Dirac and Weyl nodes in the plaquette excitations of the frustrated Heisenberg antiferromagnet on the honeycomb lattice

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Abstract

The plaquette valence bond solid (VBS) phase of the $J_1-J_2$ antiferromagnetic (AFM) Heisenberg honeycomb model has been studied using plaquette operator theory. Considering six $S = \frac{1}{2}$ spins on a single plaquette the plaquette operator representation for the spin operators in terms of plaquette-singlet and plaquette-triplet boson operators has been developed. The formalism has been applied to the $J_1-J_2$ honeycomb antiferromagnet to derive an effective interacting boson model in terms of those singlets and triplets. The effective model has been analyzed within the harmonic approximation and results have been compared with the exact diagonalization data. The evolution of Dirac and Weyl nodes with respect to $J_2/J_1$ for this model has been studied.

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I. INTRODUCTION

Recently several different theoretical approaches have been applied to study $J_1 - J_2$ model on the honeycomb lattice. Na$_2$IrO$_3$ [1], Na$_2$Co$_2$TeO$_6$[2] are examples of AFM honeycomb lattice materials. The quantum Monte Carlo results [3] for the half filled honeycomb Hubbard model has shown the existence of gapped spin-liquid phase. DMRG calculations on the $J_1 - J_2$ honeycomb model has been executed [4–6]. The coupled cluster method has been applied on this model [7]. The existence of a Néel phase, a plaquette, and a dimerized VBS phases are observed. The Heisenberg model on the honeycomb lattice has been studied using series expansions [8] and the stability of Néel, columnar, and various spiral phases has been shown. Exact diagonalization studies on the $J_1 - J_2$ Heisenberg model have shown the presence of a resonance valence bond (RVB) state providing the evidence for short-range spin-gapped phases[14]. Using a Holstein-Primakoff expansion the presence and evolution of Weyl and Dirac nodes in the FM and AfM honeycomb lattice has been examined[13].

Based on the SU(2) and SU(3) symmetry representations of the Schwinger boson approach, the complete phase diagrams of the $J_1 - J_2 - J_3$ Heisenberg honeycomb lattice has been obtained[9].

Sachdev and Bhat has been introduced the bond-operator theory to describe VBS phases of a Heisenberg model [10]. The formalism is then applied to describe dimerized phases, such as the columnar and staggered VBSs. The partial bond operator representation for the square plaquette has been introduced by Zhitomirsky and Ueda [11]. Later generalized bond-operator formalism for a four spin plaquette has been developed by Doretto [12].

In this article, the plaquette operator representation considering six $S = \frac{1}{2}$ spins on a single plaquette has been developed. The formalism is applied to the $J_1 - J_2$ AFM Heisenberg honeycomb model. The ground state energy and spin gap have been obtained. In this study, the presence of magnetic Dirac and Weyl bosons in the AFM honeycomb model has been shown by examining the presence of mode crossovers in the triplet dispersion. The evolution of the magnetic Dirac and Weyl points with the exchange interactions has been observed. The Hamiltonian of a single plaquette is introduced in Section II. In Section III plaquette operator theory is developed. In Section IV the formalism is applied to obtain a effective boson model. The model is analyzed within mean-field approximation in Section V. Section VI shows the evolution of Dirac and Weyl nodes with exchange interactions. Section VII
holds a comprehensive discussion on the results.

II. SINGLE HEXAGONAL PLAQUETTE

Spin-$\frac{1}{2}$ AFM Heisenberg Hamiltonian on the hexagonal plaquette is defined by

$$H_{\text{plaq}} = J_1 \sum_{i=1}^6 S_i \cdot S_{i+1} + J_2 \sum_{i=1}^6 S_i \cdot S_{i+2}, S_i^6 = S_i$$

($S_i^6$ is the spin-$\frac{1}{2}$ operator at the position $i$. $J_1$ and $J_2$ are the respective nearest neighbor (NN) and next nearest neighbor (NNN) exchange interaction strengths. The action of $J_2$ is totally opposite than that of $J_1$, in a sense that $J_2$ invokes frustration in this model, while $J_1$ is non-frustrating. Schematic representation of this spin model is shown in Fig.1. The z-component of the total spin, $S_T^z$ is a good quantum number. To obtain analytic expressions of eigenvalues and eigenfunctions the Hamiltonian (Eq.1) has been spanned in the different subspaces of $S_T^z$. The Hilbert space of six spins consists of $2^6$ states those states comprise to five singlets ($S_T = 0$), nine triplets ($S_T = 1$), five quintets ($S_T = 2$) and one septet ($S_T = 3$). The exact analytic expressions of all singlet($|s\rangle$), triplet($|t\rangle$), quintet($|q\rangle$) and septet($|h\rangle$) states with energy eigenvalues have been listed in the Appendix A. Only two singlets, $|s_1\rangle$ and $|s_2\rangle$ can be expressed as resonating valence bond (RVB) states. Those two particular singlets are defined by $\Psi_{\text{RVB}}$ and $\Psi'_{\text{RVB}}$. The representation of RVB states $\Psi_{\text{RVB}}(|s_1\rangle)$ and $\Psi'_{\text{RVB}}(|s_2\rangle)$ are shown in the Fig.2. Ground state is always a total spin singlet. For $J_2 < \frac{J_1}{2}$ the ground state is given by $\Psi_{\text{RVB}}$ and $\Psi'_{\text{RVB}}$ becomes the ground state for $J_2 > \frac{J_1}{2}$. Ground
state is doubly degenerate over the line, \( J_1 = 2J_2 \). \( \Psi_{RVB} \) is antisymmetric, whereas, \( \Psi'_{RVB} \) is symmetric under the rotation by \( \frac{\pi}{3} \).

\[
\Psi_{RVB} \equiv -\sqrt{\frac{2}{3}} \frac{1}{\mu_1} \begin{pmatrix}
\begin{array}{c}
\vdots \\
\end{array}
\end{pmatrix} + \frac{2}{\sqrt{3}} \frac{1}{\mu_1} C_{s_1} \begin{pmatrix}
\begin{array}{c}
\vdots \\
\end{array}
\end{pmatrix}
\]

\[
\Psi'_{RVB} \equiv \sqrt{\frac{2}{3}} \begin{pmatrix}
\begin{array}{c}
\vdots \\
\end{array}
\end{pmatrix} - \frac{1}{\sqrt{2}} \begin{pmatrix}
\begin{array}{c}
\vdots \\
\end{array}
\end{pmatrix}
\]

 FIG. 2: Pictorial representation of \( \Psi_{RVB} \) and \( \Psi'_{RVB} \).

Variation of energy eigenvalues of single plaquette against \( J_2/J_1 \) are shown in Fig.3(a). Variations of two lowest singlet energy and three lowest triplet excitations with respect to exchange interaction strengths are shown in Fig.3(b). \( E_{s_1} \) is the energy of the singlet state \( |s_1\rangle \) whereas \( E_{s_2} \) is the energy of the singlet state \( |s_2\rangle \). \( E_{t_1} \) is the energy of the three triplet states \( |t_1,\alpha\rangle \) and \( E_{t_2} = E_{t_3} \) is the energy of the six triplet states \( |t_2,\alpha\rangle \) and \( |t_3,\alpha\rangle \) where \( \alpha = x, y, z \). It has been noted the excitation gap is associated with a singlet-triplet transition (triplet gap) for \( J_2 < \frac{J_1}{2} \) and with a singlet-singlet one (singlet gap) for \( J_2 > \frac{J_1}{2} \).
III. PLAQUETTE OPERATOR THEORY

In this section, considering the case of six spins $S = \frac{1}{2}$ in a plaquette a bond-operator representation for the spin operators in terms of singlet and triplet (boson) operators has been developed. The Hilbert space of six spins in a single plaquette is consist of 64 states. A set of bosonic creation operators corresponding to these states have been introduced.

