Plaquette valence–bond solid in the square lattice $J_1$–$J_2$ antiferromagnet Heisenberg model: a bond operator approach

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We study the plaquette valence–bond solid phase of the spin–1/2 $J_1$–$J_2$ antiferromagnet Heisenberg model on the square lattice within the bond–operator theory. We start by considering four $S = 1/2$ spins on a single plaquette and determine the bond operator representation for the spin operators in terms of singlet, triplet, and quartet boson operators. The formalism is then applied to the $J_1$–$J_2$ model and an effective interacting boson model in terms of singlets and triplets is derived. The effective model is analyzed within the harmonic approximation and the previous results of Zhitomirsky and Ueda [Phys. Rev. B 54, 9007 (1996)] are recovered. By perturbatively including cubic (triplet–triplet–triplet and singlet–triplet–triplet) and quartic interactions, we find that the plaquette valence–bond solid phase is stable within the parameter region $0.34 < J_2/J_1 < 0.59$, which is narrower than the harmonic one. Differently from the harmonic approximation, the excitation gap vanishes at both critical couplings $J_2 = 0.34 J_1$ and $J_2 = 0.59 J_1$. Interestingly, for $J_2 < 0.48 J_1$, the excitation gap corresponds to a single–triplet excitation at the $\Gamma$ point while, for $J_2 > 0.48 J_1$, it is related to a single–singlet excitation at the $X = (\pi/2, 0)$ point of the tetramerized Brillouin zone.

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I. INTRODUCTION

Two–dimensional frustrated quantum antiferromagnets have been receiving a lot of attention in recent years. Here the interplay between frustration (dynamic or geometric) and quantum fluctuations may destroy magnetic long–range order (LRO) yielding to quantum paramagnetic (disordered) phases, such as valence bond solids (VBSs) with broken lattice symmetries or spin liquids, where lattice symmetries are preserved. An interesting example of a frustrated quantum magnet is the spin–1/2 $J_1$–$J_2$ antiferromagnet (AFM) Heisenberg model on the square lattice:

$$\mathcal{H} = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (1)$$

Here $\mathbf{S}_i$ is an spin–1/2 operator at site $i$ and $J_1 > 0$ and $J_2 > 0$ are, respectively, the nearest–neighbor and next–nearest–neighbor exchange couplings as illustrated in Fig. 1(a).

Several different theoretical approaches have been employed to study the $J_1$–$J_2$ model in the last few years. It is now well established that the model has semiclassical Néel magnetic LRO with ordering wave vector $\mathbf{q} = (\pi, \pi)$ for $J_2 \lesssim 0.4 J_1$, collinear magnetic LRO with $\mathbf{q} = (\pi, 0)$ or $(0, \pi)$ for $J_2 \gtrsim 0.6 J_1$, and a quantum paramagnetic (disordered) phase within the intermediate parameter region $0.4 \lesssim J_2/J_1 \lesssim 0.6$. However, the nature of such a disordered phase and the quantum phase transition at small $J_2$ are still under debate. These two issues are mainly associated with the fact that large–scale quantum Monte Carlo simulations can not be used here due to the so–called sign problem.

Different proposals have been made for the ground state of the disordered phase of the $J_1$–$J_2$ model: a columnar VBS [Fig. 2(a)], where both translational and rotational lattice symmetries are broken, and a plaquette VBS [Fig. 1(b)], where only the translational lattice symmetry is broken. A mixed columnar–plaquette VBS and gapless spin–liquids have also been found.

About the quantum phase transitions: while there are strong indications that a first–order quantum phase transition takes place at $J_2 \approx 0.6 J_1$ (the boundary between the quantum paramagnetic and the collinear phases), it is still not clear whether a first–order or a continuous quantum phase transition occurs at $J_2 \approx 0.4 J_1$ (the boundary between the Néel and the quantum paramagnetic phases). If a VBS phase sets in within the magnetic disorder region, the former scenario is in agreement with the Landau–Ginzburg framework (the Néel and the VBS phases are characterized by two different order parameters) while the latter is in favor of the so–called deconfined quantum criticality. A candidate theory for a possible continuous quantum phase transition between a $Z_2$ spin–liquid and a Néel phase is recently proposed in Ref. 33.

We should also mention that, more recently, the $J_1$–$J_2$ model on the honeycomb lattice has also been studied. Here the main motivation are quantum Monte Carlo results for the half–filled honeycomb Hubbard model which provide some evidences for a gapped spin–liquid phase within intermediate values of the on–site repulsion $U$. Density matrix renormalization group (DMRG) calculations have been performed on the honeycomb lattice $J_1$–$J_2$ model and it is found that as $J_2/J_1$ increases, a Néel phase, a plaquette and a dimerized VBS phases set in. Similar results are reported in Ref. 33 where the coupled cluster method is employed.
An useful approach to describe VBS phases of a
Heisenberg model is the bond–operator theory intro-
duced by Sachdev and Bhatt. Such a formalism can
be seen as the analog of the Holstein–Primakoff
representation, but here we consider fluctuations above a
quantum paramagnetic ground state instead of a (semi-
classical) state with magnetic LRO. The formalism de-
voped in Ref. 20 is appropriate to describe dimerized
phases, such as the columnar and (b) staggered valence bond solids. The blue ellipses indicate that the spins \( S^1 \) (open circle), \( S^2 \) (black circle), \( S^3 \) (red circle), and \( S^4 \) (green circle) form

\[ J \]

a singlet state. \( \mathbf{a}_1 \) and \( \mathbf{a}_2 \) are the primitive vectors of the tetramerized lattice. (c) Brillouin zone of the tetramerized square lattice defined by the plaquettes. Here \( X = (\pi/2, 0) \), \( Y = (0, \pi/2) \), and \( M = (\pi/2, \pi/2) \) (the lattice spacing of the original square lattice is set to one).

![FIG. 1.](image1)

**FIG. 1.** (Color online) Schematic representation: (a) \( J_1-J_2 \) AFM Heisenberg model. The dashed blue squares indicate that the spins \( S^1 \) (open circle), \( S^2 \) (black circle), \( S^3 \) (red circle), and \( S^4 \) (green circle) form a singlet state. \( \mathbf{a}_1 \) and \( \mathbf{a}_2 \) are the primitive vectors of the tetramerized lattice. (b) Plaquette valence bond solid. A generalized method suitable for
describing tetramerized phases, such as the plaquette VBS [Fig. 2(b)], was later introduced by Zhitomirsky and
Ueda. However, here only a partial bond–operator rep-
resentation for the spin operators [in terms of the lowest–
energy singlet and the triplet (boson) operators] was con-
sidered: the high–energy singlet and the quintet operators
(see below) were neglected.

In this paper, we revisit the work of Zhitomirsky and
Ueda and study the plaquette VBS phase of the \( J_1-J_2 \) model within the bond–operator theory. We derive
the full bond–operator representation (in terms of sing-
let, triplet, and quintet boson operators) for spin–1/2
operators on a single plaquette and apply such a gener-
alyzed formalism to the \( J_1-J_2 \) model. Our study is not
only restricted to the analysis at the harmonic (mean–
field) level of an effective boson model in terms of the

\[ \text{lowest–energy singlet and the triplet operators as done in Ref. 23 but we also include the high–energy singlet operator and go beyond the harmonic approximation: cubic (singlet–triplet–triplet and triplet–triplet–triplet) and quartic interactions are perturbatively considered. Our main motivations are a series of results concerning a dimerized phase of a trian-
gular lattice Heisenberg AFM, where we show that cubic (triplet–triplet–triplet) interactions have an impor-
tant role in the determination of the excitation spectrum of such a frustrated quantum magnet.} \]

### A. Overview of the results

We calculate the ground state energy [Fig. 3(b)] and
the dispersion relation of the singlet and triplet excitation
(Fig. 3) of the plaquette VBS phase within the (mean–field) harmonic and the cubic–quartic approxima-
tions. In the latter, cubic and quartic interactions are
perturbatively added to the harmonic results. Our main
findings are the following:

(a) Harmonic approximation. The plaquette phase is sta-
ble within the parameter region \( 0.26 < J_2/J_1 < 1.00 \), see Fig. 3(a). The excitation gap [Fig. 7(a)] is always finite
and it is related to a singlet–triplet excitation (triplet
gap) for \( J_2 < 0.82 J_1 \) and a singlet–singlet one (singlet
gap) for \( J_2 > 0.82 J_1 \).

(b) Cubic–quartic approximation. The region of stability

\[ \frac{J_2}{J_1} \]

![FIG. 2.](image2)

**FIG. 2.** (Color online) Schematic representation: (a) Colu-
nar and (b) staggered valence bond solids. The blue ellipses indicate that the spins \( S^1 \) (open circle) and \( S^2 \) (filled circle) form a singlet state. \( \mathbf{a}_1 \) and \( \mathbf{a}_2 \) are the primitive vectors of the dimerized lattices.

