = \frac{1}{\sqrt{3}} \left( [1, 2] \otimes [4, 3] + [1, 4] \otimes [2, 3] \right) = s^t |0\rangle ,
\]

(5.26)

\[
|t_+\rangle = -\frac{1}{\sqrt{2}} |\uparrow_1 \uparrow_3 \rangle \otimes |2, 4\rangle + \frac{1}{\sqrt{2}} |1, 3\rangle \otimes |\uparrow_2 \uparrow_4 \rangle = t^t_+ |0\rangle ,
\]

(5.27)

\[
|t_-\rangle = -\frac{1}{\sqrt{2}} |\downarrow_1 \downarrow_3 \rangle \otimes |2, 4\rangle + \frac{1}{\sqrt{2}} |1, 3\rangle \otimes |\downarrow_2 \downarrow_4 \rangle = t^t_- |0\rangle .
\]

(5.28)

\[
|t_{\pm}\rangle = -\frac{1}{\sqrt{2}} |\uparrow_1 \uparrow_3 \rangle \otimes |\downarrow_2 \downarrow_4 \rangle + \frac{1}{\sqrt{2}} |1, 3\rangle \otimes |\uparrow_2 \uparrow_4 \rangle = t^t_{\pm} |0\rangle .
\]

(5.29)

The matrix elements of the spin-operators, \( \hat{S}_x, \hat{S}_y, \) and \( \hat{S}_z \), in the subspace \(|s\rangle, |t_+\rangle, |t_-\rangle\) give the following definition of the spin-operators [46, 98]

\[
\hat{S}^z_m = -\frac{(-)^m}{\sqrt{6}} (t^t_1 s + s^t t^-_2) + \frac{1}{4} \left( t^t_+ t_+ - t^t_- t_- \right) , \quad (5.30a)
\]

\[
\hat{S}^+_m = -\frac{(-)^m}{\sqrt{3}} (t^t_1 s + s^t t_-) + \frac{1}{2\sqrt{2}} \left( t^t_+ t_+ - t^t_- t_- \right) , \quad (5.30b)
\]

\[
\hat{S}^-_m = -\frac{(-)^m}{\sqrt{3}} (t^t_1 s + s^t t_+) + \frac{1}{2\sqrt{2}} \left( t^t_+ t_+ - t^t_- t_- \right) , \quad (5.30c)
\]

where the vertices of the plaquette are labelled by \( m \). To fix the boson numbers on each plaquette, the constraint \( s^t_1 s + t^t_1 t_+ + t^t_2 t_+ + t^t_- t_- = 1 \) is applied therein. Let introduce the operators \( t_x \) and \( t_y \) as following

\[
t_x = \frac{-1}{\sqrt{2}} (t_+ + t_-) ,
\]

(5.31)

\[
t_y = \frac{1}{\sqrt{2} t} (t_+ - t_-) .
\]

(5.32)
Table 5.1: The eigen spectrum of the plaquette Hamiltonian $\mathcal{H}_p$. In the table, the singlet and triplet ($S_z = 0$) state are denoted by $[i, j]$ and $\{i, j\}$ which are defined as $[i, j] = \frac{1}{\sqrt{2}} (| \uparrow_i \downarrow_j \rangle + | \downarrow_i \uparrow_j \rangle)$, $\{i, j\} = \frac{1}{\sqrt{2}} (| \uparrow_i \downarrow_j \rangle - | \downarrow_i \uparrow_j \rangle)$.