$$|s_i\rangle = s_i^\dagger |0\rangle, |t_{a,\alpha}\rangle = t_{a,\alpha}^\dagger |0\rangle, |q_{b,\nu}\rangle = q_{b,\nu}^\dagger |0\rangle,$$

$$|q_{b,\alpha}\rangle = q_{b,\alpha}^\dagger |0\rangle, |h_\zeta\rangle = h_\zeta^\dagger |0\rangle, |h_\alpha\rangle = h_\alpha^\dagger |0\rangle$$  \hspace{1cm} (2)

where $|0\rangle$ denotes the vacuum state, $i = 1^\pm, 2, 3, 4$, $a = 1^\pm, 2^\pm, 3^\pm, 4, 5, 6$, $\alpha = x, y, z$, $b = 1, 2, 3, 4, 5$, $\nu = 1^\pm$ and $\zeta = 1^\pm, 2^\pm$. The physical constraint considering the completeness theorem

$$\sum_i s_i^\dagger s_i + \sum_{a,\alpha} t_{a,\alpha}^\dagger t_{a,\alpha} + \sum_{b,\nu} q_{b,\nu}^\dagger q_{b,\nu} + \sum_{b,\alpha} q_{b,\alpha}^\dagger q_{b,\alpha} + \sum_\zeta h_\zeta^\dagger h_\zeta + \sum_\alpha h_\alpha^\dagger h_\alpha = 1$$  \hspace{1cm} (3)

The matrix elements of each component of the six spins operators within the basis $(|s_1\rangle, |s_2\rangle, |s_3\rangle, |s_4\rangle, |t_{a,\alpha}\rangle)$ have been calculated. Considering the matrix elements $\langle s_1\pm | S_{a}^m | t_{a,\alpha}\rangle, \langle s_2 | S_{a}^m | t_{a,\alpha}\rangle, \cdots$, the three components of the six spin operators $S_{a}^m$ has been written in terms of boson operators $s$ and $t$ as $S_{a}^m$ in appendix B.

The Hamiltonian. Eq.1 assumes the form

$$H_{\text{plaq}} = \sum_{i=1^\pm, 2, 3, 4} E_{s_i} s_i^\dagger s_i + \sum_{i=1^\pm, 2^\pm, 3^\pm, 4, 5, 6} E_{t_{a,\alpha}} t_{a,\alpha}^\dagger t_{a,\alpha} + \sum_{b=1}^5 E_{q_{b,\nu}} \left( q_{b,\nu}^\dagger q_{b,\nu} + q_{b,\alpha}^\dagger q_{b,\alpha} \right) + E_{h_{\zeta}} \left( h_\zeta^\dagger h_\zeta + h_\alpha^\dagger h_\alpha \right)$$  \hspace{1cm} (4)

Considering only two singlets $(|s_1\rangle, |s_2\rangle)$ and three lowest triplets $(|t_{1-\alpha}\rangle, |t_{2-\alpha}\rangle, |t_{3-\alpha}\rangle)$ the approximate expansion for the spin operators $S_{a}^m$ becomes

$$S_{a}^m = A_{a}^n \left( t_{a-\alpha}^\dagger s_1 - s_1^\dagger t_{a-\alpha} \right) + B_{a}^n \left( t_{a-\alpha}^\dagger s_2 + s_2^\dagger t_{a-\alpha} \right) + i\epsilon^{\alpha\beta\gamma} D_{a,b}^n t_{a-\beta}^\dagger t_{b-\gamma}$$  \hspace{1cm} (5)

where $n = 1, 2, 3, 4, 5, 6$, $a = 1, 2, 3$ and $b = 1, 2$. The coefficients $A_{a}^n$, $B_{a}^n$ and $D_{a,b}^n$ are given in the appendix B.

IV. EFFECTIVE BOSON MODEL

In this section, plaquette operator theory (POT) has been employed to study the plaquette VBS phase of the $J_1$-$J_2$ AFM Heisenberg honeycomb model. The Hamiltonian (1) has
been written in terms of plaquettes of honeycomb lattice as shown in Fig. 4(b).

\[ H = \sum_{\tau_i} J_1 \left( S_{\tau_i}^1 \cdot S_{\tau_i}^2 + S_{\tau_i}^2 \cdot S_{\tau_i}^3 + S_{\tau_i}^3 \cdot S_{\tau_i}^4 + S_{\tau_i}^4 \cdot S_{\tau_i}^5 + S_{\tau_i}^5 \cdot S_{\tau_i}^6 + S_{\tau_i}^6 \cdot S_{\tau_i}^1 \right) 
+ J_2 \left( S_{\tau_i}^1 \cdot S_{\tau_i}^2 + S_{\tau_i}^2 \cdot S_{\tau_i}^3 + S_{\tau_i}^3 \cdot S_{\tau_i}^5 + S_{\tau_i}^4 \cdot S_{\tau_i}^6 + S_{\tau_i}^5 \cdot S_{\tau_i}^6 + S_{\tau_i}^1 \cdot S_{\tau_i}^2 \right) 
+ J_1 \left( S_{\tau_i}^1 \cdot S_{\tau_i+\tau_1+\tau_2}^5 + S_{\tau_i}^2 \cdot S_{\tau_i+\tau_1+\tau_2}^6 + S_{\tau_i}^3 \cdot S_{\tau_i+\tau_1+\tau_2}^3 + S_{\tau_i}^4 \cdot S_{\tau_i+\tau_1+\tau_2}^1 + S_{\tau_i}^5 \cdot S_{\tau_i+\tau_1+\tau_2}^2 + S_{\tau_i}^6 \cdot S_{\tau_i+\tau_1+\tau_2}^4 \right) \]  

Here, \( \tau_i \) is the plaquette index and the numbers \( \tau_1 \) and \( \tau_2 \) in the site indices represent the primitive vectors

\[ \tau_1 = 3a \hat{y} \quad \text{and} \quad \tau_2 = \frac{3\sqrt{3}a}{2} \hat{x} - \frac{3a}{2} \hat{y} \]

\( a \) is the lattice spacing of the original honeycomb lattice which has been set to 1. The approximate bond-operator representation (Eq. 5) has been substituted in (Eq. 6) to write the Hamiltonian in the following form

\[ H = E_0 + H_{02} + H_{20} + H_{30} + H_{21} + H_{31} + H_{40} + H_{42} \]  

(8)

\( n \) and \( m \) indicates that number of triplet and singlet operators in the expressions of \( H_{nm} \). \( E_0 \) ia a constant.

\[ E_0 = N' \left[ N_0 E_{s_{-}} - \mu (N_0 - 1) \right] \]  

(9)
with \( N' = \frac{N}{6} \) where \( N \) is the number of sites of the original honeycomb lattice. The constraint (Eq. 3) has been taken into account by adding the following term to the Hamiltonian (Eq. 8)

\[
-\mu \sum_i \left( s_{1,i}^\dagger s_{1,i} + s_{2,i}^\dagger s_{2,i} + t_{a,i,a}^\dagger t_{a,i,a} - 1 \right)
\]

(10)

where \( \mu \) is the Lagrange multiplier.

In the region \( R_1 (J_2 < \frac{J_1}{2}) \), \( s_{1,i} \) is the lowest-energy singlet. The singlet operators \( |s_{1,i}^\dagger\rangle \) is assumed to be condensed and has been replaced with a number in (Eq. 8).

\[
s_{1,i}^\dagger = s_{1,i} = \langle s_{1,i}^\dagger \rangle = \sqrt{N_0} \]

In the region \( R_2 (J_2 > \frac{J_1}{2}) \) the plaquette VBS state has been considered as a condensate of the lowest-energy singlets \( s_{2,i} \). The singlet operators \( |s_{2,i}\rangle \) has been replaced with a number in (Eq. 8).

\[
s_{2,i}^\dagger = s_{2,i} = \langle s_{2,i}^\dagger \rangle = \sqrt{N_0}
\]

Fourier transformation of the operators \( t_{a,i,a}^\dagger \) and \( s_{b,i}^\dagger \)

\[
t_{a,i,a}^\dagger = \frac{1}{\sqrt{N'}} \sum_k \exp (-i \mathbf{k} \cdot \mathbf{R}_i) t_{a,k,a}^\dagger
\]

\[
s_{b,i}^\dagger = \frac{1}{\sqrt{N'}} \sum_k \exp (-i \mathbf{k} \cdot \mathbf{R}_i) s_{b,k}^\dagger
\]

(11)

Here the momentum sums run over the 1st Brillouin zone.