![FIG. 3.](image3)

**FIG. 3.** (Color online) Region of stability of the plaquette VBS phase of the \( J_1-J_2 \) model as obtained from bond–
operator theory within (a) the harmonic and (b) the cubic–quartic approximations.
of the plaquette phase is $0.34 < J_2/J_1 < 0.59$ [Fig. 3(b)] with the excitation gap vanishing at both critical couplings $J_2 = 0.34 J_1$ and $J_2 = 0.59 J_1$ [Fig. 7(b)]. For $J_2 > 0.48 J_1$, the excitation gap is no longer associated with a singlet–triplet excitation at the $\Gamma$ point, but with a singlet–singlet one at the $X = (\pi/2, 0)$ point of the tetramerized Brillouin zone [see Fig. 2(c)]. The decay rates of the singlet and triplet excitations are also obtained [see Figs. 5 and 10].

The reader not interested in the technical details may skip Secs. II – V and go straight to Sec. VI.

B. Outline

Our paper is organized as follows: In Sec. II, we generalize the (dimer) bond–operator formalism\(^2^0\) for the case of four spins $S = 1/2$ on a single plaquette. In Sec. III, we apply the generalized bond–operator representation to the $J_1$–$J_2$ model and derive an effective model in terms of singlet and triplet boson operators. Sec. IV is devoted to the analysis of the effective boson model in the harmonic approximation. The ground state energy and the dispersion relations of the singlet and triplet excitations are calculated. In Sec. V, we consider cubic (singlet–triplet–triplet and triplet–triplet–triplet) interactions in second–order perturbation theory and quartic ones in the (no self–consistent) Hartree–Fock approximation and calculate the corrections to the harmonic results (cubic–quartic approximation). We compare our results with previous ones and discuss their implications for the $J_1$–$J_2$ model in Sec. VI. Our findings are summarized in the last section. Some details of the calculations discussed in the main part can be found in the five Appendixes.

II. BOND OPERATOR REPRESENTATION

In Ref. 20, a bond–operator representation for two spins $S = 1/2$ in a dimer is introduced. In this section, we consider the case of four spins $S = 1/2$ in a plaquette and develop a bond–operator representation for the spin operators in terms of singlet, triplet, and quintet (boson) operators. We should mention that such a formalism was already discussed in Refs. 23 and 45 but, in that case, the high–energy singlet state $\vert s_1 \rangle$ and the quintet states $\vert d_0 \rangle$, $\vert d_2 \rangle$, and $\vert d_4 \rangle$ (see below) were not considered. As far as we know, this is the first time that the complete bond–operator representation for spins in a plaquette is derived.

![Diagram](a) Schematic representation of the spin–$1/2$ $J_1$–$J_2$ AFM Heisenberg model (1) on a single plaquette, Eq. (2). (b) Eigenvalues (3) of the Hamiltonian (2) as a function of $J_2/J_1$: $E_{s0}$, $E_{s1}$, $E_{t1,2,3}$, and $E_d$ are, respectively, the energies of the singlet, triplet, and quintet states.

A. Single plaquette

Let us consider the Heisenberg model (1) restricted to four spins in a single plaquette as illustrated in Fig. 4(a):

$$\mathcal{H}_{plaq} = J_1 (S^1_1 + S^3_3) \cdot (S^2_2 + S^4_4) + J_2 (S^1_1 \cdot S^3_3 + S^2_2 \cdot S^4_4).$$

\(2\)

It is easy to show that the eigenvalues of the Hamiltonian (2) are given by

$$E_{s0} = -2J_1 + \frac{1}{2}J_2, \quad E_{s1} = -\frac{3}{2}J_2, \quad E_{t1} = -\frac{1}{2}J_2, \quad E_{t2} = -J_1 + \frac{1}{2}J_2, \quad E_{t3} = -J_1 + \frac{1}{2}J_2, \quad E_d = J_1 + \frac{1}{2}J_2.$$

\(3\)

The behaviour of the spectrum as a function of $J_2/J_1$ is shown in Fig. 4(b). For $J_2 < J_1$, the ground state is given by the singlet state $\vert s_0 \rangle$ whose energy is $E_{s0}$. There are four excited energy levels: $E_{s1}$ is the eigenvalue related to the singlet state $\vert s_1 \rangle$, $E_{t11} = E_{t12}$ is the energy of the six triplet states $\vert t_{1,1} \rangle$ and $\vert t_{1,2} \rangle$ with $\alpha = x, y, z$ while $E_{t13}$ is the energy of the three triplet states $\vert t_{3,0} \rangle$. Finally, $E_d$ is the eigenvalue associated with the five quintet states $\vert d_0 \rangle$, $\vert d_2 \rangle$, and $\vert d_4 \rangle$. Note that the excitation gap is associated with a singlet–triplet transition (triplet gap) for $J_2 < 0.5J_1$ and with a singlet–singlet one (singlet gap) for $J_2 > 0.5J_1$. We refer the reader to Appendix A for the explicit expressions of the singlet, triplet and quintet states in terms of the 16 states $\vert \uparrow \uparrow \uparrow \uparrow \rangle$, $\vert \downarrow \downarrow \uparrow \uparrow \rangle$, $\vert \uparrow \downarrow \uparrow \uparrow \rangle$, \ldots $$.
B. Boson operators

As discussed in the previous section, the Hilbert space of four spins \( S = 1/2 \) (\( S^1 \), \( S^2 \), \( S^3 \), and \( S^4 \)) in a single plaquette is made out of 16 states: two singlet, nine triplet, and five quintet states. We can introduce a set of boson operators which creates these states out of a fictitious and five quintet states. We can introduce a set of boson operators which creates these states out of a fictitious vacuum \( |0\rangle \), namely,

\[
|s_0\rangle = s_0^\dagger|0\rangle, \quad |s_1\rangle = s_1^\dagger|0\rangle, \quad |t_{a,\alpha}\rangle = t_{a,\alpha}^\dagger|0\rangle,
\]

\[
|d_0\rangle = d_0^\dagger|0\rangle, \quad |d_2\rangle = d_2^\dagger|0\rangle, \quad |d_3\rangle = d_3^\dagger|0\rangle,
\]

with \( a = 1, 2, 3 \) and \( \alpha = x, y, z \). In order to remove unphysical states from the enlarged Hilbert space, the Hamiltonian (2) assumes the form

\[
H_{\text{plaq}} = E_{s_0} s_0^\dagger s_0 + E_{s_1} s_1^\dagger s_1 + E_{t_{a,\alpha}} \sum_{a=1,2} t_{a,\alpha}^\dagger t_{a,\alpha} + E_d \left( d_0^\dagger d_0 + d_2^\dagger d_2 + d_3^\dagger d_3 \right). \quad (7)
\]

Since the bond operator representation (6) is quite involved, it is useful to consider an approximate expansion for the spin operators \( S^\mu \). In particular, neglecting the high-energy quintet states, Eq. (6) reduces to

\[
S^\mu_\alpha = C^a_\mu (t_{a,\alpha}^\dagger s_0 + s_0^\dagger t_{a,\alpha}) + \tilde{C}^a_\mu (t_{a,\alpha}^\dagger s_1 + s_1^\dagger t_{a,\alpha}) - i \epsilon^{\alpha\beta\gamma} D^{a\mu}_{\beta\gamma} d_\beta^\dagger t_{a,\gamma}, \quad (8)
\]

where \( \mu = 1, 2, 3, 4 \) and the coefficients \( C^a_\mu \), \( \tilde{C}^a_\mu \), and \( D^{a\mu}_{\beta\gamma} \) are given by

\[
C^1_{1/3} = C^2_{2/3} = \pm 1/2\sqrt{3}, \quad C^3_{1/3} = -C^4_{2/3} = 1/\sqrt{6},
\]

\[
\tilde{C}^1_{2/3} = C^2_{1/3} = \pm 1/2,
\]

\[
D^{1/3}_{1/3} = D^{2/3}_{2/3} = 1/2, \quad D^{3/4}_{3/4} = D^{1/3}_{1/3} = 1/4,
\]

\[
D^{1/3}_{1/3} = D^{2/3}_{2/3} = -D^{1/3}_{2/3} = -D^{2/3}_{1/3} = \pm 1/2\sqrt{2}, \quad (9)
\]

and zero otherwise. Eq. (8) is quite similar to the bond operator representation for two spins \( S = 1/2 \) in a dimer, see e.g., Eqs. (2.2) and (2.3) from Ref. 20.

The bond operator representation (6) can be generalized to the lattice case and the corresponding Heisenberg model can be expressed in terms of the boson operators \( s_0^\dagger, s_1^\dagger, t_{a,\alpha}^\dagger, d_0^\dagger, d_2^\dagger, d_3^\dagger, \) and \( d_{a,\alpha}^\dagger \).

III. EFFECTIVE BOSON MODEL

In this section, we apply the bond operator formalism developed above to study the plaquette VBS phase of the \( J_1-J_2 \) model. The idea is to map the Heisenberg model (1) into an effective boson model in terms of the singlet \( s_{1,i} \) and the triplet \( t_{a,i,\alpha} \) operators.

