Quantum phase diagram of distorted $J_1 - J_2$ Heisenberg $S = 1/2$ antiferromagnet in honeycomb lattice: a modified spin wave study

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Abstract
Using the modified spin wave method, we study the $J_1 - J_2$ Heisenberg model with first and second neighbor antiferromagnetic exchange interactions. For a symmetric $S = 1/2$ model, with the same couplings for all the equivalent neighbors, we find three phases in terms of the frustration parameter $\alpha = J_2/J_1$: (1) a commensurate collinear ordering with staggered magnetization (Néel.I state) for $0 < \alpha \lesssim 0.207$, (2) a magnetically gapped disordered state for $0.207 < \alpha < 0.369$, preserving all the symmetries of the Hamiltonian and lattice, which by definition is a quantum spin liquid (QSL) state and (3) a commensurate collinear ordering in which two out of the three nearest neighbor magnetizations are antiparallel and the remaining pair are parallel (Néel.II state), for $0.396 < \alpha \leq 1$. We also explore the phase diagram of a distorted $J_1 - J_2$ model with $S = 1/2$. Distortion is introduced as an inequality of one nearest neighbor coupling with the other two. This yields a richer phase diagram by the appearance of a new gapped QSL, a gapless QSL and also a valence bond crystal phase in addition to the previous three phases found for the undistorted model.

Keywords: quantum spin liquid, magnetic frustration, modified spin wave theory, honeycomb antiferromagnets

(Some figures may appear in colour only in the online journal)

1. Introduction

The recent synthesis of compounds consisting of transition metal-oxide layers with honeycomb structures has drawn attention to the magnetic properties of spin models in honeycomb lattices. Three experimental realizations of honeycomb magnetic materials are InCu$_{2/3}$V$_{1/3}$O$_3$ [1], Cu$_3$Ni$_2$SbO$_6$ [3] and Bi$_3$Mn$_4$O$_{12}$(NO$_3$) (BMNO) [2]. Cu$^{2+}$ ions with $S = 1/2$ in the first, Ni$^{2+}$ ions with $S = 1$ in the second and Mn$^{4+}$ ions with $S = 3/2$ in the third compound reside on the lattice points of weakly coupled honeycomb layers. InCu$_{2/3}$V$_{1/3}$O$_3$ develops antiferromagnetic (AF) ordering below $\sim 20$ K [4]. However, for BMNO magnetic susceptibility and specific heat measurements show no sign of magnetic ordering down to $T = 0.4$ K, in spite of high Curie–Weiss temperature $T_{CW} \approx -257$ K [2].

On the theoretical front, the large-scale quantum Monte Carlo (QMC) simulation of the half-filled Hubbard model on a honeycomb lattice proposes a gapped quantum spin liquid (QSL) phase (a magnetically disordered state preserving all the symmetries of the Hamiltonian and the lattice) for intermediate values of on-site Coulomb interaction between the AF-Mott insulating and semi-metallic phases [5]. Although later QMC simulations on larger lattice sizes refuted the existence of such a QSL phase [6–8], many researches were still devoted to the study of AF spin models in honeycomb structures [9–41].
Since a honeycomb lattice is bipartite, the Heisenberg model with nearest neighbor AF interactions in this lattice is not frustrated and develops long-range Néel ordering. However, enhanced quantum fluctuations, due to the small coordination number ($Z = 3$), reduce staggered magnetization by about half of its classical value [10, 12, 13, 16]. Therefore, the expectation for the realization of a QSL phase in honeycomb-based magnets requires the introduction of frustrating exchange interactions. The simplest model incorporating frustration effects on the honeycomb lattice is the $J_1 - J_2$ Heisenberg model, where $J_1 > 0$ and $J_2 > 0$ are the nearest and next nearest neighbor AF exchange interactions, respectively. The classical phase diagram of this model, studied by Katsura et al [9], shows that the Néel ordered phase is stable for $J_2/J_1 < 1/6$. However, for $1/6 < J_2/J_1 < 1/2$ the classical ground state becomes infinitely degenerate and can be characterized by a manifold of spiral wave vectors. Okumura et al used low-temperature expansion and Monte Carlo simulation to show that such large ground-state degeneracy can be lifted by thermal fluctuations in such a way that a broken symmetry state, with a three-fold ($C_3$) symmetry of the honeycomb lattice, would be selected [17]. In the vicinity of the AF phase boundary ($J_2/J_1 \approx 1/6$), the energy scale associated with such a thermal order-by-disorder mechanism becomes extremely small, leading to exotic spin liquid behaviors, whereby the spin structure factor would have a different pattern in comparison with the paramagnetic phase [17].

An order-by-disorder mechanism driven by quantum fluctuations has been studied by Mulder et al. They showed that spin wave corrections lower the energy of some states with particular incommensurate wave vectors in the ground-state manifold, for the classically degenerate region $1/6 < J_2/J_1 < 1/2$ [18]. They also argued that for $S = 1/2$, over a wide range of $J_2/J_1$ in the frustrated region, strong quantum fluctuations can melt this spiral ordering into a valence bond solid (VBS) with staggered dimerized ordering, which breaks the $C_3$ rotational symmetry of the lattice while preserving its translational symmetry [18]. Such a nematic ordering has already been proposed in exact diagonalization (ED) calculations [14] and in nonlinear sigma model (NLSM) formulation [15]. ED calculations in both $S = 0$, and a nearest neighbor valence bond (NNVB) basis, show that the NNVB basis provides a very good description of the ground state for $0.2 \lesssim J_2/J_1 \lesssim 0.3$ [20, 21]. Furthermore, analysis of the ground-state properties by defining appropriate structure factors suggests a plaquette VBS ground state for $0.2 \lesssim J_2/J_1 \lesssim 0.35$ which transforms to a VBS state with staggered dimerization at $J_2/J_1 \approx 0.35$ [20, 21]. The existence of a plaquette VBS has been verified by different methods, such as the functional renormalization group [22], the coupled cluster method (CCM) [23, 24], mean-field plaquette valence bond theory [25, 27] and the density matrix renormalization group (DMRG) [26, 27]. However, other methods such as the Schwinger boson mean-field approach [28, 29, 32, 33], Schwinger fermion mean-field theory [30] and variational Monte Carlo [31] propose a $Z_2$ QSL for the disordered region.

In this work we use the the modified spin wave (MSW) theory to study both symmetric and distorted $J_1 - J_2$ Heisenberg AF with an $S = 1/2$ model in the honeycomb lattice. This paper is organized as follows: the model Hamiltonian and the MSW method are introduced in section 2. The MSW phase diagram of the symmetric and distorted model is discussed in sections 3 and 4. Section 5 is devoted to conclusions.