V. MEAN-FIELD APPROXIMATION

Considering the quadratic approximation \( H_{30}, H_{21}, H_{31}, H_{40}, H_{22} \) have been neglected and

\[
H \approx E_0 + H_{02} + H_{20}
\]

(12)

The expressions of \( H_{02} \) and \( H_{20} \) in terms of singlet and triplet operators in momentum space becomes

\[
H_{02} = \sum_k (E_{s_m} - \mu) s_{m,k}^\dagger s_{m,k}
\]

with \( m = 2 \) for \( J_2 < \frac{J_1}{2} \) and \( m = 1 \) for \( J_2 > \frac{J_1}{2} \).

\[
H_{20} = \sum_k X_{k}^{ab} t_{a-,k,a}^\dagger t_{b-,k,a}^\dagger + \frac{Y_{k}^{ab}}{2} \left( t_{a-,k,a}^\dagger t_{b-,k,a}^\dagger t_{a-,k,a} t_{b-,k,a} + t_{a-,k,a} t_{b-,k,a}^\dagger t_{a-,k,a}^\dagger \right)
\]

(14)

where \( a, b = 1, 2, 3 \) and \( \alpha, \beta, \gamma = x, y, z \). The coefficients \( X_{k}^{ab} \) and \( Y_{k}^{ab} \) are given in Appendix C.
Singlet sector $H_{02}$ has been diagonalized separately and singlet excitation energy $\Omega_s = (E_{sm} - \mu)$ has been determined. The six-component vector $\Psi^\dagger_{k,\alpha} = (t^\dagger_{1,-,k,\alpha} t^\dagger_{2,-,k,\alpha} t^\dagger_{3,-,k,\alpha} t_{1,-,-,k,\alpha} t_{1,-,-,k,\alpha} t_{1,-,-,k,\alpha})$ has been introduced to diagonalize the triplet sector $H_{20}$. Eq. 12 has been written as

$$H = E'_0 + H_{02} + \frac{1}{2} \sum_k \Psi^\dagger_{k,\alpha} \hat{H}_k \Psi_{k,\alpha}$$

where

$$E'_0 = E_0 - \frac{3}{2} \sum_k \sum_{i=1,2,3} X_{k}^{aa}$$

and

$$\hat{H}_k = \begin{pmatrix} \hat{X}_k & \hat{Y}_k \\ \hat{Y}_k & \hat{X}_k \end{pmatrix}$$

$\hat{X}_k$ and $\hat{Y}_k$ are the $3 \times 3$ hermitian matrices having elements like $X_{k}^{ab}$ and $Y_{k}^{ab}$ respectively. After diagonalization the Hamiltonian assumes the form

$$H = E_{EGS} + H_{02} + \frac{1}{2} \sum_k \Phi^\dagger_{k,\alpha} \hat{H}'_k \Phi_{k,\alpha}$$

with

$$E_{EGS} = E_0 + \frac{3}{2} \sum_{a,k} (\Omega_{a,k} - X_{k}^{aa})$$

$E_{EGS}$ is the ground state energy and $\hat{H}'_k = \begin{pmatrix} \hat{h}_k & 0 \\ 0 & \hat{h}_k \end{pmatrix}$ where $\hat{h}_k = \begin{pmatrix} \Omega_{1,k} & 0 & 0 \\ 0 & \Omega_{2,k} & 0 \\ 0 & 0 & \Omega_{3,k} \end{pmatrix}$.

The eigenvectors $\Phi^\dagger_{k,\alpha}$ is given by $\Phi^\dagger_{k,\alpha} = (b^\dagger_{1,-,k,\alpha} b^\dagger_{2,-,k,\alpha} b^\dagger_{3,-,k,\alpha} b_{1,-,-,k,\alpha} b_{1,-,-,k,\alpha} b_{1,-,-,k,\alpha})$. Two sets of boson operators $t$ and $b$ are connected to each other by the following relation

$$\Phi_{k,\alpha} = \hat{M}_k \Psi_{k,\alpha}, \quad \text{where} \quad \hat{M}_k = \begin{pmatrix} U^\dagger_k & -V^\dagger_k \\ -V^\dagger_k & U^\dagger_k \end{pmatrix}$$

$U^\dagger_k$ and $V^\dagger_k$ are the $3 \times 3$ hermitian matrices having elements like Bogoliubov coefficients $u_{k}^{ab}$ and $v_{k}^{ab}$ respectively. The analytic expressions of the triplet excitation energies $\Omega_{a,k}$ and the Bogoliubov coefficients $u_{k}^{ab}$ and $v_{k}^{ab}$ in terms of the $X_{k}^{ab}$ and $Y_{k}^{ab}$ are given in Appendix C.
The following self-consistent equations for \( \mu \) and \( N_0 \) have been obtained from the saddle-point conditions \( \frac{\partial E_{\text{EGS}}}{\partial N_0} = 0 \) and \( \frac{\partial E_{\text{EGS}}}{\partial \mu} = 0 \),

\[
\mu = E_{s_1} + \frac{3}{2N'} \sum_{a,k} \left[ \frac{\partial \Omega_{a,k}}{\partial N_0} \frac{Y_{a}^{a}}{N_0} \right]
\]

\[
N_0 = 1 + \frac{3}{2N'} \sum_{a,k} \left[ \frac{\partial \Omega_{a,k}}{\partial \mu} + 1 \right]
\]

From the numerical results of \( \mu \) and \( N_0 \) the triplet \( \Omega_{a,k} \) and the singlet \( \Omega_s = E_{s_m} - \mu \) excitation energies have been obtained.

Variation of excitation gap with respect to \( J_2/J_1 \) is shown in Fig. 5(a) and 5(b). Singlet excitation (\( \Omega_s \)) is dispersionless in the mean-field approximation. In region \( R_1 \) the minimum (gap) of the triplet dispersion relation occurs at the center of the Brillouin zone (\( \Gamma \) point) for \( J_2/J_1 < 0.23 \) and then momentum associated with the excitation gap changes. For \( 0.23 < J_2/J_1 < 0.47 \) the minimum excitation gap occurs at the M point. In region \( R_2 \) momentum associated with the excitation gap changes at \( J_2/J_1 = 0.617 \). The excitation gap changes from a triplet to a singlet one at \( J_2/J_1 = 0.47 \) in region \( R_1 \) and at \( J_2/J_1 = 0.63 \) in region \( R_2 \).

Variation of ground state energy per site with respect to \( J_2/J_1 \) is shown in Fig.6(a). Result based on the POT is plotted in red line and that has been compared with the exact diagonalization data for 32 sites shown in blue points [14]. The value of \( E_{\text{EGS}}/N J_1 \) shows closer agreement with the exact diagonalization data in the disordered phases which is identified by the region shaded in pink, where this formalism stands valid. Moreover, the ground
FIG. 6: Variation of (a) Ground-state energy per site and (b) spin gap against \( J_2/J_1 \).

state energy is always higher than the true value because of the fact that POT is basically a variational approach. \( E_{\text{EGS}}/N J_1 \) shows significant departure from the exact diagonalization data in the ordered regions since POT fails to capture the quantum correlation in those regions. Similarly, spin gap \( (\Delta/J_1) \) has been evaluated through the POT and that is shown in Fig. 6(b). POT predicts a gap in the disordered phases (shaded in pink) in addition to the magnetic ordered phases. \( \Delta/J_1 \) corresponds to the gap between the ground state energy and the minimum of the triplet excitations.