We start by rewriting the Hamiltonian (1) in terms of the underline (tetramerized) square lattice defined by the plaquettes as shown in Fig. 1(b):

\[
H = \sum_i J_1 (S^1_i \cdot S^1_{i+1}) + J_2 (S^3_i \cdot S^3_{i+1}) + J_4 (S^4_i \cdot S^4_{i+1} + S^5_i \cdot S^5_{i+1})
\]

\[
+ J_2 (S^2_i \cdot S^2_{i+1} + S^3_i \cdot S^3_{i+1} + S^4_i \cdot S^4_{i+1} + S^5_i \cdot S^5_{i+1}) + J_4 (S^2_i \cdot S^2_{i+1} + S^3_i \cdot S^3_{i+1} + S^4_i \cdot S^4_{i+1} + S^5_i \cdot S^5_{i+1} + S^1_i \cdot S^1_{i+1})
\]

\[
+ J_6 (S^3_i \cdot S^3_{i+1} + S^4_i \cdot S^4_{i+1} + S^5_i \cdot S^5_{i+1} + S^1_i \cdot S^1_{i+1}). \quad (10)
\]

Here, the numbers 1 and 2 in the site indices \( i + 1, \)
\[ i + 2, \ldots, \text{etc respectively indicates the nearest-neighbor vectors} \]
\[ \tau_1 = 2a\hat{x} \quad \text{and} \quad \tau_2 = 2a\hat{y} \qquad (11) \]

with \( a \) being the lattice spacing of the original square lattice (in the following we set \( a = 1 \)). Note that the unit cell of the underline square lattice has four spins: \( S^1, S^2, S^3, \) and \( S^4 \). We then substitute Eq. \( \text{[5]} \) generalized to the lattice case into Eq. \( \text{(10)} \), i.e., we consider the approximate bond–operator representation where the high–energy quintets are neglected, and, after some algebra, find that the Hamiltonian assumes the general form:

\[ H = E_0 + H_{02} + H_{20} + H_{30} + H_{40} + H_{21} + H_{22}. \quad (12) \]

Here \( E_0 \) is a constant,

\[ E_0 = \frac{1}{4} NJ_0E_{s0} - \mu(N_0 - 1), \]

the terms \( H_{uuu} \) contain \( \mu \) triplet \( t_{a,i,a} \) and \( m \) singlet \( s_{1,i} \) operators, and the constraint \( \text{[5]} \) is taking into account by adding to the Hamiltonian \( \text{[12]} \) the term

\[ -\mu \sum_i \left( s_{0,i}^\dagger s_{0,i} + s_{1,i}^\dagger s_{1,i} + t_{a,i,a}^\dagger t_{a,i,a} - 1 \right) \]

with \( \mu \) being a Lagrange multiplier.

Within the bond operator formalism, the plaquette VBS state shown in Fig. \( \text{[1b]} \) can be seen as a condensate of the lowest–energy singlets \( s_{0,i} \). In order to implement such a (reference) state, we replace

\[ s_{0,i}^\dagger = s_{0,i} \quad (s_{0,i}^\dagger \leftrightarrow s_{0,i}) \rightarrow \sqrt{N_0} \]

in Eq. \( \text{[12]} \). We then end up with an effective Hamiltonian solely in terms of the triplet \( t_{a,i,a} \) and the singlet \( s_{1,i}^\dagger \) boson operators. Both \( \mu \) and \( N_0 \) will be self-consistently determined later.

Finally, performing a Fourier transform, i.e.,

\[ t_{a,i,a}^\dagger = N'^{-1/2} \sum_k \exp(-i\mathbf{k} \cdot \mathbf{R}_i) t_{a,k,a}, \]

\[ s_{1,i}^\dagger = N'^{-1/2} \sum_k \exp(-i\mathbf{k} \cdot \mathbf{R}_i) s_{1,k}, \]

where \( N' = N/4 \) with \( N \) being the number of sites of the original square lattice and the momentum sums run over the tetramerized Brillouin zone [Fig. \( \text{[1c]} \)], we find that in momentum space the \( H_{uuu} \) terms in Eq. \( \text{[12]} \) read

\[ H_{02} = \sum_k (E_{s1} - \mu) s_{1,k}^\dagger s_{1,k}, \quad (13) \]

\[ H_{20} = \sum_k \sum_{a,b} t_{a,k,a}^\dagger t_{b,k,a} + \frac{B_{ab}}{2} \left( t_{a,k,a}^\dagger t_{b,k,a}^\dagger + \text{H.c.} \right), \quad (14) \]

\[ H_{30} = \frac{\epsilon^{\alpha\beta\gamma}}{\sqrt{N'}} \sum_{p,k} \sum_{a,b} \sum_{c,d} \sum_{\tau,\nu} \sum_{\chi,\lambda} \lambda^{abcd} \chi^{\alpha\beta\gamma} \chi^{\alpha\beta\gamma} \left[ t_{a,p+k,a}^\dagger t_{b,p}^\dagger t_{c,p}^\dagger t_{d,p}^\dagger + \text{H.c.} \right], \quad (15) \]

\[ H_{40} = \frac{\epsilon^{\alpha\beta\gamma}}{\sqrt{N'}} \sum_{p,k} \sum_{a,b} \sum_{c,d} \sum_{\tau,\nu} \sum_{\chi,\lambda} \lambda^{abcd} \chi^{\alpha\beta\gamma} \chi^{\alpha\beta\gamma} \left[ t_{a,p+k,a}^\dagger t_{b,p}^\dagger t_{c,p}^\dagger t_{d,p}^\dagger + \text{H.c.} \right], \quad (16) \]

\[ H_{21} = \frac{1}{\sqrt{N'}} \sum_{p,k} \left[ \left[ \epsilon^{ab} t_{a,p-k,a}^\dagger t_{b,p-k,a}^\dagger s_{1,k} + \text{H.c.} \right] + \lambda^{ab} s_{1,k}^\dagger t_{a,p-k,a}^\dagger t_{b,p-k,a}^\dagger \right], \quad (17) \]

\[ H_{22} = \frac{1}{N'} \sum_{p,q,k} \left[ \epsilon^{ab} t_{a,q-k,a}^\dagger t_{b,p-k,a}^\dagger s_{1,k} + \text{H.c.} \right] + \frac{1}{2} \chi^{ab} s_{1,q-k,a}^\dagger s_{1,p-k,a}^\dagger t_{a,p-k,a}^\dagger t_{b,q-k,a}^\dagger + \text{H.c.} \right], \quad (18) \]
with \(a, b, c = 1, 2, 3\) and \(\alpha, \beta, \gamma = x, y, z\). The coefficients \(A_k^{ab}, B_k^{ab}, \xi_k^{abc}, \chi_k^{abcd}, \xi_k^{ba}, \) and \(\chi_k^{ab}\) can be found in Appendix B.

**IV. HARMONIC APPROXIMATION**

Let us now study the effective boson model (12) in the lowest-order approximation, the so-called harmonic approximation. In this case, we neglect \(H_{30}, H_{40}, H_{21},\) and \(H_{22}\) and consider

\[
H \approx E_0 + H_{02} + H_{20}. \tag{19}
\]

Note that the Hamiltonian (19) is quadratic in the singlet \(s_{1,k}\) and the triplet \(t_{a,k,\alpha}\) boson operators. Moreover, the singlet sector is already diagonalized and decoupled from the triplet one.

In order to diagonalize the triplet sector \(H_{20}\), it is useful to introduce the six-component vector

\[
\Psi_{k\alpha} = (t_{1,k,\alpha}^\dagger, t_{2,k,\alpha}^\dagger, t_{3,k,\alpha}^\dagger, t_{1,-k,\alpha}, t_{2,-k,\alpha}, t_{3,-k,\alpha})
\]

which allow us to rewrite Eq. (19) in matrix form:

\[
H = E_0' + H_{02} + \frac{1}{2} \sum_k \Psi_{k\alpha}^\dagger \hat{H}_k \Psi_{k\alpha}. \tag{20}
\]