<table>
<thead>
<tr>
<th>$S_{13}$</th>
<th>$S_{24}$</th>
<th>$S$</th>
<th>Eigen energy</th>
<th>Eigen state(s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 0 0</td>
<td>0</td>
<td>$-\frac{3}{2} J_2$</td>
<td>$[1, 2] \otimes [3, 4] - [2, 3] \otimes [4, 1]$</td>
<td></td>
</tr>
<tr>
<td>0 1 1</td>
<td>$-\frac{1}{2} J_2$</td>
<td>${[1, 3] \otimes \downarrow_2 \downarrow_4, [1, 3] \otimes \frac{1}{\sqrt{2}} (</td>
<td>\uparrow_2 \downarrow_4 \rangle +</td>
<td>\downarrow_2 \uparrow_4 \rangle), [1, 3] \otimes</td>
</tr>
<tr>
<td>1 0 1</td>
<td>$-\frac{1}{2} J_2$</td>
<td>${[\downarrow_1 \downarrow_3] \otimes [2, 4], \frac{1}{\sqrt{2}} (</td>
<td>\uparrow_1 \downarrow_3 \rangle +</td>
<td>\downarrow_1 \uparrow_3 \rangle) \otimes [2, 4],</td>
</tr>
<tr>
<td>1 1</td>
<td>$-J_1 + \frac{1}{2} J_2$</td>
<td>three triplet states$^a$</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>$J_1 + \frac{1}{2} J_2$</td>
<td>five quintet states$^b$</td>
<td></td>
</tr>
</tbody>
</table>

$^a$ $-\frac{1}{\sqrt{3}} | \downarrow_1 \downarrow_3 \rangle \otimes \{2, 4\} + \frac{1}{\sqrt{2}} \{1, 3\} \otimes | \downarrow_2 \downarrow_4 \rangle; -\frac{1}{\sqrt{2}} | \uparrow_1 \uparrow_3 \rangle \otimes | \downarrow_2 \downarrow_4 \rangle + \frac{1}{\sqrt{2}} | \downarrow_1 \downarrow_3 \rangle \otimes | \uparrow_2 \uparrow_4 \rangle;$
$-\frac{1}{\sqrt{2}} | \uparrow_1 \uparrow_3 \rangle \otimes \{2, 4\} + \frac{1}{\sqrt{2}} \{1, 3\} \otimes | \uparrow_2 \uparrow_4 \rangle.$

$^b$ $| \downarrow_1 \downarrow_3 \rangle \otimes | \downarrow_2 \downarrow_4 \rangle; \frac{1}{\sqrt{3}} | \downarrow_1 \downarrow_3 \rangle \otimes \{2, 4\} + \frac{1}{\sqrt{2}} \{1, 3\} \otimes \downarrow_2 \downarrow_4 \rangle; \sqrt{\frac{3}{2}} \{1, 3\} \otimes \{2, 4\} + \frac{1}{\sqrt{6}} | \downarrow_1 \downarrow_4 \rangle \otimes | \uparrow_2 \uparrow_4 \rangle + \frac{1}{\sqrt{6}} | \uparrow_1 \downarrow_3 \rangle \otimes \downarrow_2 \downarrow_4 \rangle; \frac{1}{\sqrt{2}} | \uparrow_1 \downarrow_3 \rangle \otimes \{2, 4\} + \frac{1}{\sqrt{2}} \{1, 3\} \otimes | \uparrow_2 \uparrow_4 \rangle; | \uparrow_1 \uparrow_3 \rangle \otimes | \uparrow_2 \uparrow_4 \rangle.$
5.2. Triplon mean-field theory

Now, the constraint equation becomes

\[ s^t s + t_x^t t_x + t_y^t t_y + t_z^t t_z = 1, \]

and the spin-operators shown in Eq. (5.30) can be written as

\[
\hat{S}_m^\alpha = -\frac{(-)^m}{\sqrt{6}} (t^\dagger s + s^\dagger t^\alpha) - \frac{i}{4} \epsilon_\alpha\beta\gamma t^\beta_\beta t^\gamma,
\]

(5.33)

where \( \alpha, \beta, \gamma = x, y, z \) and \( \epsilon_\alpha\beta\gamma \) is Levi-Civita antisymmetric tensor.