VI. TRIPLET DISPERSION

The 3D representation of triplet dispersion has been shown in Fig. 7(a)-7(c). The triplet dispersion along the high-symmetry pathway (\( \Gamma, M, K, \Gamma \)) in the region \( R_1 \) of the AFM configuration of the honeycomb lattice has been shown in Fig. 7(d)-7(f). AFM configuration consists of three distinct modes. The number of mode crossing points changes with \( J_2/J_1 \). The 1st and 2nd mode crossing are represented by red circles and the crossing of 2nd and 3rd mode are represented by the blue circles. The magnon triple points are shown by the pink circles. The mode crossing at the point \( \mathbf{k} = (0, 4\pi/9) \) remains independent with exchange interaction strengths whereas the position of other mode crossing point changes. This crossing point is considered as Dirac node and represented by the green circle. The others magnon bands crossing points are considered as Weyl magnon node. Red circles are considered as W1 node while the blue circles are taken as W2 node. In region \( R_1 \) the nodal points located at \( \Gamma \) and \( K \) change from W1 node to W2 node as \( J_2/J_1 \) crosses the value 0.34. At \( J_2/J_1 = 0.34 \) magnon triple points exist at those nodal points. The production of the nodal points in \( \mathbf{k} \)
FIG. 7: Evolution of Dirac and Weyl nodes in region $R_1$ with $J_2/J_1$, $J_2/J_1 = (a),(d),(g) 0.30$, (b),(e),(h) 0.34, (c),(f),(i) 0.45.

space as exchange interaction strengths changes are shown in the Fig.7(g)-7(i). The six Dirac nodal points are located at $(0, \frac{4\pi}{9}), (0, -\frac{4\pi}{9}), (\frac{2\pi}{3\sqrt{3}}, \frac{2\pi}{9}), (-\frac{2\pi}{3\sqrt{3}}, -\frac{2\pi}{9}), (\frac{2\pi}{3\sqrt{3}}, -\frac{2\pi}{9})$ and $(-\frac{2\pi}{3\sqrt{3}}, -\frac{2\pi}{9})$ for any values of $J_2/J_1$. W1 nodes are located at $\Gamma, K$ for $J_2/J_1 < 0.34$ and magnon triple points for $J_2/J_1 = 0.34$. For $J_2/J_1 > 0.34$ W1 nodes are located at 3 points around $K$ and at 6 points around $\Gamma$. W2 nodes are located at 2 points near $K$ and at 6 points around $\Gamma$. 

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The 3D representation of triplet dispersion has been shown in Fig. 7(a)-7(d). The triplet dispersion along the high-symmetry pathway (Γ, M, K, Γ) in the region $R_2$ of the AFM configuration of the honeycomb lattice has been shown in Fig. 8(e)-8(h). Dirac node exists in this region also at $k = (0, \frac{4\pi}{9})$ for any values of $J_2/J_1$. Magnon triple points exist at at Γ and K for $J_2/J_1 = 0.60$. As interaction strengths crosses that critical value the nodal points located at Γ and K are changes from W1 node to W2 node. The lowest excitation remains dispersionless for $J_2/J_1 < 0.60$. The production of the nodal points in $k$ space as exchange interaction strengths changes are shown in the Fig. 8(i)-8(l). The position of six Dirac nodal points are same as in region $R_1$. W1 nodes are located at Γ, K for $J_2/J_1 < 0.60$ and magnon triple points for $J_2/J_1 = 0.60$. For $J_2/J_1 > 0.60$ W1 nodes are located on a semicircle around K and on a circle around Γ. W2 nodes are located around the Dirac nodes for $J_2/J_1 > 0.65$. 
FIG. 8: Evolution of Dirac and Weyl nodes in region $R_2$ with $J_2/J_1$, $J_2/J_1 = (a),(e),(i) 0.55$, $(b),(f),(j) 0.60$, $(c),(g),(k) 0.65$, $(d),(h),(l) 0.75$.
Fig. 9(a) and 9(b) shown the evolution of Dirac and Weyl nodes along $k$ with $J_2/J_1$. In region $R_1$ the no of Weyl nodes changes from 2 to 7 as $J_2/J_1$ crosses the value 0.34. Dirac node remains independent of exchange interaction strengths in both regions which is denoted by the green dots. In region $R_2$ as $J_2/J_1$ crosses the value 0.60 Weyl nodes are produced around the Dirac node.

FIG. 10: Two modes crossing and formation of Dirac node at $k_d = (0, \frac{4\pi}{9})$ in the Brillouin zone.

VII. DISCUSSION

In this paper the plaquette operator representation has been introduced and the plaquette VBS phase of the $J_1 - J_2$ AFM Heisenberg honeycomb model has been studied. It has been found that in the mean-field approximation the plaquette VBS phase of the $J_1 - J_2$ model is stable in the region $0.1 < J_2/J_1 < 1.0$. Mean-field results shows closer agreement
with the exact diagonalization data in the region $0.2 < J_2/J_1 < 0.4$. The Phase diagram of the frustrated honeycomb lattice has been studied by A. F. Albuquerque, D. Schwandt, B. Hetényi, S. Capponi, M. Mambrini, and A. M. Läuchli [15]. The authors found a magnetically disordered phase forming a plaquette valence-bond crystal in the region which is closed to this region. The ground state of spin-$\frac{1}{2}$ $J_1 - J_2$ AFM Heisenberg honeycomb model exhibits two different kinds of long range orders in two extreme locations, Néel order for $0 \leq J_2/J_1 \leq 0.2$ and spiral order for $0.4 \leq J_2/J_1 \leq 0.5$. Intermediate region exhibits two types of disordered phases, gapped spin liquid phase for $0.20 \leq J_2/J_1 \leq 0.37$ and staggered dimer valence bond solid phase for $0.437 \leq J_2/J_1 \leq 0.40$ [16]. The creation and evolution of Dirac and Weyl nodes in the magnetic honeycomb lattice has been studied using linear spin-wave theory by D. Boyko, A. V. Balatsky, and J. T. Haraldsen [13]. The authors found that the AFM configuration does not produce two modes. Therefore, Dirac modes are absent in AFM configuration. In this paper same model has been studied using POT within mean-field approximation. It has been found that AFM configuration consists of three distinct modes and the crossing of these modes produces Dirac and Weyl nodes. Two main phenomena have been observed in the AFM configuration. The Dirac nodes are not shifted in k space with change in exchange interaction strengths while multiple Weyl nodes have been produced and shifted with exchange interaction strengths.

VIII. ACKNOWLEDGMENTS

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Appendix A: ENERGY EIGENVALUES AND EIGENSTATES

In this section, we provide the expressions of all the eigenvectors and corresponding eigenvalues of the single hexagonal plaquette, Eq. 1. All energy eigenvalues are given below.

\[
\begin{align*}
E_{s_1^\pm} &= -J_1 \pm \frac{1}{2} d_s, & E_{s_2} &= \frac{3}{2} (-J_1 - J_2), & E_{s_3} &= E_{s_4} = -\frac{1}{2} (J_1 + 3 J_2), \\
E_{t_1^\pm} &= -J_1 \pm \frac{1}{2} d_t, & E_{t_2^\pm} &= E_{t_3^\pm} = -\frac{1}{4} (J_1 + 3 J_2 \pm d_t), \\
E_{t_4} &= E_{t_5} = -J_1, & E_{t_6} &= \frac{1}{2} (J_1 - 3 J_2), & E_{q_1} &= E_{q_2} = J_1, \\
E_{q_3} &= E_{q_4} = 0, & E_{q_5} &= \frac{1}{2} (-J_1 + 3 J_2), & E_h &= \frac{3}{2} (J_1 + J_2)
\end{align*}
\]

(A1)

To write down all the eigenstates following notations have been used.