\section{Model Hamiltonian and MSW formalism}

The $J_1 - J_2$ Heisenberg AF Hamiltonian is defined by

\begin{equation}
H = \frac{1}{2} \sum_{nn} J_{nn} S_i \cdot S_j + \frac{J_2}{2} \sum_{nnn} S_i \cdot S_j,
\end{equation}

in which $nn$ and $nnn$ denote the nearest and next nearest neighbors, respectively, and the exchange couplings $J_{nn} > 0$ and $J_2 > 0$ denote the first and second neighbor couplings. Here we consider the case where the nearest neighbor couplings are equal to $J_1$, for the bond denoted by the vector $\delta_1$ and $\delta_2$ for the bonds denoted by $\delta_2$ and $\delta_3$ (see figure 1). Now we redefine the couplings as follows

\begin{equation}
J'_1 = J_1 (1 - \bar{\delta}), \quad J_2 = J_2 (1 + 2\bar{\delta}), \quad \bar{\delta} = J_2/J_1,
\end{equation}

where $\bar{\delta} = \frac{\delta + 2\delta'_2}{3}$ and the dimensionless quantities $\delta$ and $\bar{\delta}$ denote the distortion and frustration, respectively.

Now, we give a brief introduction to the formalism of MSW theory in a bipartite lattice, and then apply it to the Hamiltonian (1). The MSW was introduced by Takahashi [42] and its basic assumption is that the ground state of the spin Hamiltonian in the classical limit ($S \rightarrow \infty$) is long-range ordered. It has been shown that the minimum energy condition for the classical $J_1 - J_2$ Heisenberg model gives rise to planar states [9, 14]. Hence, the translational invariance requires that the ordered ground state is characterized by a planar wave vector $\mathbf{Q}$. Under this assumption, it is convenient to rotate the coordinates axes $(x, y, z)$ locally to $(\eta_i, \zeta_i, \xi_i)$ at each site $i$, in such a way that $\zeta_i$ represents the local symmetry breaking axis. For this purpose, we introduce the following spin transformations for the honeycomb lattice which contains two lattice points per unit cell.
Figure 2. Schematic representation of (a) Néel.I spin configuration comprised of two magnetic sublattices A and B, and (b) Néel.II and (c) Néel.III states consisting of four magnetic sublattices A–D.

\[
S_{i,j}^{\gamma} = -\sin(Q \cdot r_i + (\gamma - 1)\phi)S_{i,j}^{0} \\
+ \cos(Q \cdot r_i + (\gamma - 1)\phi)S_{i,j}^{0} \\
S_{i,j}^{0} = -S_{i,j}^{0} \\
\]

where \( r_i \) denotes the position of each spin, \( \gamma = 1, 2 \) refers to the two lattice points (A, B sublattices) in the unit cell identified by the vectors \( \delta_0 = 0 \) and \( \delta_1 = \frac{a}{\sqrt{2}} \) (see figure 1) and the

\[
H = \frac{1}{2} \sum_{n,m} J_n [(S_i^0)^2(S_j^0)^2 + S_i^{S_j,2} + S_i^{S_j,2} \cos(Q \cdot (r_i - r_j) + \phi) + (S_i^{S_j,2})^2] \\
+ \frac{J_2}{2} \sum_{n,m} \sum_{\gamma=1}^{2} [S_i^{S_j,\gamma} + S_i^{S_j,\gamma}] \cos(Q \cdot (r_i - r_j) + \phi) + S_i^{S_j,\gamma}] \\
+ \frac{J_2}{2} \sum_{n,m} \sum_{\gamma=1}^{2} [S_i^{S_j,\gamma} + S_i^{S_j,\gamma}] \cos(Q \cdot (r_i - r_j) + \phi) + S_i^{S_j,\gamma}] \\
+ \frac{J_2}{2} \sum_{n,m} \sum_{\gamma=1}^{2} [S_i^{S_j,\gamma} + S_i^{S_j,\gamma}] \cos(Q \cdot (r_i - r_j) + \phi) + S_i^{S_j,\gamma}] \\
+ \frac{J_2}{2} \sum_{n,m} \sum_{\gamma=1}^{2} [S_i^{S_j,\gamma} + S_i^{S_j,\gamma}] \cos(Q \cdot (r_i - r_j) + \phi) + S_i^{S_j,\gamma}] \\
\]

We use Dyson–Maleev (DM) transformations to obtain a bosonic representation of the spin Hamiltonian. For a bipartite lattice, like the honeycomb lattice, DM transformations are given by

\[
S_{i,j}^{(0)} = \frac{1}{\sqrt{2S}} (2S - a^+_ia_i), \quad S_{i,j}^{(1)} = \frac{1}{\sqrt{2S}} (2S - b^+_ib_i), \\
S_{i,j}^{(1)} = S - a^+_ia_i, \quad S_{i,j}^{(2)} = S - b^+_ib_i, \\
\]

in which \( a, b \) represent the bosonic operators in the A and B sublattices, respectively, and \( S \) is the value of the spins. In the above transformations the quantization axes are taken to be the local \( \zeta \) axes and \( S_{i,j}^{(1)} \equiv S_{i,j}^{(1)} \mp iS_{i,j}^{(2)} \). The commutation relations \([S_{i,j}^{(0)}, S_{i,j}^{(1)}] = \epsilon_{\alpha\beta\gamma} S_{i,j}^{(\alpha)} S_{i,j}^{(\beta)} S_{i,j}^{(\gamma)} \) are satisfied by the bosonic algebra between the \( a \) and \( b \) operators, i.e., \([a, a^+_j] = \delta_{ij}, \quad [b, b^+_j] = \delta_{ij}, \quad [a, a^+_i] = [b, b^+_i] = [a, b] = 0\).

Substituting the transformations (5) into the Hamiltonian (3), one finds the following bosonic Hamiltonian

\[
H = \frac{1}{4} \sum_{n,m} J_n [2S(a^+_ib_j + a^+_jb_i) - a^+_ib_ip_j - a^+_ia_ip_j + 3S(a^+_ia_j + a^+_aj + a^+_ia_i) \cos(Q \cdot (r_i - r_j) + \phi)] \\
\]

angle \( \phi \) denotes the relative rotation of the symmetry breaking axes within a unit cell. Unlike ordinary spin-wave theory, we do not make any assumption on the ordering vector \( Q \) which may differ from the classical ordering wave vector.