Here

\[
E_0' = E_0 - \frac{3}{2} \sum_{a=1,2,3} \sum_k A_k^{aa}
\]

and the \(6 \times 6\) matrix \(\hat{H}_k\) reads

\[
\hat{H}_k = \begin{pmatrix} \hat{A}_k & \hat{B}_k \\ \hat{B}_k & \hat{A}_k \end{pmatrix} \tag{21}
\]

with \(\hat{A}_k\) and \(\hat{B}_k\) being \(3 \times 3\) Hermitian matrices whose elements are \(A_k^{ab}\) and \(B_k^{ab}\) respectively. Although the diagonalization of the \(6 \times 6\) problem is quite involved (we briefly outline the analytical procedure in Appendix C), it is possible to show that, after the diagonalization, Eq. (20) acquires the form

\[
H = E_{EGS} + H_{02} + \frac{1}{2} \sum_k \Phi_{k\alpha}^\dagger \hat{H}'_k \Phi_{k\alpha}. \tag{22}
\]

where

\[
E_{EGS} = E_0 + \frac{3}{2} \sum_{a,k} (\Omega_{a,k} - A_k^{aa}) \tag{23}
\]

is the ground state energy, the \(6 \times 6\) matrix \(\hat{H}'_k\) reads

\[
\hat{H}'_k = \begin{pmatrix} \hat{h}_k & 0 \\ 0 & \hat{h}_k \end{pmatrix}
\]

with \(\hat{h}_k\) being

\[
\begin{pmatrix} \Omega_{1,k} & 0 & 0 \\ 0 & \Omega_{2,k} & 0 \\ 0 & 0 & \Omega_{3,k} \end{pmatrix}
\]
and the six–component vector $\Phi_{\mathbf{k}a}^\dagger$ is given by
\[
\Phi_{\mathbf{k}a}^\dagger = \left( b_{1,\mathbf{k},\alpha}^\dagger, b_{3,\mathbf{k},\alpha}^\dagger, b_{1,-\mathbf{k},\alpha}, b_{2,-\mathbf{k},\alpha}, b_{3,-\mathbf{k},\alpha} \right).
\]
The relation between the two set of boson operators $t$ and $b$ is
\[
\Phi_{\mathbf{k}a} = \hat{M}_k \Psi_{\mathbf{k}a}, \quad \text{where} \quad \hat{M}_k = \left( \begin{array}{cc} \hat{U}_k & \hat{V}_k \\ -\hat{V}_k & \hat{U}_k \end{array} \right)
\]
with $\hat{U}_k$ and $\hat{V}_k$ being $3 \times 3$ matrices whose elements are the Bogoliubov coefficients $u_{ab}^k$ and $v_{ab}^k$. The explicitly expressions of the triplet excitation energies $\Omega_{a,k}$ and the the Bogoliubov coefficients $u_{ab}^k$ and $v_{ab}^k$ in terms of the $A_{2ab}^k$ and $B_{2ab}^k$ functions can be found in Appendix C.

Finally, from the saddle points conditions $\partial E_0/\partial N_0 = 0$ and $\partial E_0/\partial \mu = 0$, self–consistent equations for $\mu$ and $N_0$ follow, namely
\[
\mu = -2J_1 + \frac{1}{2}J_2 + \frac{3}{2N^2} \sum_{a,k} \left[ \frac{\partial \Omega_{a,k}}{\partial N_0} - \frac{1}{N_0} B_{a0}^k \right],
\]
\[
N_0 = 1 + \frac{3}{2N^2} \sum_{a,k} \left[ 1 + \frac{\partial \Omega_{a,k}}{\partial \mu} \right].
\]
(25)

Once $\mu$ and $N_0$ are numerically calculated, the triplet $\Omega_{a,k}$ and the singlet $\Omega_s = E_{s1} - \mu$ excitation energies are completely determined.

We numerically solve the self–consistent equations (25) and find solutions within the range $0.26 < J_2/J_1 < 1.0$ as indicated in Fig. 3(a). The behaviour of the parameters $N_0$ and $\mu$ and the ground state energy (23) as a function of $J_2/J_1$ are respectively shown in Figs. 3(a) and (b). One sees that $N_0$ has a maximum at $J_2 = 0.58 J_1$ and that $E_{\text{EGS}}$ monotonically increases with $J_2/J_1$. For comparison, we include the ground state energy of the dimerized columnar [Fig. 2(a)] and staggered [Fig. 2(b)] VBS phases as obtained from the (dimer) bond–operator theory at the harmonic level (see Appendix D for details). Note that the plaquette VBS state is the lowest–energy one and that it extends over a region of the parameter space much larger than the dimerized VBSs: the columnar VBS is stable for $0.38 < J_2/J_1 < 0.57$ while the staggered VBS only for $0.44 < J_2/J_1 < 0.56$.

Figures 3(a) and (b) shows the energy of the triplet $\Omega_{a,k}$ (solid and dotted–dashed lines) and the singlet $\Omega_s$ (dashed line) excitations for $J_2 = 0.48$ and 0.56 $J_1$, respectively. Recall that $\Omega_s$ is dispersionless in the harmonic approximation. One sees that for $J_2 = 0.48 J_1$, the minimum (gap) of the triplet dispersion relation occurs at the center of the tetramerized Brillouin zone $[\Gamma$ point, see Fig. 4(c)] while for $J_2 = 0.56 J_1$, at the X point. As shown in Fig. 7(a), such a changing in the momentum associated with the excitation gap takes place at $J_2 = 0.50 J_1$. Interestingly, the gap changes from a triplet gap to a singlet one at $J_2 = 0.82 J_1$. Finally, note that the excitation gap is always finite within the parameter region $0.26 < J_2/J_1 < 1.0$, i.e., there is no indication of a continuous quantum phase transition at any critical coupling $J_2$.

V. CUBIC–QUARTIC APPROXIMATION

Since the energy of the singlet $s_1$ and the triplet $t_1$ excitations are quite close for $J_2 \approx 0.5 J_1$ [see Figs. 3(a) and (b)], it is interesting to consider the effects of the cubic interaction $\mathcal{H}_{21}$ [Eq. (14)]. Moreover, we have recently shown that cubic (triplet–triplet–triplet) interactions provide important renormalizations to the harmonic (mean–field) excitation spectrum of a dimerized VBS phase in a frustrated quantum magnet. Motivated by these two points, in this section we consider both cubic terms $\mathcal{H}_{30}$ and $\mathcal{H}_{21}$ with second–order perturbation theory and calculate the corrections to the harmonic re-

FIG. 7. (Color online) The excitation gap as a function of $J_2/J_1$ at (a) the harmonic and (b) the cubic–quartic approximations. $\Omega_{\Gamma,\Gamma}/\Omega_{1,\Gamma}$ (thick solid black line) and $\Omega_{1,X}/\Omega_{1,\Gamma}$ (dashed red line) are respectively the energy of the lowest–energy triplet excitation at the $\Gamma = (0,0)$ and $X = (\pi/2,0)$ points, see Figs. 2(a and b). $\Omega_{\Gamma,\Gamma}/\Omega_{1,\Gamma}$ (thick–dashed line) and $\Omega_{1,X}/\Omega_{1,\Gamma}$ (dotted–dashed line) are respectively the energy of the singlet excitations at the $\Gamma$ and $X$ points.
sults determined in the previous section. We also consider the quartic terms \( \mathcal{H}_{30} \) [Eq. (10)] and \( \mathcal{H}_{21} \) [Eq. (18)] within the (no self-consistent) Hartree–Fock approximation. Although the quartic terms provide very small corrections to the harmonic results, they are important in the determination of the critical couplings. Such a procedure constitutes the so-called cubic–quartic approximation.

The first step is to express \( \mathcal{H}_{30} \) and \( \mathcal{H}_{21} \) in terms of the bosons \( b \). With the help of Eq. (24), it is possible to show that

\[
\mathcal{H}_{30} = \frac{1}{\sqrt{N^3}} \sum_{k,p} \left[ \sum_{\alpha,\beta,\gamma} \Gamma^{abc}_{1,k,p} \left( \beta^{\dagger}_{\alpha,k-p,\alpha} \beta^{\dagger}_{\beta,p,\beta} \beta_{\gamma,k,\gamma} + \text{H.c.} \right) \right]
\]

and

\[
\mathcal{H}_{21} = \frac{1}{\sqrt{N^3}} \sum_{k,p} \left[ \Gamma^{ab}_{1s,k,p} \beta^{\dagger}_{\alpha,k-p,\alpha} \beta_{\beta,p,\beta} s_{1,k} + \text{H.c.} \right]
\]

\[
+ \Gamma^{ab}_{3s,k,p} \beta^{\dagger}_{\alpha,k-p,\alpha} \beta_{\beta,p,\beta} s_{1,k} + \text{H.c.} \right]
\]

\[
+ \Gamma^{ab}_{4s,k,p} \beta^{\dagger}_{\alpha,k-p,\alpha} \beta_{\beta,p,\beta} s_{1,k} + \text{H.c.} \right] .
\]

Here \( a, b, c = 1, 2, 3 \) (summation over repeated indices is assumed), the sum over \( \alpha, \beta, \gamma \) has only three terms, \((\alpha, \beta, \gamma) = (x, y, z), (z, x, y), (y, z, x)\), and the expressions of the renormalized cubic vertices \( \Gamma^{abc}_{1s,k,p} \) and \( \Gamma^{ab}_{1s/3s/4s,k,p} \) [see Fig. 8(a)] in terms of the Bogoliubov coefficients \( u^a_k \) and \( v^b_k \) are given in Appendix E.