\[
d_s = \sqrt{13J_4^2 + 9J_2^2 - 18J_1J_2}, \quad d_t = \sqrt{5J_4^2 + 9J_2^2 - 10J_1J_2}, \quad d_{t_2} = \sqrt{17J_4^2 + 9J_2^2 - 10J_1J_2},
\]

\[
\begin{align*}
|\psi_n^3\rangle &= T^{n-1} |3\rangle \ (n = 1), \ |3\rangle = |\uparrow\uparrow\uparrow\uparrow\uparrow\rangle, \\
|\psi_n^2\rangle &= T^{n-1} |2\rangle \ (n = 1, 2, 3, 4, 5, 6), \ |2\rangle = |\uparrow\uparrow\uparrow\downarrow\rangle, \\
|\psi_n^1_0\rangle &= T^{n-1} |1\rangle_0 \ (n = 1, 2, 3, 4, 5, 6), \ |1\rangle_0 = |\uparrow\uparrow\downarrow\downarrow\rangle, \\
|\psi_n^1_1\rangle &= T^{n-1} |1\rangle_1 \ (n = 1, 2, 3, 4, 5, 6), \ |1\rangle_1 = |\downarrow\uparrow\downarrow\uparrow\rangle, \\
|\psi_n^1_2\rangle &= T^{n-1} |1\rangle_2 \ (n = 1, 2, 3), \ |1\rangle_2 = |\downarrow\uparrow\uparrow\uparrow\rangle, \\
|\psi_n^0_0\rangle &= T^{n-1} |0\rangle_0 \ (n = 1, 2, 3, 4, 5, 6), \ |0\rangle_0 = |\uparrow\uparrow\downarrow\downarrow\downarrow\rangle, \\
|\psi_n^0_1\rangle &= T^{n-1} |0\rangle_1 \ (n = 1, 2, 3, 4, 5, 6), \ |0\rangle_1 = |\uparrow\downarrow\downarrow\downarrow\rangle, \\
|\psi_n^0_2\rangle &= T^{n-1} |0\rangle_2 \ (n = 1, 2, 3, 4, 5, 6), \ |0\rangle_2 = |\downarrow\uparrow\downarrow\downarrow\rangle, \\
|\psi_n^0_3\rangle &= T^{n-1} |0\rangle_3 \ (n = 1, 2), \ |0\rangle_3 = |\downarrow\uparrow\uparrow\downarrow\rangle, \\
|\psi_n^{-1}_0\rangle &= T^{n-1} |-1\rangle_0 \ (n = 1, 2, 3, 4, 5, 6), \ |-1\rangle_0 = |\downarrow\downarrow\uparrow\uparrow\downarrow\rangle, \\
|\psi_n^{-1}_1\rangle &= T^{n-1} |-1\rangle_1 \ (n = 1, 2, 3, 4, 5, 6), \ |-1\rangle_1 = |\uparrow\downarrow\downarrow\uparrow\downarrow\rangle, \\
|\psi_n^{-1}_2\rangle &= T^{n-1} |-1\rangle_2 \ (n = 1, 2, 3), \ |-1\rangle_2 = |\uparrow\downarrow\uparrow\downarrow\rangle, \\
|\psi_n^{-2}_0\rangle &= T^{n-1} |-2\rangle \ (n = 1, 2, 3, 4, 5, 6), \ |-2\rangle = |\downarrow\downarrow\uparrow\downarrow\uparrow\rangle, \\
|\psi_n^{-3}_0\rangle &= T^{n-1} |-3\rangle \ (n = 1), \ |-3\rangle = |\downarrow\downarrow\downarrow\uparrow\downarrow\rangle.
\end{align*}
\]

Here $T$ is a unitary cyclic right shift operator. $T |abcdef\rangle = |fabcde\rangle$ where $|abcdef\rangle = |a\rangle \otimes |b\rangle \otimes |c\rangle \otimes |d\rangle \otimes |e\rangle \otimes |f\rangle$. All the energy eigenstates have been listed below.
\[ |s_1\rangle = \frac{1}{\mu_{s_1} \sqrt{2}} \left( \sum_{n=1,6} (-1)^{n-1} (\sqrt{2}C_{s_1,3}|\psi^0_n\rangle + C_{s_1,2}(|\psi^0_n\rangle + |\psi^0_n\rangle_2) + \sqrt{2}C_{s_1,1} \sum_{n=1}^3 (-1)^{n-1} |\psi^0_n\rangle_3 \right) \]

\[ |s_2\rangle = \frac{1}{\sqrt{2}} \left( \sum_{n=1,6} \left( |\psi^0_n\rangle_2 - |\psi^0_n\rangle_1 \right) \right) \]

\[ |s_3\rangle = \frac{1}{2} \left( \sum_{n=1,6} (-1)^n |\psi^0_n\rangle_0 + \sum_{n=1,4} (-1)^n (|\psi^0_n\rangle_1 + |\psi^0_n\rangle_2) + \frac{1}{6} \sum_{n=1}^6 (-1)^{n-1} (|\psi^0_n\rangle_0 + |\psi^0_n\rangle_1 + |\psi^0_n\rangle_2) \right) \]

\[ |s_4\rangle = \frac{1}{\sqrt{2}} \left( \sum_{n=1}^2 |\psi^0_n\rangle_0 - \sum_{n=1}^5 |\psi^0_n\rangle_0 + \sum_{n=1,2} |\psi^0_n\rangle_1 + |\psi^0_n\rangle_2 - \frac{1}{3} \sum_{n=2}^3 (|\psi^0_n\rangle_1 + |\psi^0_n\rangle_2) \right) \]

\[ |t_{1,+,\alpha}\rangle = \frac{\lambda_\alpha}{\mu_{t_{1,+,\alpha}} \sqrt{2}} \left( \sum_{n=1,1} \left( C_{t_{1,+,\alpha},2} |\psi^0_n\rangle_0 + C_{t_{1,+,\alpha},1} (|\psi^0_n\rangle_1 + |\psi^0_n\rangle_2) + \sqrt{2}C_{t_{1,+,\alpha},0} \sum_{n=1}^3 (|\psi^0_n\rangle_2 + |\psi^0_n\rangle_3) \right) \right) \]

\[ |t_{2,+,\alpha}\rangle = \frac{\lambda_\alpha}{\mu_{t_{2,+,\alpha}} \sqrt{24}} \left( \sum_{n=1,2,5} \left( C_{t_{2,+,\alpha},1} (|\psi^0_n\rangle_0 + |\psi^0_n\rangle_1) + C_{t_{2,+,\alpha},2} (|\psi^0_n\rangle_1 + |\psi^0_n\rangle_2) \right) \right) \]

\[ |t_{3,+,\alpha}\rangle = \frac{\lambda_\alpha}{\mu_{t_{3,+,\alpha}} \sqrt{2}} \left( \sum_{n=1,4} \left( C_{t_{3,+,\alpha},1} (|\psi^0_n\rangle_0 + |\psi^0_n\rangle_1) + C_{t_{3,+,\alpha},2} (|\psi^0_n\rangle_1 + |\psi^0_n\rangle_2) \right) \right) \]