Applying the transformations (3) to the Hamiltonian (1), we find

\[
H = \frac{1}{2} \sum_{n,m} J_n [(S_i^0)^2(S_j^0)^2 + S_i^{S_j,2} + S_i^{S_j,2} \cos(Q \cdot (r_i - r_j) + \phi) + (S_i^{S_j,2})^2] \\
+ \frac{J_2}{2} \sum_{n,m} \sum_{\gamma=1}^{2} [S_i^{S_j,\gamma} + S_i^{S_j,\gamma}] \cos(Q \cdot (r_i - r_j) + \phi) + S_i^{S_j,\gamma}] \\
+ \frac{J_2}{2} \sum_{n,m} \sum_{\gamma=1}^{2} [S_i^{S_j,\gamma} + S_i^{S_j,\gamma}] \cos(Q \cdot (r_i - r_j) + \phi) + S_i^{S_j,\gamma}] \\
+ \frac{J_2}{2} \sum_{n,m} \sum_{\gamma=1}^{2} [S_i^{S_j,\gamma} + S_i^{S_j,\gamma}] \cos(Q \cdot (r_i - r_j) + \phi) + S_i^{S_j,\gamma}] \\
+ \frac{J_2}{2} \sum_{n,m} \sum_{\gamma=1}^{2} [S_i^{S_j,\gamma} + S_i^{S_j,\gamma}] \cos(Q \cdot (r_i - r_j) + \phi) + S_i^{S_j,\gamma}] \\
\]

where \( r_j = r_i - r_j \) is equal to \( \delta_1, \delta_2, \delta_3 \) for the nearest neighbors and \( \pm \delta_1, \pm \delta_2, \pm \delta_3 \) for the next nearest neighbors (figure 1). Now, we use mean field theory to find an expression for the expectation value of the Hamiltonian (6), i.e., \( E = \langle H \rangle \). For this purpose, we use Wick’s theorem to calculate the expectation value of the quartic terms; hence we find
\[
E = -\frac{N}{2} \sum_{\delta} J(\delta) (\Delta + f(0) + g(\delta)^2 (1 - \cos(\mathbf{Q} \cdot \delta + \phi)) - [S + \frac{1}{2} - f(0) + g(\delta)^2 (1 + \cos(\mathbf{Q} \cdot \delta + \phi))]
\]

\[
J_2 \sum_{\delta} ((S + \frac{1}{2} - f(0) + g(\delta)^2 (1 - \cos(\mathbf{Q} \cdot \delta')))
\]

\[
- [S + \frac{N}{2} - f(0) + g(\delta')^2 (1 + \cos(\mathbf{Q} \cdot \delta'))],
\]

(7)

in which \(\delta\) and \(\delta'\) denote the first and second neighbors, respectively, \(J(\delta) = J_0, J(\delta_2) = J(\delta_3) = J_1 \) and \(N\) is the number of sites. Functions \(f\) and \(g\) denote the expectation value of the hopping and pairing of DM bosons defined as

\[
\langle a_i \rangle = \langle a_i \rangle = \langle b_i \rangle = \langle b_i \rangle = f(\delta) - \frac{1}{2} \delta_{ij},
\]

\[
\langle a_i \rangle = \langle a_i \rangle = \langle b_i \rangle = \langle b_i \rangle = g(\delta),
\]

\[
\langle a_i^\dagger \rangle = \langle a_i \rangle = \langle b_i \rangle = \langle b_i \rangle = g(\delta),
\]

(8)

Then using equations (7) and (8), one finds the following expression for the ground-state energy per site, \(E_0 = E/N\), in mean field approximation

\[
E_0 = \epsilon_0 + \epsilon_1 \cos(\phi)
\]

\[
+ \left\{ \cos(\frac{Q_1}{2} + \frac{\sqrt{3} Q_x}{2} + \phi) + \cos(\frac{Q_1}{2} + \frac{\sqrt{3} Q_x}{2} + \phi) \right\}
\]

\[
+ \epsilon_2 \left\{ \cos(\frac{Q_1}{2} + \frac{\sqrt{3} Q_x}{2}) + \cos(\frac{Q_1}{2} + \frac{\sqrt{3} Q_x}{2}) \right\}
\]

\[
(9)
\]

where

\[
\epsilon_0 = \sum_{\delta} J(\delta) (f(\delta)^2 - g(\delta)^2) + \frac{J_2}{2} \sum_{\delta} (f(\delta')^2 - g(\delta')^2),
\]

\[
\epsilon_1 = \frac{J_1}{2} (f(\delta^2) + g(\delta^2)), \epsilon_1 = \frac{J_1}{4} \sum_{i=1}^{\delta} (f(\delta^2) + g(\delta^2)),
\]

\[
\epsilon_2 = \frac{J_2}{12} \sum_{\delta} (f(\delta')^2 + g(\delta')^2).
\]

The first step in the MSW procedure is to minimize the energy (9) with respect to the ordering vector \(\mathbf{Q}\). This incorporates the competition between states with long-range ordering (LRO) at different ordering vectors \(\mathbf{Q}\) which may not necessarily be stable at the classical level [43]. The next step is to minimize \(E_0\) with respect to \(f_0\) and \(g_0\). In the absence of an external field, this minimization is done under the constraint that the expectation value of spins along the local quantization axes vanishes. The constraint \(\langle S_i \rangle = 0\), introduced by Takahashi [42], keeps the physical requirement that the number of DM bosons per site be less than 2\(S\) (\(n \leq 2S\)).

\[
\langle S_i \rangle = -S + \langle a_i^\dagger a_i \rangle = -S + \langle b_i^\dagger b_i \rangle = -S + \langle f(0) \rangle = 0.
\]

(11)

Takahashi’s constraint reduces the Hilbert space dimension available to the DM bosons by reducing their average density to \(S\). In a bipartite lattice, one can in fact show a significant reduction of the Hilbert space dimension from \(2^S\) to \(\frac{4^2 \phi}{\pi} \) for \(S = 1/2\) [44].