Figures (b) and (c) show the lowest-order diagrams that contribute to the (normal) triplet self-energies \( \Sigma_a(k, \omega) \) with \( a = 1, 2, 3 \), while Fig. 8(d) shows the ones related to the singlet self-energy \( \Sigma_s(k, \omega) \). The solid line in each diagram corresponds to the bare (harmonic) \( b \) triplet propagator,

\[
G_{0,a}^{-1}(k, \omega) = \omega - \Omega_{a,k} + i\delta,
\]
and the dashed line denotes the bare $s_1$ singlet propagator,

$$G_{0,s,k}^{-1}(\omega) = \omega - \Omega_s + i\delta,$$

(29)

with $\Omega_s = E_{s1} - \mu$. Hereafter, we omit the $a$ index in the triplet Green’s functions and self-energies since the $x, y, z$ triplet branches for each $a$ are degenerate.

Note that there are no bare anomalous $b$ propagators. Although they can be generated in perturbation theory, we neglect them in the following (for details, see note 40 from Ref. 41). Using standard diagrammatic techniques for bosons at zero temperature, we find that only the diagrams (b1), (b4), (c1), (c3), (d2), and (d3) shown in Fig. 8 are finite, and therefore,

$$\Sigma_a(k, \omega) = \Sigma_a^{(b1)}(k, \omega) + \Sigma_a^{(b4)}(k, \omega)$$

$$+ \Sigma_a^{(c1)}(k, \omega) + \Sigma_a^{(c3)}(k, \omega),$$

(30)

and

$$\Sigma_s(k, \omega) = \Sigma_s^{(d2)}(k, \omega) + \Sigma_s^{(d3)}(k, \omega).$$

(31)

The expressions of the different components of the self-energies (30) and (31) are shown in Appendix E.

Turning to the quartic terms $H_{40}$ and $H_{22}$, it is possible to show that

$$H_{40} = E_{EGS}^{(4)} + \sum_{a,b,\alpha} \sum_{p} A_{ab,p}^{HF} b_{a,p,\alpha}^\dagger b_{b,p,\alpha}$$

$$+ \left[ B_{ab,p}^{HF} b_{a,p,\alpha}^\dagger b_{b,-p,\alpha} + \text{H.c.} \right] + \mathcal{O}(b^4)$$

(32)

and

$$H_{22} \approx \sum_{p} A_{s,p}^{HF} s_{1,p}^\dagger s_{1,p} + \frac{1}{2} \left( B_{s,p}^{HF} s_{1,-p}^\dagger s_{1,p} + \text{H.c.} \right),$$

(33)

where the constant $E_{EGS}^{(4)}$ and the coefficients $A_{ab,s,p}^{HF}$ and $B_{ab,s,p}^{HF}$ can be found in Appendix E. Note that Eq. (32) is not diagonal in the $a$ and $b$ indices. The (normal) triplet and singlet self-energies are then respectively given by

$$\Sigma_{ab}^{HF}(k) = A_{ab,p}^{HF}, \quad \Sigma_s^{HF}(k) = A_{s,p}^{HF},$$

(34)

The renormalized singlet $\bar{\Omega}_{s,k}$ and triplet $\bar{\Omega}_{a,k}$ excitation energies and the decay rates $\bar{\Gamma}_{s/a,k}$ are given by the poles of the corresponding Green’s function $G_{s/a}(k, \omega)$:

$$G_{s/a}^{-1}(k, \omega) = \omega - \bar{\Omega}_{s,a} - \Sigma_{s/a}(k, \omega) - \Sigma_{s/a}^{HF}(k) = 0.$$

(35)

Note that in addition to the anomalous Hartree-Fock self-energies, the normal ones with $\alpha \neq b$ are also neglected, since it significantly simplifies the determination of the poles of the Green’s function. The above equation is solved within the on-shell approximation, where the self-energy is evaluated at the bare (harmonic) single-particle energy:

$$\bar{\Omega}_{s/a} = -i \bar{\Gamma}_{s/a} - \bar{\Omega}_{s/a} - \Sigma_{s/a}(k, \Omega_{s/a}) - \Sigma_{s/a}^{HF}(k) = 0.$$
Such a procedure, which is less involved than the off–shell approximation adopted in Ref. 44, provides reasonable results for the excitation spectra (see below) without the discontinuities and logarithmic singularities reported in Ref. 45.

Finally, the ground state energy reads

\[ E_{\text{EGS}} = E_{\text{EGS}} + E_{\text{EGS}}^{(3)} + E_{\text{EGS}}^{(4)} , \] (35)

where \( E_{\text{EGS}} \) is the harmonic term \(^{23}\) and the expressions of the corrections due to cubic \( E_{\text{EGS}}^{(3)} \) and quartic \( E_{\text{EGS}}^{(4)} \) interactions are presented in Appendix E.

The renormalized singlet \( \Omega_{\alpha,k} \) and triplet \( \Omega_{\alpha,k} \) excitation spectra for \( J_2 = 0.48 \) and 0.56 \( J_1 \) are respectively shown in Figs. 9(a) and (b) while the corresponding decay rates \( \Gamma_{s/a,k} \), in Figs. 9(a) and (b). One sees that the excitation energies decrease as compared to the harmonic ones, an effect similar to what we have found in the triangular lattice quantum magnet.\(^{11}\) In particular, the singlet excitation branch, which now acquires a dispersion, is the lowest–energy excitation for both configurations. We find that the contributions of \( \Sigma_{\alpha}^{(3)} (k, \omega) \) and \( \Sigma_{\alpha}^{(4)} (k, \omega) \) to the renormalized triplet spectra \( \Omega_{s,k} \) are much larger than the ones associated with \( \Sigma_{\alpha}^{(1)} (k, \omega) \) and \( \Sigma_{\alpha}^{(3)} (k, \omega) \). Moreover, we also find that the renormalizations due to the cubic vertices are stronger than the ones associated with the quartic interactions. The most important contributions of \( \Sigma_{HF} (k, \omega) \) to the triplet excitation spectra occurs around \( J_2 = 0.30 J_1 \).

The behaviour of the excitation gap as a function of \( J_2/J_1 \) is shown in Fig. 7(b). Note that the gap vanishes at the critical couplings \( J_2 = 0.34 \) and 0.59 \( J_1 \), indicating that the plaquette VBS phase is stable only within the parameter region \( 0.34 < J_2/J_1 < 0.59 \). Such a result sharply contrasts with the ones obtained within the harmonic approximation [see Figs. 3(a) and (b)]. Moreover, as \( J_2/J_1 \) increases, the excitation gap changes from a triplet gap to a singlet one: for \( 0.34 < J_2/J_1 < 0.48 \), the gap is associated with a singlet–triplet excitation at the \( \Gamma \) point while, for \( 0.48 < J_2/J_1 < 0.59 \), with a singlet–singlet excitation at \( \mathbf{k} = (\pi/2, 0) \).

In addition to renormalize downward the excitation energies, the cubic vertices may also enable two–particle decay of the singlet and triplet modes [Figs. 9(a) and (b)]. In particular, note that for \( J_2 = 0.48 \) and 0.56 \( J_1 \), the triplet decay rate \( \Gamma_{1,1} \approx 0 \) while the singlet one \( \Gamma_{s,X} \) is finite. Indeed, while the former is constant, the latter has an almost monotonous behaviour, decreasing with \( J_2/J_1 \), see Fig. 10. Such a result indicates that the excitation gap acquires a finite decay rate for \( J_2 > 0.48 J_1 \), that decreases and (almost) vanishes close to the critical coupling \( J_2 = 0.59 J_1 \).\(^{34}\)

Finally, we should note that cubic and quartic vertices provide very small corrections to the harmonic ground state energy, see Fig. 9(b).

VI. DISCUSSION

According to the harmonic bond–operator theory (Sec. IV), the plaquette VBS phase has lower energy than the dimerized columnar [Fig. 2(a)] and staggered [Fig. 2(b)] ones. Moreover, the ground state energy of the plaquette phase monotonically increases with \( J_2/J_1 \) while, for the dimerized phases, \( E_{\text{EGS}} \) is a convex function with a minimum around \( J_2 = 0.5 J_1 \). The behaviour of the plaquette ground state energy qualitatively agrees with exact diagonalization data, which show that \( E_{\text{EGS}} \) monotonically increases with \( J_2/J_1 \), reaches a maximum around \( J_2 = 0.6 J_1 \), and then decreases.\(^{39,33,37}\) Such an agreement could be seen as a further indication that the plaquette phase might set in within the disordered region of the \( J_1–J_2 \) model. A similar behaviour for the ground state energy is also observed in coupled cluster\(^{10}\) and tensor network states\(^{25}\) calculations.