\[ |t_{4,+,\alpha}\rangle = \frac{\lambda_\alpha}{\mu_{t_{4,+,\alpha}} \sqrt{8}} \left( \sum_{n=1,3,6} \left( C_{t_{4,+,\alpha},1} (|\psi^0_n\rangle_0 + |\psi^0_n\rangle_1) + C_{t_{4,+,\alpha},2} (|\psi^0_n\rangle_1 + |\psi^0_n\rangle_2) \right) \right) \]
\[ |t_{6,\alpha} \rangle = \frac{\lambda_\alpha}{\sqrt{12}} \sum_{n=1}^{6} (-1)^n (|\psi_{n,0}^1\rangle + |1\rangle) \]
\[ |t_{6,\omega} \rangle = \frac{1}{\sqrt{12}} \left( \sum_{n=1}^{6} (-1)^{n-1} (|\psi_{n,1}^0\rangle - |\psi_{n,2}^0\rangle) \right) \]
\[ |q_{1,1\pm} \rangle = \frac{1}{2\sqrt{2}} \left( \sum_{n=1}^{2} (|\psi_{n,0}^0\rangle \pm |\psi_{n,2}^0\rangle) - \sum_{n=4}^{5} (|\psi_{n,0}^2\rangle \pm |\psi_{n,2}^2\rangle) \right) \]
\[ |q_{1,\alpha} \rangle = \frac{\lambda_\alpha}{4\sqrt{2}} \left( \sum_{n=1}^{6} (-1)^{n-1} (|\psi_{n,1}^1\rangle + |\psi_{n,1}^{-1}\rangle_o) + 3 \sum_{n=1,4}^{1} (-1)^n (|\psi_{n,1}^1\rangle + |\psi_{n,1}^{-1}\rangle_o) \right. \]
\[ \left. + \sum_{n=2}^{6} (|\psi_{n,0}^0\rangle + |\psi_{n,0}^{-1}\rangle_o) \right) \]
\[ |q_{1,\omega} \rangle = \frac{1}{\sqrt{24}} \left( 2 \sum_{n=1}^{6} |\psi_{n,0}^0\rangle - 2 \sum_{n=4}^{5} |\psi_{n,0}^2\rangle + \sum_{n=2}^{3} (|\psi_{n,0}^1\rangle + |\psi_{n,0}^{-1}\rangle_o) - \sum_{n=5}^{6} (|\psi_{n,0}^1\rangle + |\psi_{n,0}^{-1}\rangle_o) \right) \]
\[ |q_{2,1\pm} \rangle = \frac{1}{2\sqrt{2}} \left( \sum_{n=1}^{6} (-1)^{n-1} (|\psi_{n,2}^0\rangle \pm |\psi_{n,2}^2\rangle) + 3 \sum_{n=1,4}^{1} (-1)^n (|\psi_{n,2}^0\rangle + |\psi_{n,2}^{-1}\rangle_o) \right) \]
\[ |q_{2,\alpha} \rangle = \frac{\lambda_\alpha}{\sqrt{96}} \left( 3 \sum_{n=2}^{6} (|\psi_{n,0}^1\rangle + |\psi_{n,0}^{-1}\rangle_o) - 3 \sum_{n=1,4}^{5} (|\psi_{n,2}^1\rangle + |\psi_{n,2}^{-1}\rangle_o) + 3 \sum_{n=1,4}^{1} (-1)^n (|\psi_{n,0}^1\rangle + |\psi_{n,0}^{-1}\rangle_o) \right. \]
\[ \left. + 3 \sum_{n=2}^{5} (-1)^{n-1} (|\psi_{n,1}^1\rangle + |\psi_{n,1}^{-1}\rangle_o) \right) \]
\[ |q_{2,\omega} \rangle = \frac{1}{\sqrt{72}} \left( 6 \sum_{n=1,4}^{3} (-1)^n |\psi_{n,0}^0\rangle + 3 \sum_{n=1,4}^{5} (-1)^{n-1} (|\psi_{n,2}^1\rangle + |\psi_{n,2}^{-1}\rangle_o) \right) \]
\[ + \frac{1}{\sqrt{72}} \sum_{n=1}^{6} (-1)^n (|\psi_{n,1}^0\rangle + |\psi_{n,1}^2\rangle) - 2 |\psi_{n,0}\rangle \}
\[ |q_{3,1\pm} \rangle = \frac{1}{2\sqrt{2}} \left( \sum_{n=1}^{6} (|\psi_{n,0}^0\rangle \pm |\psi_{n,0}^{-1}\rangle_o) - \sum_{n=2}^{6} (|\psi_{n,0}^2\rangle \pm |\psi_{n,0}^{-2}\rangle) \right) \]
\[ |q_{3,\alpha} \rangle = \frac{\lambda_\alpha}{\sqrt{96}} \left( 3 \sum_{n=2,5}^{3} (|\psi_{n,0}^1\rangle \mp |\psi_{n,0}^{-1}\rangle_o) + (|\psi_{n,1}^0\rangle + |\psi_{n,1}^{-1}\rangle_o) - 6 \sum_{n=1}^{5} (|\psi_{n,0}^1\rangle + |\psi_{n,0}^{-1}\rangle_o) + (|\psi_{n,1}^0\rangle + |\psi_{n,1}^{-1}\rangle_o) \right) \]
\[ + 2 \sum_{n=2}^{3} (|\psi_{n,2}^1\rangle + |\psi_{n,2}^{-1}\rangle_o) - 4 (|\psi_{1,2}^1\rangle + |\psi_{1,2}^{-1}\rangle_o) \}
\[ |q_{3,\omega} \rangle = \frac{1}{\sqrt{8}} \left( \sum_{n=2,5}^{3} (|\psi_{n,0}^0\rangle \mp |\psi_{n,0}^2\rangle) + \sum_{n=6}^{3} (|\psi_{n,0}^0\rangle \mp |\psi_{n,0}^2\rangle) \right) \]
\[ |q_{4,1\pm} \rangle = \frac{1}{2\sqrt{2}} \left( \sum_{n=1,4}^{3} (|\psi_{n,0}^0\rangle \pm |\psi_{n,0}^{-2}\rangle) - 3 \sum_{n=1}^{6} (|\psi_{n,0}^2\rangle \pm |\psi_{n,0}^{-2}\rangle) \right) \]
\[ |q_{4,\alpha} \rangle = \frac{\lambda_\alpha}{4\sqrt{2}} \left( \sum_{n=1,4}^{3} (|\psi_{n,0}^1\rangle + |\psi_{n,0}^{-1}\rangle_o) + (|\psi_{n,1}^1\rangle + |\psi_{n,1}^{-1}\rangle_o) - \sum_{n=3,6}^{5} (|\psi_{n,0}^1\rangle + |\psi_{n,0}^{-1}\rangle_o) + (|\psi_{n,1}^1\rangle + |\psi_{n,1}^{-1}\rangle_o) \right) \]
\[ + 2 \sum_{n=1}^{3} (-1)^n (|\psi_{n,0}^1\rangle + |\psi_{n,0}^{-1}\rangle_o) \}
\[ |q_{4,\omega} \rangle = \frac{1}{\sqrt{24}} \left( 3 \sum_{n=1,4}^{3} (|\psi_{n,0}^0\rangle - |\psi_{n,0}^2\rangle) + \sum_{n=1}^{6} (|\psi_{n,0}^0\rangle - |\psi_{n,0}^2\rangle) \right) \]
\[ |q_{5,1\pm} \rangle = \frac{1}{12} \left( \sum_{n=1}^{6} (-1)^{n-1} (|\psi_{n,0}^0\rangle \pm |\psi_{n,0}^{-2}\rangle) \right) \]
where the upper and lower signs respectively refer to $\alpha = x$ and $y$, $\lambda_x = -1$ and $\lambda_y = i$.

\[
|q_{\alpha}\rangle = \frac{\lambda_\alpha}{\sqrt{12}} \left( \sum_{n=1}^{6} (-1)^{n-1} (|\psi^\alpha_n\rangle_1 + |\psi^{-\alpha}_n\rangle_1) \right)
\]

\[
|q_{\alpha}\rangle = \frac{1}{6} \left( \sum_{n=1}^{6} (-1)^{n-1} (|\psi^\alpha_n\rangle_0 + |\psi^\alpha_n\rangle_2) + 3 \sum_{n=1}^{2} (-1)^{n-1} |\psi^\alpha_n\rangle_3 \right)
\]

\[
|h_{1+}\rangle = \frac{1}{\sqrt{2}} (|\psi^\alpha_6\rangle \pm |\psi^{-\alpha}_6\rangle)
\]

\[
|h_{2+}\rangle = \frac{1}{\sqrt{12}} \sum_{n=1}^{6} (|\psi^\alpha_n\rangle \pm |\psi^{-\alpha}_n\rangle)
\]

\[
|h_\alpha\rangle = \frac{\lambda_\alpha}{\sqrt{20}} \left( \sum_{n=1}^{6} (|\psi^\alpha_n\rangle_0 + |\psi^\alpha_n\rangle_1 + |\psi^\alpha_n\rangle_2 + \sum |\psi^\alpha_n\rangle_3 \right)
\]

\[
|h_\lambda\rangle = \frac{1}{\sqrt{2}} (|\psi^\lambda_6\rangle \pm |\psi^{-\lambda}_6\rangle)
\]

\[
|h_{1-}\rangle = \frac{1}{\sqrt{2}} (|\psi^\lambda_6\rangle \pm |\psi^{-\lambda}_6\rangle)
\]

\[
|h_{2-}\rangle = \frac{1}{\sqrt{12}} \sum_{n=1}^{6} (|\psi^\lambda_n\rangle \pm |\psi^{-\lambda}_n\rangle)
\]

\[
|h_{\lambda\alpha}\rangle = \frac{\lambda_\alpha}{\sqrt{20}} \left( \sum_{n=1}^{6} (|\psi^\lambda_n\rangle_0 + |\psi^\lambda_n\rangle_1 + |\psi^\lambda_n\rangle_2 + \sum |\psi^\lambda_n\rangle_3 \right)
\]