For a given ordering wave vector \(\mathbf{Q}\) and rotation \(\phi\), an appropriate set of Bogoliubov transformations are defined, in terms of which the Hamiltonian equation (6) can be diagonalized in mean field approximation. Moreover, the quantities \(f\) and \(g\) defined by equation (8) can be parameterized in terms of the coefficients of the Bogoliubov transformations, allowing us to minimize the total energy with respect to these coefficients, under Takahashi’s constraint (11). To satisfy Takahashi’s constraint we need to introduce a Lagrange multiplier \(\mu\) which plays the role of chemical potential for the DM bosons. In bosonic language, a magnetically ordered state can be translated to a Bose–Einstein condensate (BEC), for which \(\mu = 0\) [45]. For the magnetic disordered states the spontaneous magnetization is zero; hence there is no reason for the vanishing of the chemical potential. In this case \(\mu\) has to be calculated self-consistently to give the gap of the magnon dispersion. The MSW gives a set of self-consistent equations for \(g\) and \(f\), whose outputs are the ground-state energy, magnon energy spectrum, magnetization and spin–spin correlations. The details of this procedure are given in appendices A and B. In the next section we apply MSW theory to the symmetric \(J_1 - J_2\) model.

3. MSW phase diagram of symmetric \(J_1 - J_2\) model

For the symmetric model \(J_1 = J_1\), minimizing the total energy (9) with respect to \(Q_1, Q_2, \phi\) gives rise to numerous commensurate and incommensurate solutions. The commensurate minima are achieved by a two-sublattice collinear ordering, given by \(Q = (0, 0), \phi = \pi\) (Néel.1), and two types of four-sublattice collinear ordering with \(Q = (\pi, \frac{\pi}{\sqrt{3}}), \phi = \pi\) (Néel.1 (MSW) * Néel.1 (LSW) * QSL.1 (MSW) * Néel.2 (MSW) *)

Figure 3. Ground-state energy per site (in units of \(J_1\)) of symmetric \(J_1 - J_2\) model versus the frustration parameter \(\alpha\) for \(S = \frac{1}{2}\). The ground state is Néel.1 ordered for \(0.0 \leq \alpha \leq 0.207\) (pink triangles), quantum spin liquid with Néel.1 type symmetry (QSL.1) for \(0.207 \leq \alpha \leq 0.396\) (blue rhomboids) and Néel.2 ordered for \(0.396 \leq \alpha \leq 1.0\) (red diamonds). The gray squares show the ground-state energies per site, obtained in linear spin wave (LSW) approximation in the Néel.1 phase.
II), and \( Q = (0, \frac{2\pi}{\sqrt{3}}), \phi = 0 \) (Néel.III). The schematic spin configurations in these states are illustrated in figure 2. The incommensurate solutions are given by the spiral states \( Q = (2 \cos^{-1}(\pm \frac{3\pi/2}{\phi}), 0 \text{ or } \frac{2\pi}{3}), \phi = 0 \text{ or } \pi, \) and \( Q = (0, \frac{2\pi}{\sqrt{3}}(\sin^{-1}(\frac{1}{\sqrt{2}} \sin(\phi)) - \phi)), \phi = \cos^{-1}(\frac{2\pi}{\phi} - \frac{3\pi}{4\epsilon_2} + \pi), \) where \( \epsilon_1 \) and \( \epsilon_2 \) are given by equation (10).

Having a long-range ordered ground state requires the gapless excitation spectrum as a result of the Goldstone theorem. This condition leads to the vanishing of the chemical potential \( \mu \) (defined by equation (A.6)) in the ordered state as a requirement of BEC transition [45]. To calculate the energy and magnetization for each type of ordering one needs to solve the self-consistent equations (A.7)–(A.11), with \( \mu = 0 \). After convergence, these equations give the spontaneous magnetization \( M_0 \) and the functions \( f_0 \) and \( g_\mu \), and then substitution of \( f_0 \) and \( g_\mu \) in equation (7) gives the ground-state energy per site \( E_0 \). The magnon excitation spectrum is given by equation (A.12), and spin–spin correlations can be calculated by equations (A.13) and (A.14). For Néel.II and III orderings, it is more convenient to use a four-sublattice unit cell (figures 2(b) and (c)), where the ordering wave vector is \( \mathbf{Q} = 0 \). Using a larger unit cell in real space leads to the reduction of the size of the magnetic Brillouin zone in \( \mathbf{K} \)-space and thus of the number of singular points, hence making the convergence of corresponding self-consistent equations much easier (see appendix B for details). In this case, the physical quantities of interest can be calculated by solving the set of equation (B.5).

Following the above procedure, the MSW results in the following: within the possible ordered state, Néel.I and Néel.II acquire the minimum energy for \( 0 \lesssim \tilde{\alpha} \lesssim 0.207 \) and \( \tilde{\alpha} \gtrsim 0.25 \), respectively. The dependence of the ground-state energy per site \( E_0 \) and the corresponding spontaneous magnetization \( (M_0) \) on the frustration parameter \( \tilde{\alpha} \) is illustrated in figures 3 and 4, respectively. In figures 3 and 4, \( E_0 \) and \( M_0 \) obtained by linear spin wave (LSW) theory are also represented for the Néel.I state. The LSW indicates that the Néel.I phase is stable only to \( \tilde{\alpha} \approx 0.11 \). Therefore, a comparison between the LSW and MSW shows that nonlinear interactions, taken into account by the MSW in the mean field approximation, lower the energy of the Néel.I phase and increase its stability against frustration up to \( \tilde{\alpha} \approx 0.207 \).

On the other hand, for \( \tilde{\alpha} > 0.25 \) we found that Néel.II has lower energy with respect to Néel.III and the spiral states. The energy per site and magnetization, corresponding to this type of ordering, are plotted for the range \( 0.25 < \tilde{\alpha} \lesssim 0.8 \) in figures 3 and 4. It is important to mention that the Néel.II state is not a classically stable state. Indeed assuming such an ordering and using LSW approximation, it is found that complex numbers appear in its spin excitation spectrum which makes this state unstable. Hence, the stability of this phase in the MSW can be attributed to nonlinear magnon–magnon interactions.

For the interval \( 0.207 \lesssim \tilde{\alpha} \lesssim 0.25 \), however, no ordered state is found to be stable. Indeed, the magnetization of the Néel.II state falls continuously to zero at \( \tilde{\alpha} \approx 0.207 \), above which no stable solution of self-consistent equations corresponding to Néel.II ordering is possible with the BEC condition \( \mu = 0 \). However, starting from the Néel.I state and relaxing the BEC condition and setting \( M_0 = 0 \), it is possible to obtain from MSW equations a magnetically disordered state with finite chemical potential and vanishing magnetization for \( \tilde{\alpha} \gtrsim 0.207 \). In this case the chemical potential, \( \mu \), has to be considered as a quantity which is to be found self-consistently.