As mentioned in the Introduction (Sec. I), Zhitomirsky and Ueda\(^{23}\) studied the plaquette VBS phase of the \( J_1–J_2 \) model within the bond–operator theory at the harmonic level without including the high–energy singlet state \( |s\rangle \). They found that the plaquette phase is stable for \( 0.08 < J_2/J_1 < 0.80 \) and that it has lower energy than the dimerized columnar VBS. In particular, for \( J_2 = 0.50 J_1 \), they found that the excitation gap \( \Delta = 0.85 J_1 \) while the ground state energy \( E_{\text{EGS}} = -0.466 J_1 \). Although the region of stability of the plaquette phase that we arrive at [see Fig. 2(a)] differs from their results, both harmonic (mean–field) calculations show that the plaquette VBS phase extends over a region much larger than the \( J_1–J_2 \) model paramagnetic one \((0.4 \lesssim J_2/J_1 \lesssim 0.6, \text{ see Sec. I})\). Our mean–field results are in reasonable agreement with Ref. 23. Recall that we also find that the plaquette VBS state is more stable than the dimerized columnar state [Fig. 5(b)]. Moreover, for \( J_2 = 0.50 J_1 \), the gap \( \Delta = 0.89 J_1 \) and the ground state energy \( E_{\text{EGS}} = -0.472 J_1 \).

As described in Sec. IV, cubic VBS phase is strongly modified by the harmonic singlet and triplet excitation spectra of the \( J_1–J_2 \) model, similar to what we have recently found for a triangular lattice AFM.\(^{11}\) One important consequence is that the region of stability of the plaquette VBS phase \((0.34 < J_2/J_1 < 0.59)\) is reduced as compared with the harmonic one [see Figs. 3(a) and (b)] and it is now quite close to the disordered region of the \( J_1–J_2 \) model found in previous calculations, see Sec. I. Such a result shows that cubic and quartic interactions are indeed relevant for a proper description of the plaquette VBS phase within the bond operator approach. We should note that although the cubic corrections to the harmonic results are much larger than the quartic ones, the latter has an important role in the determination of the lower critical coupling: including only the cubic vertices, we find that the region of stability of the plaquette phase is \( 0.29 < J_2/J_1 < 0.59 \).

Although the region of stability derived within the
cubic–quartic approximation almost agrees with the paramagnetic region of the $J_1$–$J_2$ model, the lower critical coupling $J_2 = 0.34 J_1$ is smaller than the ones reported in the literature, i.e. $J_2 \approx 0.40 J_1$, see Sec. I. In particular, it is even smaller than the one derived within linear spin–wave theory, $J_2 \approx 0.38 J_1$, (corrections up to second order in the $1/S$ expansion of the sublattice magnetization even increase the lower critical coupling, i.e., the region of stability of the Néel phase increases when $1/S$ corrections are added to the linear spin–wave results, see Ref. [6] for details). Differently from spin–wave theory, where $1/S$ can be taken as a small parameter, the bond operator formalism lacks such a quantity (in principle, the density of excited triplets can be considered as a small parameter, see Ref. [33] for details) and therefore, it is difficult to systematically determine corrections to the mean–field results. We believe that the results derived here could be improved once: (a) the full singlet and triplet propagators, instead of the bare ones, are employed in the calculation of the normal triplet [Eqs. (30)] and singlet [31] self–energies; (b) the anomalous cubic and quartic self–energies are considered; (c) the influence of the exact diagonalization data; and/or (d) an alternative treatment of the constraint (3) is employed (see Sec. II.C from Ref. [11] for details). However, it is difficult to say which one is the most relevant contribution to the determination of the phase boundary.

The nature of the excitation gap of the plaquette VBS phase is also affected by cubic and quartic vertices: for $J_2 < 0.48 J_1$, we find a triplet gap while for $J_2 > 0.48 J_1$, a singlet one. It should be contrasted with the harmonic approximation: the gap changes from a triplet gap to a singlet one at $J_2 = 0.82 J_1$. Interestingly, one of the first exact diagonalization data[8] for the $J_1$–$J_2$ model indicates that the excitation gap is associated with a singlet–singlet excitation for $0.50 < J_2/J_1 < 0.60$. We should also note that: (i) The hierarchical mean–field approach[27,28,49] also indicates that the excitation gap changes from a triplet to a singlet one, but at $J_2 \approx 0.57 J_1$; (ii) The DMRG calculations recently reported in Ref. [11] which find some evidences for a $Z_2$ spin–liquid phase, point to a singlet gap smaller than the triplet one within the whole disordered region.

Cubic and quartic vertices also influence the nature of the phase transitions at small and large $J_2$. Recall that (Sec. [1]) for $J_2 = 0.34 J_1$, a triplet gap vanishes, indicating a continuous quantum phase transition either to an ordered phase or to a mixed phase[50] (Néel phase with plaquette modulation). As discussed in the Introduction, the former scenario is in favor of the deconfined quantum criticality[37] for the Néel–VBS transition while the latter scenario is in agreement with the Landau–Ginzburg framework. On the other hand, for $J_2 = 0.59 J_1$, a singlet gap vanishes which, in principle, points to a continuous quantum phase transition to a dimerized columnar VBS phase: note that a suitable linear combination of $\ket{S_0}$ and $\ket{S_1}$ [see Eq. (A2) and Fig. [11]] yields a (columnar) dimer state. Here, a continuous transition to a mixed phase (columnar VBS with plaquette modulation) should not be excluded either[50] Such a result is in contradiction with previous ones (see Sec. [1]) which indicate that a first–order quantum phase transition takes place at $J_2 \approx 0.60 J_1$ from a quantum paramagnetic phase to a collinear (ordered) one (see discussion below).

Finally, in order to check the accuracy of our results, it is interesting to compare the ground state energy and the excitation gap for $J_2 = 0.5 J_1$, which is deep in the disordered phase, with the available data. Within the cubic–quartic approximation (Sec. [v]), we find that $E_{EGS} = −0.477 J_1$, which is in reasonable agreement with (plaquette) series expansion results[22] and a very recent DMRG (Ref. [51]) Green function Monte Carlo[23] hierarchical mean–field [see also note (i) above] and exact diagonalization[24] approaches. Recall that [Fig. 7(b)] we arrive at $\Delta = 0.30$ and $0.46 J_1$ for the singlet and triplet excitation gaps, respectively, which are larger than the values reported in the literature. As discussed above, such excitation gaps could decrease if, for instance, cubic and quartic interactions are self–consistently considered.

A. Consequences for the $J_1$–$J_2$ model

The results that we have derived within the bond–operator theory (cubic–quartic approximation) allow us to state that if a plaquette VBS phase sets in for $J_2 \approx 0.5 J_1$, then such a phase displays a singlet excitation gap. This is the same feature of a possible spin liquid phase described by recent DMRG simulations[49]. Therefore, the determination of the nature of the excitation gap is not enough to make a distinction between the plaquette VBS phase and a $Z_2$ spin–liquid for $J_2 \approx 0.5 J_1$.

The fact that a singlet gap vanishes at $J_2 = 0.59 J_1$ disagrees with previous calculations (see Sec. [1]). Such a result could indicate that: (a) the plaquette–columnar VBS transition is indeed a true quantum phase transition and a first–order columnar VBS–collinear quantum phase transition takes place at a larger $J_2$, (b) a first–order quantum phase transition to the collinear phase preempts the plaquette–columnar VBS transition, or (c) a mixed columnar–plaquette phase[59] may set in within the disordered region. It should be mentioned that the possibility of a series of intermediate paramagnetic phases between the Néel and the collinear phases [scenario (a)] is discussed in Ref. [53] and that the plaquette–columnar quantum phase transition was studied by Kotov et al. [54]
who showed that such a quantum critical point belongs to the O(1) universality class (equivalent to 3D Ising).

We intend to investigate the above scenario (c) within the bond–operator theory in a future publication.

VII. SUMMARY

In this paper, we revisited the work of Zhitomirsky and Ueda and studied the plaquette VBS phase of the square lattice $J_1-J_2$ AFM model within the (tetramerized) bond–operator theory. We improved the previous analysis by including the high–energy singlet state within the description and perturbatively taking into account the effects of cubic (singlet–triplet–triplet and triplet–triplet–triplet) and quartic vertices above the harmonic (mean–field) results. We showed that cubic and quartic interactions play an important role in the determination of the singlet and the triplet excitation spectra. As a consequence, the region of stability of the plaquette phase is smaller than the harmonic one. Interesting, we found that at $J_2 = 0.48 J_1$, the excitation gap of the plaquette VBS phase changes from a triplet gap to a singlet one, which vanishes at $J_2 = 0.59 J_1$.

We would like in the near future to apply the formalism discussed here to study the stability of the plaquette VBS phase in some extensions of the $J_1-J_2$ model. For instance, the square lattice $J_1-J_2-J_3$ AFM model, where there are some evidences\cite{18,26} that the inclusion of a next–next–nearest–neighbor AFM coupling $J_3$ favors the stability of the plaquette phase. We also believe that effects of anisotropy in the plaquette VBS phase can also be addressed. In this case, one candidate is the square lattice $J_{1XZ}-J_{2XZ}$ AFM model recently considered in Ref. \cite{55}.

As a final remark, we would like to mention that it would also be interesting to consider the AFM $J_1-J_2$ model on the honeycomb lattice within the procedure developed here. There are numerical evidences that a plaquette VBS phase may set in within the zero temperature phase diagram not only in the $J_1-J_2$ model\cite{10,11} but also in the $J_1-J_2-J_3$ model\cite{56,57}. However, this is a much more involved task since the Hilbert space of six spins $S = 1/2$ on a hexagon has 64 states: five singlet, 27 triplet, 25 quintet, and seven septet states. In this case, it is very difficult to determine the bond operator representation, i.e., the equivalent of Eq. (6), for the spin operators.