Appendix B: DETAILS BOND OPERATOR THEORY

The six spin operators $S^\alpha_\alpha$ can be written in terms of singlet and triplet boson operators as
\[ S^\alpha_n = \left[ \left( \frac{1}{2^{n-1}} \right) \left( \frac{C_{\ell \pm 1} C_{\ell \pm 2} C_{\ell \pm 1} C_{\ell \pm 2} + C_{\ell \pm 3} C_{\ell \pm 3}}{3} + \frac{C_{\ell \pm 1} C_{\ell \pm 3} C_{\ell \pm 3}}{3} \right) \right] t_{1 \pm , \alpha} \]

\[
\left. + \left( - \frac{1}{6} \right) \left( \frac{\lambda_n \lambda_{n+1}}{2} \right) \left( C_{\ell \pm 1} C_{\ell \pm 2} C_{\ell \pm 1} + 2 C_{\ell \pm 3} C_{\ell \pm 3} \right) t_{1 \pm , \alpha} + \frac{\lambda_n - \lambda_{n+1}}{3} \left( C_{\ell \pm 1} C_{\ell \pm 2} \right) t_{1 \pm , \alpha} - \frac{\lambda_n \lambda_{n+1}}{2} \left( C_{\ell \pm 1} C_{\ell \pm 2} t_{1 \pm , \alpha} \right) \right] s_{1 \pm} \]

\[
\left. + \left( - \frac{1}{6} \right) \left( \frac{\lambda_n \lambda_{n+1}}{2} \right) \left( C_{\ell \pm 1} C_{\ell \pm 2} C_{\ell \pm 1} + 2 C_{\ell \pm 3} C_{\ell \pm 3} \right) t_{1 \pm , \alpha} + \frac{\lambda_n - \lambda_{n+1}}{3} \left( C_{\ell \pm 1} C_{\ell \pm 2} \right) t_{1 \pm , \alpha} - \frac{\lambda_n \lambda_{n+1}}{2} \left( C_{\ell \pm 1} C_{\ell \pm 2} t_{1 \pm , \alpha} \right) \right] s_{2 \pm} \]

\[
\left. + \left( - \frac{1}{6} \right) \left( \frac{\lambda_n \lambda_{n+1}}{2} \right) \left( C_{\ell \pm 1} C_{\ell \pm 2} C_{\ell \pm 1} + 2 C_{\ell \pm 3} C_{\ell \pm 3} \right) t_{1 \pm , \alpha} + \frac{\lambda_n - \lambda_{n+1}}{3} \left( C_{\ell \pm 1} C_{\ell \pm 2} \right) t_{1 \pm , \alpha} - \frac{\lambda_n \lambda_{n+1}}{2} \left( C_{\ell \pm 1} C_{\ell \pm 2} t_{1 \pm , \alpha} \right) \right] s_{3 \pm} \]

\[
\left. + \left( - \frac{1}{6} \right) \left( \frac{\lambda_n \lambda_{n+1}}{2} \right) \left( C_{\ell \pm 1} C_{\ell \pm 2} C_{\ell \pm 1} + 2 C_{\ell \pm 3} C_{\ell \pm 3} \right) t_{1 \pm , \alpha} + \frac{\lambda_n - \lambda_{n+1}}{3} \left( C_{\ell \pm 1} C_{\ell \pm 2} \right) t_{1 \pm , \alpha} - \frac{\lambda_n \lambda_{n+1}}{2} \left( C_{\ell \pm 1} C_{\ell \pm 2} t_{1 \pm , \alpha} \right) \right] s_{4 \pm} \]

\[
\left. + \left( - \frac{1}{6} \right) \left( \frac{\lambda_n \lambda_{n+1}}{2} \right) \left( C_{\ell \pm 1} C_{\ell \pm 2} C_{\ell \pm 1} + 2 C_{\ell \pm 3} C_{\ell \pm 3} \right) t_{1 \pm , \alpha} + \frac{\lambda_n - \lambda_{n+1}}{3} \left( C_{\ell \pm 1} C_{\ell \pm 2} \right) t_{1 \pm , \alpha} - \frac{\lambda_n \lambda_{n+1}}{2} \left( C_{\ell \pm 1} C_{\ell \pm 2} t_{1 \pm , \alpha} \right) \right] \]
The values of $\lambda_{n,k}$ are given below where $k = 1, 2, \cdots, 19$. Here $\alpha, \beta, \gamma = x, y, z$ and $\epsilon^{\alpha \beta \gamma}$ is the completely antisymmetric tensor with $\epsilon^{xyz} = 1$. Summation convention over repeated indices is implied here.

| $\lambda_{n,k}$ | $n = 1, 4$ | $n = 2, 5$ | $n = 3, 6$ | $\lambda_{n,k}$ | $n = 1, 4$ | $n = 2, 5$ | $n = 3, 6$ |
|-----------------|-------------|-------------|-------------|-----------------|-------------|-------------|-------------|
| $\lambda_{n,1}$ | 1           | $-\frac{1}{2}$ | $-\frac{1}{2}$ | $\lambda_{n,9}$ | 1           | 1           | 2           |
| $\lambda_{n,2}$ | 0           | 1           | 1           | $\lambda_{n,10}$ | 1           | -2          | -2          |
| $\lambda_{n,3}$ | 0           | -1          | 1           | $\lambda_{n,11}$ | 2           | $\frac{1}{7}$ | $\frac{1}{7}$ |
| $\lambda_{n,4}$ | 1           | -1          | 0           | $\lambda_{n,12}$ | 1           | 2           | 3           |
| $\lambda_{n,5}$ | 1           | 1           | -2          | $\lambda_{n,13}$ | 3           | 0           | 3           |
| $\lambda_{n,6}$ | 1           | 0           | -1          | $\lambda_{n,14}$ | 1           | 0           | 0           |
| $\lambda_{n,7}$ | 1           | -2          | 1           | $\lambda_{n,15}$ | 1           | 4           | 1           |
| $\lambda_{n,8}$ | 1           | 1           | $\frac{2}{5}$ | $\lambda_{n,16}$ | 3           | 2           | 1           |

The analytic expressions of the coefficients $A^n_a$, $B^n_a$ and $D^n_{ab}$ are given below

\[
A_1^n = \frac{(-1)^{n-1}}{2\mu_{s_1}\mu_{t_1}} \left( C_{s_0,1}C_{t_1,2} + \frac{C_{s_0,2}C_{t_1,1}}{3} + \frac{C_{s_0,3}C_{t_1,3}}{3} \right),
\]

\[
A_2^n = \frac{(-1)^{n-1}\lambda_{n,1}}{3\sqrt{2}\mu_{s_2}\mu_{t_2}} \left( C_{s_1,2}C_{t_2,3} - 2C_{s_1,3}C_{t_2,2} \right),
\]

\[
A_3^n = \frac{(-1)^{n-1}\lambda_{n,3}}{2\sqrt{6}\mu_{s_2}\mu_{t_2}} \left( C_{s_1,2}C_{t_3,1} - 2C_{s_1,3}C_{t_3,2} \right),
\]

\[
B_1^n = 0, \quad B_2^n = \frac{\lambda_{n,3}}{2\sqrt{2}\mu_{t_2}} C_{t_{2,1}}, \quad B_3^n = -\frac{\lambda_{n,1}}{\sqrt{6}\mu_{t_3}} C_{t_{3,1}},
\]

\[
D_{11}^n = \frac{1}{6}, \quad D_{22}^n = \frac{1}{6\mu_{t_2}^2} \left( \lambda_{n,11}(C_{t_{1,3}}C_{t_{2,1}} + C_{t_{1,2}}C_{t_{2,3}}) - \lambda_{n,10}C_{t_{2,3}}C_{t_{3,2}} \right),
\]

\[
D_{33}^n = \frac{1}{2\mu_{t_2}^2} \left( \lambda_{n,2}\left( C_{t_{1,2}}C_{t_{3,1}} + C_{t_{1,1}}C_{t_{3,2}} \right) + \lambda_{n,14}C_{t_{3,2}}C_{t_{3,1}} \right),
\]

\[
D_{12}^n = \frac{\lambda_{n,1}}{3\sqrt{2}\mu_{t_1}\mu_{t_2}} \left( C_{t_{1,3}}C_{t_{2,1}} + C_{t_{1,2}}C_{t_{2,3}} - 2C_{t_{1,3}}C_{t_{2,2}} \right),
\]

\[
D_{31}^n = \frac{\lambda_{n,3}}{2\sqrt{6}\mu_{t_1}\mu_{t_3}} \left( C_{t_{1,3}}C_{t_{3,1}} + C_{t_{1,2}}C_{t_{3,2}} - 2C_{t_{1,3}}C_{t_{3,2}} \right),
\]

\[
D_{23}^n = -\frac{\lambda_{n,3}}{4\sqrt{3}\mu_{t_2}\mu_{t_3}} \left( C_{t_{2,3}}C_{t_{3,1}} + C_{t_{2,2}}C_{t_{3,2}} - 2C_{t_{2,3}}C_{t_{3,2}} \right).
\]