In addition to the \( SU(2) \) symmetry of the spin Hamiltonian (1), such a disordered phase preserves all the symmetries of the lattice, i.e. the \( C_3 \) and \( C_6 \) rotational and translational symmetries. In fact all attempts to find a solution with broken rotational symmetry, for example a solution with unequal pairing and hopping functions on different bonds, were unsuccessful. Such a magnetically disordered state which respects all the symmetries of the Hamiltonian and the lattice is called the QSL state. As can be seen from figure 3, the energy curve of the Néel.I state connects smoothly to the QSL state, an indication of a continuous phase transition between these two ground states. Moreover the calculation of the spin gap, illustrated in figure 5, shows the continuous rise of the magnon gap.
in this phase. Interestingly, the stability of the QSL state goes beyond \( \alpha = 0.25 \) and its energy is lower than that of the Néel II phase up to \( \hat{\alpha} \approx 0.397 \) where it crosses the energy curve of Néel II. As a conclusion, the transition between these two phases is of the first order. Figure 5 shows that at this transition point the spin gap drops discontinuously to zero.

Since this gapped QSL phase is obtained by starting from the Néel.I state it possesses all the symmetries of Néel.I; hence we call it QSL.I. Starting from Néel.II and III, it is also possible to find QSL states, albeit with higher energy with respect to QSL.I. Calculation of spin–spin correlations for QSL.I shows the existence of short-range Néel.I type correlations in this phase (see table 1 and figure 9(b)).

Figure 6 represents the magnon dispersion along the symmetry directions in the magnetic Brillouin zone of the Néel.I (panel-(a)), QSL (panel-(b)) and Néel.II (panel-(c)) phases. In panel-(a), the LSW magnon dispersion is also shown to have lower energy with respect to the MSW dispersion, indicating the greater rigidity of the ordered phase as a result of magnon–magnon interaction. In panel-(c) of figure 6 only the lower branch of the magnon dispersion, given by equation (B.6), is plotted.

To close this section, we compare the MSW results with those of some other methods. Figure 7 displays such a comparison; in the top panel of this figure the MSW ground-state energies are co-plotted with similar results obtained by the DMRG [26], variational Monte Carlo (VMC) approaches based on projected fermionic states (VMC + fermionic state) [31], and VMC based on spin wave states (VMC + SW state) [39]. A comparison of the MSW results with those of CCM [23] and Schwinger boson mean field approaches SB1 [28] and SB2 [33] is also illustrated in the bottom panel of this figure.

The top panel clearly shows that the MSW ground-state energies in the three phases lie below the energies obtained by the DMRG and VMC. Specifically, for the disordered region the QSL state proposed by the MSW has lower energy with respect to the plaquette valence bond (PVB) and staggered dimerized (SD) state (both proposed by the DMRG). For \( \hat{\alpha} \gtrsim 0.4 \), the Néel.II state obtained by the MSW has lower energy than the spin state proposed by VMC + SW [39]. VMC based upon fermionic states yields a Néel.I phase for \( 0 \leq \hat{\alpha} \lesssim 0.08 \), a Z2 QSL state for \( 0.08 \lesssim \hat{\alpha} \lesssim 0.3 \) and an SD state for \( \hat{\alpha} \gtrsim 0.3 \) [31].

On the other hand, the bottom panel shows that the MSW results are in very good agreement with the Schwinger boson mean field approach [28, 33] in the Néel.I and QSL phases, but the MSW gives lower energy for these phases with respect to the CCM [23]. For the Néel.II state, although the MSW result agrees well with SB2 and the CCM for \( \hat{\alpha} \gtrsim 0.6 \), the energy lies below the ones obtained by the other two for \( \hat{\alpha} \lesssim 0.6 \). Hence, the transition point from QSL to Néel.II which is \( \hat{\alpha} \approx 0.42 \) for SB2 moved to a smaller value \( \hat{\alpha} \approx 0.396 \) for the MSW. Moreover, by calculation of the PVB and SD susceptibilities, the CCM predicts a PVB state for \( 0.207 < \hat{\alpha} < 0.385 \) and SD ground states for \( 0.385 < \hat{\alpha} < 0.65 \). SB1 [28] also results in SD ordering for \( 0.373 \lesssim \hat{\alpha} \lesssim 0.398 \) with a competitive energy with QSL.I as found by the MSW. SB1 also gives rise to a spiral ground state for \( 0.398 < \hat{\alpha} < 0.5 \) with a higher energy than the Néel.II obtained by the MSW.

Like the MSW, the entangled pair variational ansatz (not shown in figure 7) [34] yields a Néel.I ordered state for \( 0 \leq \hat{\alpha} \lesssim 0.2 \), a Néel.II state for \( 0.4 \lesssim \hat{\alpha} < 1 \) and a symmetry-preserving disordered state for \( 0.2 \leq \hat{\alpha} \lesssim 0.4 \), though with higher energies with respect to the MSW.

### 4. Phase diagram of distorted model

In this section we discuss the phase diagram of the \( S = 1/2 \) distorted model Hamiltonian (1). The MSW phase diagram of the model is presented in figure 8 in the plane of the distortion parameter \( \delta \) and frustration \( \bar{\alpha} \). Like the symmetric model, the magnetically ordered phases in the presence of distortion are found to be the collinear states Néel.I and II.

Figure 8 shows that the maximum stability of the Néel.I state occurs for the isotropic model \( \delta = 0 \). Distortion in both \( c \) positive (\( \delta > 0 \) i.e. \( J_1 > J'_2 \)) and negative (\( \delta < 0 \) i.e. \( J_1 < J'_2 \)) cases makes this phase more fragile against frustration. For \( |\delta| \approx 0.3 \), the Néel.I phase becomes totally unstable for any \( \bar{\alpha} \). The stability region of the Néel.I state versus distortion is in agreement with the results of renormalization group (RG) calculations made on the NLSM presentation of the model [15]. However, the RG–NLSM underestimates the stability range of this phase against frustration, i.e. it finds the maximum stability range \( 0 \leq \bar{\alpha} \lesssim 0.11 \) for the symmetric model (\( \delta = 0 \)).

For the Néel.II phase, while positive distortion (\( \bar{\alpha} > 0 \)) has a destructive effect on the stability of this phase against frustration, negative distortion (\( \bar{\alpha} < 0 \)) extends its stability to a lower value of frustration. Note that in order to make \( J_1 \) and \( J'_2 \) positive, the distortion parameter should be in the interval \([-0.5, 1.0]\).