In the simplified triplon mean-field theory, we ignore the mixing of different triplet bosons and higher order triplet interactions. Then, the interaction within plaquette and inter-plaquette exchange become as following

\[
H_p(R) = \left( -2J_1 + \frac{1}{2} J_2 \right) s^t(R)s(R) + \left( -J_1 + \frac{1}{2} J_2 \right) \sum_\alpha t^\dagger_\alpha(R) t_\alpha(R),
\]

(5.34)

\[
\sum_\alpha S_m^\alpha(R) S_m^\alpha(R') \approx \frac{(-)^{m+n}}{6} \sum_\alpha [t_\alpha(R)t^\dagger_\alpha(R') + t_\alpha(R')t^\dagger_\alpha(R) + t_\alpha(R)t_\alpha(R') + t^\dagger_\alpha(R)t^\dagger_\alpha(R')],
\]

(5.35)

where \( R \) and \( R' \) are the position vectors of different plaquettes.

Let the singlet Bose condensation amplitude on the plaquettes (see the shaded plaquettes in Fig. 5.6) is some c-number \( \bar{s} \), i.e. \( \langle s^t \rangle = \langle s \rangle = \bar{s} \). The global constraint is imposed on each plaquette: \( \mu \sum_R [s^2 + \sum_\alpha t^\dagger_\alpha(R)t_\alpha(R) - 1] \). The mean-field Hamiltonian now becomes

\[
H_{mf} = E_0 + \sum_{k, \alpha} [\Lambda_k \left( t_{ka} t_\dagger_{ka} + t^\dagger_{ka} t_{ka} \right) + \Delta_k \left( t_{ka} t_{-ka} + t^\dagger_{ka} t^\dagger_{-ka} \right)],
\]

(5.36)

where

\[
\begin{align*}
E_0 &= -\frac{1}{4} J_1 + \frac{1}{8} J_2 + \frac{1}{2} K - \frac{5}{4} \Lambda + \frac{1}{2} \lambda \bar{s}^2 - \bar{s}^2 \left( \frac{1}{4} J_1 + \frac{1}{12} K - \frac{1}{36} K \bar{s}^2 \right), \\
\Lambda_k &= \lambda + \frac{1}{9} K \bar{s}^2 + \Delta_k, \\
\Delta_k &= \frac{1}{3} \bar{s}^2 \left[ \left( -J_1 + J_2 + 2J_3 + \frac{4}{9} K \bar{s}^2 \right) (\cos 2k_x + \cos 2k_y) + J_2 \cos 2k_x \cos 2k_y \right].
\end{align*}
\]

(5.37)

(5.38)

(5.39)

In the above equations, we have used the definition of renormalized effective potential, that is, \( \lambda = \frac{1}{2} \left( -J_1 + \frac{1}{2} J_2 - \mu \right) \). We diagonalize the mean-field Hamiltonian Eq. (5.36) by Bogoliubov transformation: \( t_{ka} = \gamma_{ka} \cosh \theta_k + \gamma^\dagger_{-ka} \sinh \theta_k \). Thus, we get the following diagonalized form of the mean-field Hamiltonian

\[
H_{mf} = E_0 + 2 \sum_{k, \alpha} E_k \left( \gamma^\dagger_{ka} \gamma_{ka} + \frac{1}{2} \right),
\]

(5.40)
where $E_k = \sqrt{\lambda_k^2 - \Delta_k^2} \geq 0$ is the triplon dispersion. The minimization of the ground state energy with respect to unknown mean-field parameters $\lambda$ and $\Delta^2$ leads to the self-consistent equations. The methodology of finding solution to these equations similar to as discussed in Ref. [33, 34].

5.3 Results and discussion

In this section, we discuss the results obtained by triplon mean-field theory. We considered three different cases of the model, that is, (a) $J_3 = 0$, (b) $J_1 = 0$, and (c) $J_2 = 0$. In these cases, the parametrization of the exchange couplings is as follows

Case (a): $J_3 = 0$

$$J_1 = (1 - \zeta)J, \quad J_2 = \zeta J, \quad K = \kappa, \quad J + \kappa = 1,$$

(5.41)

Case (b): $J_1 = 0$

$$J_2 = (1 - \eta)J, \quad J_3 = \eta J, \quad K = \kappa, \quad J + \kappa = 1,$$

(5.42)

Case (c): $J_2 = 0$

$$J_1 = (1 - \delta)J, \quad J_3 = \delta J, \quad K = \kappa, \quad J + \kappa = 1.$$

(5.43)

where all introduced parameters are positive as the couplings $J_1$, $J_2$, $J_3$, and $K$ are antiferromagnetic. In the following discussion, we focus on three three coupling spaces.