Note added. We recently became aware of DMRG calculations\cite{51} which indicates that the plaquette VBS phase is stable for $0.50 < J_2/J_1 < 0.61$. The authors also found that the Néel order vanishes for $J_2 > 0.44 J_1$ and that a possible gapless spin liquid phase may set in for $0.44 < J_2/J_1 < 0.50$.

FIG. 11. (Color online) Schematic representation of the singlet states $|s_0\rangle$ and $|s_1\rangle$ [Eq. (A2)]. The symbols are the same as in Figs. 1 and 2.

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Appendix A: Single–plaquette Hilbert space

In this section, we provide the expansion of the eigenvectors of the Hamiltonian (2) (the two singlet, $|s_0\rangle$ and $|s_1\rangle$, nine triplet, $|t_{a,\alpha}\rangle$ with $a = 1, 2, 3$ and $\alpha = x, y, z$, and five quintet, $|d_0\rangle$, $|d_2\rangle$, and $|d_\alpha\rangle$, states) in terms of the 16 states $|\uparrow\uparrow\uparrow\uparrow\rangle$, $|\downarrow\downarrow\uparrow\uparrow\rangle$, $|\uparrow\downarrow\uparrow\uparrow\rangle$, $|\downarrow\uparrow\downarrow\uparrow\rangle$, ..., etc. It is
possible to show that
\[ |s_0\rangle = \frac{1}{2\sqrt{3}} (2| \uparrow \uparrow \downarrow \rangle + 2| \downarrow \uparrow \uparrow \rangle - | \uparrow \downarrow \downarrow \rangle - | \downarrow \uparrow \uparrow \rangle), \]
\[ |s_1\rangle = \frac{1}{2} (| \downarrow \uparrow \uparrow \downarrow \rangle + | \uparrow \downarrow \downarrow \uparrow \rangle - | \uparrow \uparrow \downarrow \downarrow \rangle - | \downarrow \uparrow \downarrow \uparrow \rangle), \]
\[ |t_{1,a}\rangle = \frac{\lambda_a}{2} (| \uparrow \uparrow \uparrow \downarrow \rangle - | \uparrow \downarrow \downarrow \uparrow \rangle), \]
\[ |t_{1,z}\rangle = \frac{\lambda_z}{2} (| \uparrow \uparrow \uparrow \downarrow \rangle - | \uparrow \downarrow \downarrow \uparrow \rangle), \]
\[ |t_{2,\alpha}\rangle = \frac{\lambda_{\alpha}}{2} (| \uparrow \uparrow \downarrow \downarrow \rangle - | \uparrow \downarrow \downarrow \downarrow \rangle), \]
\[ |t_{3,\alpha}\rangle = \frac{\lambda_{\alpha}}{2\sqrt{2^3}} (| \uparrow \uparrow \downarrow \downarrow \rangle - | \uparrow \downarrow \downarrow \downarrow \rangle), \]
\[ |t_{3,z}\rangle = \frac{1}{\sqrt{2}} (| \uparrow \uparrow \uparrow \downarrow \rangle - | \downarrow \uparrow \uparrow \downarrow \rangle), \]
\[ |d_0\rangle = \frac{1}{\sqrt{6}} (| \uparrow \downarrow \downarrow \downarrow \rangle + | \downarrow \uparrow \downarrow \uparrow \rangle + | \uparrow \uparrow \downarrow \uparrow \rangle + | \downarrow \downarrow \uparrow \uparrow \rangle) \]
\[ + | \uparrow \uparrow \downarrow \downarrow \rangle | \uparrow \downarrow \downarrow \uparrow \rangle + | \downarrow \uparrow \downarrow \uparrow \rangle + | \downarrow \downarrow \uparrow \uparrow \rangle), \]
\[ |d_n\rangle = \frac{\lambda_n}{2\sqrt{2^3}} (| \uparrow \downarrow \uparrow \uparrow \downarrow \rangle + | \downarrow \uparrow \downarrow \uparrow \downarrow \rangle + | \uparrow \uparrow \uparrow \downarrow \downarrow \rangle + | \uparrow \uparrow \downarrow \uparrow \downarrow \rangle), \]
\[ |d_1\rangle = \frac{1}{\sqrt{2}} (| \downarrow \downarrow \downarrow \downarrow \rangle - | \uparrow \uparrow \uparrow \uparrow \rangle), \]
\[ |d_2\rangle = \frac{1}{\sqrt{2}} (| \downarrow \downarrow \downarrow \downarrow \rangle + | \uparrow \uparrow \uparrow \uparrow \rangle), \]
\[ \text{(A1)} \]

where the upper and lower signs respectively refer to \( \alpha = x \) and \( \gamma \), \( \lambda_x = 1 \), and \( \lambda_{\gamma} = i \). In particular, the singlet states can also be written as \[ |s_0\rangle = \frac{1}{\sqrt{3}} (|1, 2| [4, 3] + |1, 4| [2, 3]), \]
\[ |s_1\rangle = |1, 2| [4, 3] - |1, 4| [2, 3], \]
\[ \text{(A2)} \]

where \([i, j]| = i, j = 1, 2, 3, 4\) denotes that the spins \( S^i \) and \( S^j \) form a singlet, see Figs. \[1\] and \[11\]. Note that \(|s_0\rangle\) is even while \(|s_1\rangle\) is odd under a \( \pi/2 \) rotation.

**Appendix B: Details: effective boson model**

Here, we quote the explicitly expressions of the coefficients \( A_{k}^{ab}, B_{k}^{ab}, c_{k}^{abc}, \chi_{k}^{abcd}, \bar{c}_{k}^{abc}, \) and \( \chi_{k}^{abcd} \) see Eqs. \[14\]–\[18\] of
\[ A_{k}^{ab} = (E_{l1} - \mu) (\delta_{a,1}\delta_{b,1} + \delta_{a,2}\delta_{b,2}) \]
\[ + (E_{l3} - \mu) \delta_{a,3}\delta_{b,3} + B_{k}^{ab}, \]
\[ B_{k}^{ab} = N_{0} \sum_{n} [g_{2}^{ab}(n)e^{i\mathbf{k}\cdot\mathbf{n}} + g_{2}^{ba}(n)e^{-i\mathbf{k}\cdot\mathbf{n}}], \]
\[ \xi_{k}^{abc} = -i N_{0}^{1/2} \sum_{n} [g_{3}^{abc}(n)e^{-i\mathbf{k}\cdot\mathbf{n}} + g_{3}^{bac}(n)e^{i\mathbf{k}\cdot\mathbf{n}}], \]
\[ \chi_{k}^{abcd} = - \frac{1}{2} \sum_{n} [g_{4}^{abcd}(n)e^{i\mathbf{k}\cdot\mathbf{n}} + g_{4}^{dabc}(n)e^{-i\mathbf{k}\cdot\mathbf{n}}], \]
\[ \bar{c}_{k}^{abc} = N_{0}^{1/2} \sum_{n} [g_{5}^{abc}(n)e^{-i\mathbf{k}\cdot\mathbf{n}} + g_{5}^{bac}(n)e^{i\mathbf{k}\cdot\mathbf{n}}], \]
\[ \chi_{k}^{abc} = \sum_{n} [g_{6}^{abc}(n)e^{-i\mathbf{k}\cdot\mathbf{n}} + g_{6}^{bac}(n)e^{i\mathbf{k}\cdot\mathbf{n}}], \]
\[ \text{(B1)} \]