In addition to these two ordered/disordered phases we find four magnetically distinct disordered phases: (i) a valence bond crystal (VBC) phase for large positive distortion, (ii) a gapped QSL originating from the Néel.I state (gapped QSL.I) for intermediate positive and small negative distortions, (iii) a gapped QSL originating from the Néel.II state (gapped QSL.II) for negative distortions and intermediate frustration and (iv) gapless QSL originating from Néel.II (gapless QSL.I) for large negative distortion and small frustration. In contrast to gapped QSL.II, all the other three disordered phases VBC and gapped and gapless QSL.I are self-consistent solutions of MSW equations started from the Néel.I ordering state, but with vanishing spontaneous magnetization. On the other hand, in the stability region of gapped QSL.II, starting from Néel.II ordering, the self-consistent equations do not converge to any stable solution. However, in this region assuming a Néel.II type ordering, a stable disordered state comes out of MSW equations.

To gain insight into the nature of the disordered states, we calculated the spin–spin correlation for the nearest and next nearest neighbor spins. The correlation data are given in table 1 for a representative point in each phase. These results are also displayed schematically in figure 9.
As evident from the first three rows of table 1 and panels (a)–(c) of figure 9, in QSL.I there are short-range correlations inherited from Néel.I ordering, i.e. nearest neighbor negative correlations.

Figure 6. Magnon energy dispersion $\omega_k$ (in units of $J_1$) along symmetry directions in the magnetic first Brillouin zone (1BZ) for (a) Néel.I, (b) QSL, and (c) Néel.II phases. The insets illustrate the magnetic 1BZ corresponding to each phase. The magnetic 1BZ of the Néel.I state is the same as the 1BZ of the honeycomb lattice as shown by the inset of panel (b). The solid and dashed lines in panel (a) represent the magnon dispersion obtained in LSW approximation.

Figure 7. Comparison of MSW results with (top) density-matrix renormalization group (DMRG) [26], variational Monte Carlo (VMC) based on Jastrow and projected fermionic states (VMC + fermionic state) [31] and spin wave states (VMC + SW state) [39], and (bottom) the coupled cluster method (CCM) [23] and Schwinger boson approach SB1 [28] and SB2 [33]. SD denotes the staggered dimerized state.

Figure 8. MSW phase diagram of $S = 1/2$ distorted $J_1 - J_2$ honeycomb antiferromagnet. QSL.I and II denote the quantum spin liquid states originating from Néel.I and Néel.II, respectively. VBC stands for valence bond crystal state.
Table 1. Spin–spin correlation functions of nearest neighbors \((S_1 \cdot S_{1+\delta_1}), (S_1 \cdot S_{1+\delta_2}), (S_1 \cdot S_{1+\delta_3})\) and next nearest neighbors \((S_1 \cdot S_{1+\delta_{12}}), (S_1 \cdot S_{1+\delta_{13}}), (S_1 \cdot S_{1+\delta_{23}})\) in different phases of distorted honeycomb antiferromagnet.

| \(\delta\) | \(\delta\) | \(S_1 \cdot S_{1+\delta_1}\) | \(S_1 \cdot S_{1+\delta_2}\) | \(S_1 \cdot S_{1+\delta_3}\) | \(S_1 \cdot S_{1+\delta_{12}}\) | \(S_1 \cdot S_{1+\delta_{13}}\) | \(S_1 \cdot S_{1+\delta_{23}}\) | State          |
|------------|------------|----------------|----------------|----------------|----------------|----------------|----------------|--------------|
| 0.02       | -0.4       | -0.0065        | -0.3276        | 0.0282         | 0.0282         | Gapped QSL.I   |                |
| 0.25       | 0          | -0.1720        | -0.1720        | 0.0411         | 0.0411         | Gapped QSL.1 (symmetric) |
| 0.3125     | 0.125      | -0.3217        | -0.05680       | 0.0122         | 0.0122         | Gapped QSL.I   |                |
| 0.425      | 0.350      | -0.3753        | -4.2 \times 10^{-7} | 5.0 \times 10^{-8} | 5.0 \times 10^{-8} | VBC            |                |
| 0.1875     | -0.125     | 3 \times 10^{-11} | -0.2230        | -4.3 \times 10^{-9} | 0.0615         | Gapped QSL.II  |                |

Note: The vectors \(\delta_1, \delta_2, \delta_3, \delta_{12}, \delta_{13}\) and \(\delta_{23}\) are shown in figure 1.

Figure 9. Schematic representation of spin–spin correlations for the first and second neighbors for a representative point in the phases: (a) gapless QSL.I, (b) symmetric gapped QSL.I \((\delta = 0)\), (c) distorted gapped QSL. I, (e) valence bond solid (VBC) and (d) gapped QSL. II. The solid red and dotted blue lines denote negative (AF) and positive (F) correlations. The thickness of the lines is proportional to the correlation magnitude.

(AF) and next nearest neighbor positive (F) correlations. In the absence of distortion \((\delta = 0)\) the correlations are the same in all directions. However, in the presence of distortion, AF correlations are stronger for nearest neighbor bonds with larger exchange coupling (figures 9(a) and (c)).

While for positive and small negative distortion the QSL.I state is gapped, for small frustration and large negative distortions \((-0.5 \leq \delta \leq -0.3)\) this phase is gapless. It can be seen from the first row of table 1 that the AF correlations along \(J\)-bonds are higher than the ones along \(J\)-bonds, by two orders of magnitude; hence if \(J_2\) is small enough, the honeycomb spin system in this case can be considered as a system of weakly coupled chains with coupling \(J'\). Therefore, the fact that the ground state of a \(S = 1/2\) Heisenberg chain is a gapless spin liquid state would be a justification for the QSL.I state in this region being gapless.

For large positive distortions, there are vanishing correlations between the nearest neighbor correlations in the \(\delta_2\) and \(\delta_3\) directions as well as between all the second neighbors. In this case, the spins residing on \(J_1\)-bonds (\(\delta_1\) directions) form strong singlet valence bonds (figure 9(d)). In such a strong dimerized state, singlets are prevented from hopping to the neighboring bonds and are frozen. This is the reason for its being called a valence bond crystal (VBC).

Finally, in the region of stability for gapped QSL.II (negative distortions and moderate frustration) the AF correlation along \(J_1\)-bonds as well as the positive correlations along \(\delta_1\) and \(\delta_2\) correlations is negligible (the last row of tables 1 and 9(c)). In this phase the system can also be considered as effectively decoupled chains with nearest negative and next nearest positive correlations. It seems the enhanced frustrating interaction \(J_2\) between the second neighbors pushes the two spins within each unit cell into their high-spin state; hence, roughly speaking, this spin system can be effectively described by \(S = 1\) chains for which spin excitations are gapped.