Appendix C: MEAN-FIELD APPROXIMATION DETAILS

The analytic expressions of the coefficients $X_{k}^{ab}$ and $Y_{k}^{ab}$ are given as
\[ X_{k}^{ab} = (E_{t_a} - \mu) (\delta_{a,1} \cdot \delta_{b,1} + \delta_{a,2} \cdot \delta_{b,2} + \delta_{a,3} \cdot \delta_{b,3}) + Y_{k}^{ab}, \]
\[ Y_{k}^{ab} = N_0 \sum_{n} 2g^{ab}(n) \cos (k \cdot n) \]
\[ g^{ab}(n) = J_1 (A^1_a A^6_b \delta_{n,1} + A^2_a A^5_b \delta_{n,1+2} + A^3_a A^6_b \delta_{n,2}) + J_2 \left( (A^1_a A^5_b + A^1_a A^3_b + A^2_a A^4_b) + A^6_a A^4_b \delta_{n,1} + (A^2_a A^6_b + A^3_a A^4_b + A^3_a A^5_b + A^6_a A^6_b \delta_{n,2} + (A^1_a A^5_b + A^2_a A^6_b + A^3_a A^4_b + A^5_a A^6_b) \delta_{n,1+2} \right) \]

Here \( a, b = 1, 2, 3 \) and \( E_{t_a} \) is the triplet energy of the single plaquette.

The analytical procedure used to diagonalize the triplet sector of the harmonic Hamiltonian (15) has been described below. Instead of \( \hat{H}_k \), \( \hat{I}_B \hat{H}_k \) has been diagonalized, where

\[ \hat{I}_B = \begin{pmatrix} \hat{I} & 0 \\ 0 & -\hat{I} \end{pmatrix} \]

with \( \hat{I} \) is the \( 3 \times 3 \) identity matrix. The characteristic equation and (positive) eigenvalues of the matrix (C2) are

\[ \Omega_k^6 + a_{2,k} \Omega_k^4 + a_{1,k} \Omega_k^2 + a_{0,k} = 0 \]
\[ \Omega_{a,k} = 2 \sqrt{-Q \cos\left(\frac{\theta}{3} - \frac{2\pi p}{3}\right) - \frac{a_{2,k}}{3}} \]
\[ Q_k = \frac{3a_{1,k} - a_{2,k}^2}{9} \]
\[ R_k = \frac{9a_{2,k}a_{1,k} - 27a_{0,k} - 2a_{1,k}^3}{54} \]
\[ \cos(\theta) = \frac{-R_k}{Q_k \sqrt{-Q_k^2}} \]

\( p = 0, 1, 2 \) for \( a = 1, 2, 3 \).

The coefficients \( a_{i,k} \) are given below
\[ a_{2,k} = - \left( w_{11,k}^2 + w_{22,k}^2 + w_{33,k}^2 \right) \]
\[ a_{1,k} = w_{11,k}^2 w_{22,k}^2 + w_{11,k}^2 w_{33,k}^2 + w_{22,k}^2 w_{33,k}^2 - 4(Y_{12}^2) (X_{11}^k - Y_{11}^k) (X_{33}^k - Y_{22}^k) \]
\[ - 4(Y_{23}^2) (X_{33}^k - Y_{22}^k) (X_{33}^k - Y_{33}^k) - 4(Y_{13}^2) (X_{11}^k - Y_{11}^k) (X_{33}^k - Y_{33}^k) \]
\[ a_{0,k} = \left( X_{11}^k - Y_{11}^k \right) \left( X_{33}^k - Y_{22}^k \right) \left( X_{33}^k - Y_{33}^k \right) \left[ 4(Y_{12}^2) (X_{33}^k + Y_{33}^k) \right. \]
\[ + 4(Y_{23}^2) (X_{33}^k + Y_{11}^k) + 4(Y_{13}^2) (X_{33}^k + Y_{22}^k) - 16Y_{12}^2 Y_{13}^2 Y_{23}^2 \]
\[ \left. - \left( X_{11}^k + Y_{11}^k \right) \left( X_{33}^k + Y_{22}^k \right) \left( X_{33}^k + Y_{33}^k \right) \right] \]

where \( w_{ij,k}^2 = (X_{ij}^k)^2 - (Y_{ij}^k)^2 \) with \( i, j = 1, 2, 3 \).

Using the procedure described in [17] the Bogoliubov coefficients has been determined. The Bogoliubov coefficients \( u_{ab}^k \) and \( v_{ab}^k \) are given below

\[ u_{ab}^k = \frac{\phi_{ab}^k + \psi_{ab}^k}{2} \]  \hspace{1cm} \[ v_{ab}^k = \frac{\phi_{ab}^k - \psi_{ab}^k}{2} \]  \hspace{1cm} (C5)

with \( a, b = 1, 2 \) and 3. Where,
\[ \phi_{b,k}^{1b} = x_{b,k} \sqrt{X_{11}^b - Y_{11}^b} \]
\[ \phi_{b,k}^{2b} = y_{b,k} \sqrt{X_{22}^b - Y_{22}^b} \]
\[ \phi_{b,k}^{3b} = z_{b,k} \sqrt{X_{33}^b - Y_{33}^b} \]
\[ \psi_{b,k}^{1b} = x_{b,k} (X_{11}^b + Y_{11}^b) \sqrt{X_{11}^b - Y_{11}^b} + 2y_{b,k} Y_{12}^b \sqrt{X_{22}^b - Y_{22}^b} + 2z_{b,k} Y_{13}^b \sqrt{X_{33}^b - Y_{33}^b} \]
\[ \psi_{b,k}^{2b} = 2x_{b,k} Y_{12}^b \sqrt{X_{11}^b - Y_{11}^b} + y_{b,k} (X_{22}^b + Y_{22}^b) \sqrt{X_{22}^b - Y_{22}^b} + 2z_{b,k} Y_{23}^b \sqrt{X_{33}^b - Y_{33}^b} \]
\[ \psi_{b,k}^{3b} = 2x_{b,k} Y_{13}^b \sqrt{X_{11}^b - Y_{11}^b} + 2y_{b,k} Y_{23}^b \sqrt{X_{22}^b - Y_{22}^b} + z_{b,k} (X_{33}^b + Y_{33}^b) \sqrt{X_{33}^b - Y_{33}^b} \]

\[ x_{b,k} = \frac{M_{b,k}}{\sqrt{G_{b,k}}} \]
\[ y_{b,k} = \frac{1}{\sqrt{G_{b,k}}} \]
\[ z_{b,k} = \frac{N_{b,k}}{\sqrt{G_{b,k}}} \]
\[ M_{b,k} = \frac{A_{k} C_{k} - (w_{22,k} - \Omega_{2,k}^2) B_{k}}{A_{k} B_{k} - (w_{11,k} - \Omega_{2,k}^2) C_{k}} \]
\[ N_{b,k} = \frac{C_{k} + B_{k} M_{b,k}}{(\Omega_{2,k}^2 - w_{33,k}^2)} \]
\[ G_{b,k} = \Omega_{b,k} [1 + M_{b,k}^2 + N_{b,k}^2] \]
\[ A_{k} = 2Y_{12}^b \sqrt{X_{11}^b - Y_{11}^b} \sqrt{X_{22}^b - Y_{22}^b} \]
\[ B_{k} = 2Y_{13}^b \sqrt{X_{11}^b - Y_{11}^b} \sqrt{X_{33}^b - Y_{33}^b} \]
\[ C_{k} = 2Y_{23}^b \sqrt{X_{22}^b - Y_{22}^b} \sqrt{X_{33}^b - Y_{33}^b} \]

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