Case (a): In this coupling space, we solve triplon mean-field equations as follows. First, we solve the self-consistent equations obtained by dimer triplon mean-field theory on SS dimer state. It gives the phase boundary between collinear phase and SS dimer state. Importantly, the SS dimer state does not compete with Néel state as the SS dimer state is also an eigenstate for the nearest-neighbor Heisenberg antiferromagnetic interaction term, $J_1 \sum_{(i,j)} S_i \cdot S_j$. Thus, we get an overestimated phase boundary of the collinear state. We can evade from this overestimation by formulating the triplon mean-field theory on the columnar dimer state. This is legitimate as for $J_1 - J_2$ model the triplon mean-field results show columnar singlet dimerization in the range $0.19 \lesssim J_2/J_1 \lesssim 0.67$ [44, 33]. The triplon mean-field analyses on the CD-VBC state show two commensurate magnetically ordered phases: Néel and collinear, and an incommensurate phase with ordering wave vector $(0, \varphi)$, where $\varphi = \cos^{-1} \left( \frac{J_2}{J_1} \right)$. The triplon mean-field theory on the plaquette state gives an energetically more favorable plaquette phase inside the CC-VBC phase. The resultant quantum phase diagram is shown in Fig. 5.7.
Figure 5.7: The quantum phase diagram for the $J_1$-$J_2$-$K$ model ($J_3 = 0$). The solid black lines denote continuous phase transitions whereas broken lines and small circles indicate the first-order transitions. In the phase diagram, we have three valence bond crystal phases: the columnar (C-VBC), the Shastry-Sutherland (SS-VBC), and the plaquette (P-VBC) phases. The ordering wave vectors of the Neel and collinear phases are respectively $(0, \pi)$ and $(0, 0)$. The incommensurate ordered phase has ordering wave vector $(0, \varphi)$, where $\varphi = \cos^{-1} \left( \frac{J_2}{2J_3} \right)$. The wave vectors are with respect to C-VBC unit cell.

The phase diagram has many antiferromagnetic and paramagnetic phases. The phase transitions between them are quite interesting. The Neel phase shows continuous phase transition with CD-VBC phase. The collinear phase also has continuous quantum phase transitions with CD-VBC and SS-VBC phases. The incommensurate magnetic phase shows first order phase transition (i.e., level crossing) with SS-VBC phase and continuous phase transition with CD-VBC phase. The paramagnetic phases, CD-VBC and SS-VBC, show first order quantum phase transition with each other. The exact diagonalization study shows the boundary point of the collinear phase is $\kappa = 0.87$ at $J_2 = 1$ axis whereas our study underestimate and giving 0.52. At $J_1 = 1$ axis, the Neel phase is bounded by $\kappa \lesssim 0.2$ (exact diagonalization), 0.08 (triplon mean-field theory). Again, triplon mean-field theory underestimating. However, the SS-VBC phase exists for $\kappa \gtrsim 0.58$ at $J_1 = 1$ axis in triplon mean-field theory which is very close to numerical value, 0.6, obtained by exact diagonalization on 32 site cluster. Gellé et al. [32] proposed many kind of phases between the Neel phase and the SS dimer phase. This is happening because of the presence of different competing
5.3. Results and discussion

dimer correlations as revealed by triplon mean-field analyses.