5. Conclusion

Taking advantage of DM transformation, which is exact and hence unlike Holstein–Primakoff transformation needs not be truncated, the MSW provides a powerful tool to extract the phase diagram of spin systems. Using the MSW, we explored the ground state of symmetric and distorted \(S = 1/2\) Heisenberg \(J_1 - J_2\) antiferromagnets in a honeycomb lattice. For the symmetric model, where all equivalent bonds in the honeycomb lattice have equal exchange couplings, we found two types of collinear ordering in low and high frustration limits, namely a two-sublattice ordering Néel.I for \(0 \leq J_2/J_1 \lesssim 0.207\) and a four-sublattice ordering Néel.II for \(J_2/J_1 \gtrsim 0.396\). Néel.II is not a classical solution and so is unstable when quantum fluctuations are taken into account by LSW theory. Indeed, for \(S = 1/2\) enhanced nonlinear quantum fluctuations tend to stabilize this phase. For intermediate frustration \(0.207 \lesssim J_2/J_1 \lesssim 0.396\) a magnetically disordered state whose symmetries of the system is found to be the ground state—that is, a gapped QSL. The short-range correlations in this QSL have the symmetries of Néel.I, so we coined the name QSL.I for this phase. We found that these two phases transform to each other by a continuous phase transition. However, the symmetries of QSL.I are different from those of Néel.II, and so a first-order transition is found between these two states as expected. As a conclusion the order–disorder transitions in this system can be described in the framework of Landau–Ginzburg theory. We compared our
results with those obtained with other methods and found very good agreement between MSW and Schwinger boson mean field theory [33], knowing that both benefit from mean field approximation.

Introducing distortion to the model breaks its $C_3$ symmetry. This leads to the emergence of new phases as the result of the interplay between distortion and frustration. These new phases, all magnetically disordered, are a gapless QSL.I originating from Néel.I ordering, a gapped QSL.II originating from Néel.II and a VBS state where singlet dimers are frozen on bonds with larger coupling. We discussed that in both the gapless QSL.I and gapped QSL.II phases, the model can be effectively described in terms of weakly coupled zigzag chains.

The main advantage of the MSW over other methods, such as the DMRG, VMC and ED, is that it is free from the finite size effect. However, the validity of the mean field approximation incorporated in this method might be in question when quantum fluctuations become large. Quantum fluctuations are significantly large in disordered states where spontaneous magnetization, or in terms of bosons the condensate, vanishes. This suggests that the QSL states proposed for the disordered region of the phase diagram have to be considered cautiously. Therefore, to improve the validity of MSW states, one could consider them as the initial wave function for variational methods.

Appendix A. Derivation of MSW self-consistent equations

To diagonalize the Hamiltonian (6) in mean field approximation, we need to define the Bogoliubov transformations

\[
\begin{align*}
\alpha_k &= \cosh(\theta_k)a_k - \sinh(\theta_k)b_k^\dagger, \\
\beta_k^\dagger &= -\sinh(\theta_k)a_k + \cosh(\theta_k)b_k^\dagger,
\end{align*}
\]  

(A.1)

where $a_k$ and $b_k^\dagger$ are the Fourier transformations of $a_i$ and $b_i^\dagger$ (defined by equation (5)),

\[
\begin{align*}
a_k &= \frac{1}{\sqrt{N}} \sum_{j \in A} e^{-i k \cdot r_j} a_j, \\
b_k^\dagger &= \frac{1}{\sqrt{N}} \sum_{j \in B} e^{-i k \cdot r_j} b_j,
\end{align*}
\]  

(A.2)

in which $N$ is the total number of sites. The mean field Hamiltonian in its diagonalized form, in terms of noninteracting Bogoliubov quasiparticles, is written as

\[
H_{\text{MF}} = \sum_k \omega_k (\alpha_k^\dagger \alpha_k + \beta_k^\dagger \beta_k) + N E_0,
\]  

(A.3)

where $\omega_k$ is the excitation energy spectrum and $E_0$ is the ground-state energy per site given by equation (9). Substituting $a_i$ and $b_i$ in equation (8) in terms of Bogolon operators (A.1), for a pair of DM bosons at a given displacement vector $r_{ij} = r_i - r_j$, one finds for hopping ($f_{ij} = f(r_{ij})$) and pairing ($g_{ij} = g(r_{ij})$) the expectation functions

\[
f_{ij} = \frac{1}{N} \sum_{k} \cosh(2\theta_k) \exp(-i k \cdot r_{ij}),
\]  

(A.4)

with $i, j \in A$ or $B$, and

\[
g_{ij} = \frac{1}{N} \sum_{k} \sinh(2\theta_k) \exp(-i k \cdot r_{ij}),
\]  

(A.5)

with $i \in A$ and $j \in B$. Otherwise $f_{ij}$ and $g_{ij}$ vanish. In equations (A.4) and (A.5), $\sum_k$ denotes the sum of over half of the Brillouin zone.

We then minimize the mean field energy (7) with respect to $\theta_k$, under the constraint (11), that is

\[
\frac{\partial [E - \mu f(0)]}{\partial \theta_k} = 0,
\]  

(A.6)

where the Lagrange multiplier $\mu$ can be considered the chemical potential needed to fix the number of DM bosons in order to fulfill Takahashi’s constraint. Minimization (A.6) yields the following set of self-consistent equations

\[
f_{ij} = M_0 + \frac{1}{N} \sum_{k \neq 0} \delta_{ij} e^{ik \cdot r_k},
\]  

(A.7)

\[
g_{ij} = M_0 + \frac{1}{N} \sum_{k \neq 0} \delta_{ij} e^{-ik \cdot r_k},
\]  

(A.8)

and

\[
M_0 = S + \frac{1}{2} - \frac{1}{N} \sum_{k \neq 0} \delta_{ij} e^{ik \cdot r_k}.
\]  

(A.9)

Here $A_k$ and $B_k$ are given by

\[
A_k = \frac{1}{2} \sum_{\delta} J(\delta) g(\delta) e^{i k \cdot \delta} + \frac{J}{2} \sum_{\delta'} g(\delta') e^{i k \cdot \delta'}
\]  

(A.10)

and

\[
B_k = \frac{1}{2} \sum_{\delta} J(\delta)(g(\delta) - f(\delta)(1 - e^{ik \cdot \delta}))
\]  

\[
+ \frac{J}{2} \sum_{\delta'} [g(\delta') - f(\delta')(1 - e^{ik \cdot \delta'})] + \mu.
\]  

(A.11)

In equation (A.9), $M_0$ denotes the spontaneous magnetization. In terms of DM bosons, $M_0$ would be the order parameter of BEC transition; hence the physical meaning of $M_0 = \langle a_{k=0}^\dagger a_{k=0} \rangle / N = \langle b_{k=0}^\dagger b_{k=0} \rangle / N$ is the number of bosons condensed in zero energy. Therefore, the nonzero value of the condensate is an indication of the existence of LRO in the magnetic state of the spin system.

The magnon energy spectrum is also given by

\[
\omega_k = \sqrt{B_k^2 - A_k^2},
\]  

(A.12)

At $k = 0$, we obtain from equations (A.10) and (A.11) that $B_{k=0} = A_{k=0} + \mu$.

For each ordering wave vector $Q$, the spin–spin correlation function can be obtained as
\begin{align}
\langle S_i, S_j \rangle &= \frac{1}{2} \left[ (S + \frac{1}{2} - f(0) + g_{ij})^2 (1 - \cos(Q \cdot r_{ij} + \phi)) 
- (S + \frac{1}{2} - f(0) + f_{ij})^2 (1 + \cos(Q \cdot r_{ij} + \phi)) \right], \\
(\text{A.13})
\end{align}

for \( i \in A \) and \( j \in B \) and

\begin{align}
\langle S_i, S_j \rangle &= \frac{1}{2} \left[ (S + \frac{1}{2} - f(0) + g_{ij})^2 (1 - \cos(Q \cdot r_{ij})) 
- (S + \frac{1}{2} - f(0) + f_{ij})^2 (1 + \cos(Q \cdot r_{ij})) \right]. \\
(\text{A.14})
\end{align}

for \( i \) and \( j \) in \( A \) or \( B \).

**Appendix B. Derivation of self-consistent equations for Néel.II and III states**

For the Néel.II and III states, owing to their four-sublattice magnetic pattern (figures 2(b) and (c)), we define the Bogoliubov transformations as

\begin{align}
\alpha_{i,k} &= \frac{1}{\sqrt{2}} \left( \cosh(\theta_{i,k}^{\prime}) a_k - \sinh(\theta_{i,k}^{\prime}) b_{i,k}^\dagger - \sinh(\theta_{i,k}^{\prime}) c_{i,k}^\dagger \right) \\
+ \cosh(\theta_{i,k}^{\prime}) d_k, \\
(\text{B.1})
\end{align}

\begin{align}
\beta_{i,k} &= \frac{1}{\sqrt{2}} \left( -\sinh(\theta_{i,k}^{\prime}) a_k + \cosh(\theta_{i,k}^{\prime}) b_{i,k}^\dagger + \cosh(\theta_{i,k}^{\prime}) c_{i,k}^\dagger \right) \\
- \sinh(\theta_{i,k}^{\prime}) d_k, \\
\beta_{2,k} &= \frac{1}{\sqrt{2}} \left( -\sinh(\theta_{2,k}^{\prime}) a_k + \cosh(\theta_{2,k}^{\prime}) b_{2,k}^\dagger - \cosh(\theta_{2,k}^{\prime}) c_{2,k}^\dagger \right) \\
+ \sinh(\theta_{2,k}^{\prime}) d_k. \\
(\text{B.2})
\end{align}

Then following a similar approach discussed in appendix A, after minimizing the energy with respect to \( \theta_{i,k}^{\prime} \) and \( \theta_{2,k}^{\prime} \), that is

\begin{align}
\frac{\partial[E - \mu f(0)]}{\partial \theta_{i,k}^{\prime}} &= 0, \\
(\text{B.2})
\end{align}

and defining \( A_{k}^\pm \) and \( B_{k}^\pm \) as

\begin{align}
A_{k}^\pm &= \frac{1}{2} \sum_\delta J(\delta) g(\delta) e^{i k \cdot r_\delta} \pm \frac{f_2}{2} \sum_\delta^* g(\delta^*) e^{i k \cdot r_\delta}, \\
(\text{B.3})
\end{align}

and

\begin{align}
B_{k}^\pm &= \frac{1}{2} \sum_\delta J(\delta) [g(\delta) - f(\delta)(1 \pm e^{i k \cdot r_\delta})] \\
+ \frac{f_2}{2} \sum_\delta^* [g(\delta^*) - f(\delta^*)(1 - e^{i k \cdot r_\delta})] + \mu, \\
(\text{B.4})
\end{align}

one finds the following set of self-consistent equations

\begin{align}
M_0 &= S + \frac{1}{2} \frac{1}{2 N} \sum_{k=0}^{3} \left( \frac{B_{k}^+}{\omega_k} + \frac{B_{k}^-}{\omega_k} \right), \\
f_{ij} &= M_0 + \frac{1}{2 N} \sum_{k=0}^{3} \left( \frac{B_{k}^+}{\omega_k} - \frac{B_{k}^-}{\omega_k} \right) e^{i k \cdot r_{ij}}, \\
\text{for } (i \in A; j \in D) \text{ or } (i \in B; j \in C), \\
f_{ij} &= M_0 + \frac{1}{2 N} \sum_{k=0}^{3} \left( \frac{A_{k}^+}{\omega_k} + \frac{A_{k}^-}{\omega_k} \right) e^{i k \cdot r_{ij}}, \\
\text{for } (i, j) \in A, B, C, \text{ or } D, \\
g_{ij} &= M_0 + \frac{1}{2 N} \sum_{k=0}^{3} \left( \frac{A_{k}^+}{\omega_k} - \frac{A_{k}^-}{\omega_k} \right) e^{i k \cdot r_{ij}}, \\
\text{for } (i \in A; j \in B) \text{ or } (i \in C; j \in D), \\
g_{ij} &= M_0 + \frac{1}{2 N} \sum_{k=0}^{3} \left( \frac{A_{k}^+}{\omega_k} - \frac{A_{k}^-}{\omega_k} \right) e^{i k \cdot r_{ij}}, \\
\text{for } (i \in A; j \in C) \text{ or } (i \in B; j \in D), \\
otherwise \end{align}

otherwise \( f_{ij} = g_{ij} = 0 \).  

Néel.II consists of four sublattices, and then there are two branches of magnon excitations for this phase given by

\begin{align}
\omega_{1,k} &= \sqrt{B_{k}^2 - A_{k}^2}, \\
\omega_{2,k} &= \sqrt{B_{k}^2 - A_{k}^2}. \\
(\text{B.6})
\end{align}

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