Whenever we are close to an magnetically ordered phase, the triplon mean-field results vary from exact numerical results. For example, in the case of $J_1 - J_2$ model, the exact diagonalization predicts the paramagnetic region in the range $0.4 \lesssim J_2/J_1 \lesssim 0.6$ [40, 43] whereas the triplon mean-field gives $0.19 \lesssim J_2/J_1 \lesssim 0.67$ [44, 33]. This is because, in triplon mean-field theory, we always assume singlet condensation and triplons are created on the top of singlets. This means there is always phase coexistence in ordered phase, that is, $\langle \tilde{s} \rangle \neq 0$ and $\langle t_\alpha \rangle \neq 0$. We also explored plaquette valence-bond crystal phase in the quantum phase diagram as earlier studies show the existence of plaquette phase in the paramagnetic region of the $J_1 - J_2$ model on square lattice [46, 52]. The plaquette phase is energetically lower than columnar phase (see shaded region in Fig. 5.7) and hence a level crossing happens between them.

![Diagram](image)

Figure 5.8: The quantum phase diagram for the model when $J_1 = 0$. In this phase diagram, we have three magnetically ordered phases with ordering wave vectors $(0, \pi)_c, (0, 0)_c$, and $(\pi/2, \pi/2)_s$ where the subscripts $c$ and $s$ denote the sublattice-CDS and SS states respectively.

Case (b) & (c): In the absence of $J_1$ interaction, the model consists only second- and third-nearest neighbor interactions. This leads to competition between a antiferromagnetic phase and SS dimer phase. Moreover, there is also an energetically lower columnar dimer on same sublattices state for $J \gg K$. The resultant quantum phase diagram in this case is shown in Fig. 5.8. Here, we formulated the triplon mean-field theory on the SS dimer and sublattice columnar dimer states. There are three magnetically ordered phases with ordering wave vectors $(0, \pi)_c, (0, 0)_c,$
5.3. Results and discussion

Figure 5.9: The quantum phase diagram for the model when \( J_2 = 0 \). The solid black lines denote the continuous transitions and broken line represents first-order quantum phase transition. The \((0, \pi)\) phase is Néel phase. The incommensurate phase has ordering wave vector \((\pi/2, \pi - \varphi)\), where \( \varphi = \cos^{-1}\left(\frac{J_1}{4J_3}\right) \). All the wave-vectors defined with respect to the columnar dimer unit cell.

and \((\pi/2, \pi/2)\), where the subscripts \( c \) and \( s \) denote the referenced columnar dimer on same sub-lattices and SS states respectively. The SS dimer state gives continuous quantum phase transitions with \((\pi/2, \pi/2)\) phase and level crossing with columnar dimer. The columnar dimer shows the continuous transitions with \((0, \pi)_c\) and \((0, 0)_c\) phases. The phase \((0, 0)_c\) shows level crossing with \((\pi/2, \pi/2)_c\). The quantum phase diagram for the remaining case is shown in Fig. 5.9. We carried out the triplon mean-field analyses on the columnar dimer and SS dimer states. The phase diagram contains Néel phase, an incommensurate phase with respective ordering wave vectors \((0, \pi)\) and \((\pi/2, \pi - \varphi)\) on the columnar dimer unit cell, where \( \varphi = \cos^{-1}\left(\frac{J_1}{4J_3}\right) \). The SS dimer state shows energy level crossings with the incommensurate phase and columnar dimer phase. The columnar dimer phase shows continuous quantum phase transitions with Néel and incommensurate phase. In Ref. [52], the authors considered \( J_1, J_2, \) and \( J_3 \) Heisenberg exchange interactions on the square lattice. The numerical results also support many of the phases obtained in triplon mean-field theory.
5.4 Summary

We study the model proposed in Ref. [32] by extending it with an additional third neighbor Heisenberg exchange interaction. This additional interaction is a natural competitor with the SS dimer generating multiple spin-exchange interaction. We divide our study in three parts: (a) $J_3 = 0$, (b) $J_1 = 0$, and (c) $J_2 = 0$. In the analyses, we used dimer triplon mean-field theory [44] and plaquette triplon mean-field theory [46]. The mean-field analyses suggest many antiferromagnetic and gapped phases.