with \( a, b, c, d = 1, 2, \) and \( 3 \). \( E_{l1} \) and \( E_{l3} \) are the triplet eigenvalues \( \mathbf{3} \) of the single plaquette Hamiltonian \( \mathbf{2} \) and the \( g \) coefficients are given by
\[ g_{n}^{ab}(n) = J_{1} (C_{a}^{2} C_{b}^{1} + C_{a}^{3} C_{b}^{4}) \delta_{n,1} \]
\[ + J_{1} (C_{a}^{1} C_{b}^{4} + C_{a}^{3} C_{b}^{2}) \delta_{n,2} \]
\[ + J_{2} C_{a}^{2} C_{b}^{1} (\delta_{n,1} + \delta_{n,-2} + \delta_{n,1-2}) \]
\[ + J_{2} C_{a}^{3} C_{b}^{1} (\delta_{n,1} + \delta_{n,-2} + \delta_{n,1+2}) \]
\[ g_{3}^{abc}(n) = J_{1} (C_{a}^{2} D_{bc}^{1} + C_{a}^{3} D_{bc}^{4}) \delta_{n,1} \]
\[ + J_{1} (C_{a}^{1} D_{bc}^{4} + C_{a}^{3} D_{bc}^{2}) \delta_{n,2} \]
\[ + J_{2} C_{a}^{2} D_{bc}^{1} (\delta_{n,1} + \delta_{n,-2} + \delta_{n,1-2}) \]
\[ + J_{2} C_{a}^{3} D_{bc}^{1} (\delta_{n,1} + \delta_{n,-2} + \delta_{n,1+2}) \]
\[ \tilde{g}_{3}^{abc}(n) = J_{1} (D_{a}^{2} C_{c}^{1} + D_{a}^{3} C_{c}^{4}) \delta_{n,1} \]
\[ + J_{1} (D_{a}^{1} C_{c}^{4} + D_{a}^{3} C_{c}^{2}) \delta_{n,2} \]
\[ + J_{2} D_{a}^{2} C_{c}^{1} (\delta_{n,1} + \delta_{n,-2} + \delta_{n,1-2}) \]
\[ + J_{2} D_{a}^{3} C_{c}^{1} (\delta_{n,1} + \delta_{n,-2} + \delta_{n,1+2}) \].
\[ g_{abcd}(n) = J_1 \left( D_{ab}^2 D_{cd}^1 + D_{ab}^1 D_{cd}^2 \right) \delta_{n,1} + J_1 \left( D_{ab}^4 D_{cd}^1 + D_{ab}^1 D_{cd}^4 \right) \delta_{n,2} + J_2 D_{ab}^2 D_{cd}^4 \left( \delta_{n,1} + \delta_{n,-2} + \delta_{n,-1,2} \right) + J_2 D_{ab}^3 D_{cd}^1 \left( \delta_{n,1} + \delta_{n,2} + \delta_{n,1,2} \right), \quad (B2) \]

\[ g_{ab}(n) = g_{so}(n) \] with the replacements \( C \leftrightarrow C, g_{so}(n) = g_{ab}(n) \) with the replacements \( C \leftrightarrow C \), and \( g_{so}(n) = g_{ab}(n) \) with \( C \rightarrow C \). Here \( n = 1, 2 \) corresponds to the nearest-neighbor vectors \[(11)\] and the \( C, C \), and \( D \) coefficients are shown in Eq. \[0\].

**Appendix C: Diagonalization harmonic Hamiltonian**

In this section, we briefly summarize the analytical procedure used to diagonalize the triplet sector of the harmonic Hamiltonian \[(20)\]. In order to deal with such a \( 6 \times 6 \) problem, we follow the procedure described in Refs. \[58\] and \[59\]. It should be mentioned that we have recently employed this scheme to diagonalize a similar \( 4 \times 4 \) problem \[(22)\].

Since we are considering a bosonic system, instead of \( \hat{H}_k \) [see Eq. \[(20)\]], we should diagonalize

\[ \hat{I}_B \hat{H}_k, \quad \text{with} \quad \hat{I}_B = \begin{pmatrix} \hat{I} & 0 \\ 0 & -\hat{I} \end{pmatrix}, \quad (C1) \]

where \( \hat{I} \) is the \( 3 \times 3 \) identity matrix. It is easy to show that the (positive) eigenvalues of the matrix \[(C1)\] are (roots of a cubic polynomial)

\[ \Omega_{1/2,k} = \left[ -\frac{1}{3} a_{2,k} - \text{Re}(S_k) \pm \sqrt{3} \text{Im}(S_k) \right]^{1/2}, \quad (C2) \]

\[ \Omega_{3,k} = \left[ -\frac{1}{3} a_{2,k} + 2\text{Re}(S_k) \right]^{1/2}, \]

where

\[ S_k = \left( R_k + i\sqrt{D_k} \right)^{1/3}, \quad D_k = -Q_k^2 - R_k, \]

\[ Q_k = \frac{1}{9} \left( 3a_{1,k} - a_{2,k}^2 \right), \quad (C3) \]

\[ R_k = \frac{1}{54} \left( 9a_{2,k}a_{1,k} - 27a_{0,k} - 2a_{2,k}^3 \right). \]

The coefficients \( a_{i,k} \) read

\[ a_{0,k} = \left( A_{11}^k - B_{11}^k \right) \left( A_{22}^k - B_{22}^k \right) \left( A_{33}^k - B_{33}^k \right) \]

\[ 4(B_{12}^k)^2 \left( A_{33}^k + B_{33}^k \right) + 4(B_{23}^k)^2 \left( A_{11}^k + B_{11}^k \right) \]

\[ + 4(B_{13}^k)^2 \left( A_{22}^k + B_{22}^k \right) - 16B_{12}^k B_{23}^k B_{13}^k \]

\[ - \left( A_{11}^k + B_{11}^k \right) \left( A_{22}^k + B_{22}^k \right) \left( A_{33}^k + B_{33}^k \right) \]

\[ a_{1,k} = \omega_{1,k}^2 \left( 2 + \omega_{2,k}^2 + \omega_{3,k}^2 \right) \]

\[ - 4(B_{12}^k)^2 \left( A_{11}^k - B_{11}^k \right) \left( A_{22}^k - B_{22}^k \right) \]

\[ - 4(B_{23}^k)^2 \left( A_{22}^k + B_{22}^k \right) \left( A_{33}^k - B_{33}^k \right) \]

\[ - 4(B_{13}^k)^2 \left( A_{11}^k + B_{11}^k \right) \left( A_{33}^k + B_{33}^k \right), \quad (C4) \]

where \( \omega_{i,k} = \left( A_{ij}^k \right)^2 - \left( B_{ij}^k \right)^2 \), with \( i = 1, 2, 3 \), and \( A_{ij}^k \) and \( B_{ij}^k \) [see Eq. \[(B1)\]] are respectively the elements of the \( 3 \times 3 \) Hermitian \( A_k \) and \( B_k \) [see Eq. \[(21)\)].

The determination of the Bogoliubov coefficients \( u_{ij}^k \) and \( v_{ij}^k \), the elements of the \( 3 \times 3 \) matrices \( \hat{U}_k \) and \( \hat{V}_k \), is quite involved. Using the properties of the matrix \( \hat{M}_k \) (see Sec. 5 from Ref. \[58\]) and after some lengthy algebra, it is possible to show that

\[ u_{ij}^k = \frac{\mu_{ij}^k}{G_{ij}^k} \left( B_{ij}^k - A_{ij}^k - \Omega_{ij}^k \right) \left( \Omega_{ij}^k + A_{33}^k - B_{33}^k \right), \]

\[ u_{33}^k = -\frac{i}{G_{33}^k} \left[ (\Omega_{33}^k + A_{33}^k) \nu_{33}^k + 2\lambda_{33}^k \left( A_{22}^k - B_{22}^k \right) \right] \]

\[ + 2\mu_{33}^k \left( A_{11}^k - B_{11}^k \right), \quad (C5) \]

where \( j = 1, 2 \),

\[ \mu_{12}^k = 2B_{12}^k B_{23}^k \left( A_{22}^k - B_{22}^k \right) + B_{13}^k \left( \Omega_{23}^k \right)^2 - \omega_{2,k}^2 \left( \Omega_{12}^k \right)^2, \]

\[ \mu_{23}^k = 2B_{23}^k B_{13}^k \left( A_{11}^k - B_{11}^k \right) + B_{23}^k \left( \Omega_{13}^k \right)^2 - \omega_{1,k}^2 \left( \Omega_{23}^k \right)^2, \]

\[ \nu_{ij}^k = \left( \Omega_{ij}^k \right)^2 - \omega_{ij}^2 \left( \Omega_{ij}^k \right)^2 \]

\[ - 4(B_{ij}^k)^2 \left( A_{11}^k - B_{11}^k \right) \left( A_{22}^k - B_{22}^k \right), \]
In this section, we study the dimerized columnar [Fig. 2(a)] and staggered [Fig. 2(b)] VBS phases of the $J_1$–$J_2$ model within the (dimer) bond–operator formalism \cite{000} at the harmonic approximation. We only quote the main results and refer the reader to Secs. II and III from Ref. \cite{000} for more details.

The effective model [the equivalent of Eq. (12)] in terms of the boson triplet operators $t_{k\alpha}$ with $\alpha = x, y, z$ reads

$$H = -3J_1 N/8 - \mu N (N_0 - 1)/2$$

$$+ \sum_{k} \left[ A_k^{\dagger} t_{k\alpha}^{\dagger} t_{k\alpha} + \frac{1}{2} B_{k} t_{k\alpha}^{\dagger} t_{k\alpha}^{\dagger} + H.c. \right]$$

$$+ \frac{1}{2\sqrt{N}} \xi_{k} [t_{k\alpha}^{\dagger} t_{k\beta}^{\dagger} t_{k\gamma} t_{k\lambda} + H.c.]$$

$$+ \frac{1}{2\sqrt{N}} \epsilon_{\alpha\beta\lambda} \sum_{p,q} \gamma_{k} t_{p\alpha}^{\dagger} t_{q\beta}^{\dagger} t_{p\gamma} t_{q\lambda}.$$  \hspace{1cm} (D1)

Here, $N' = N/2$ with $N$ being the number of sites of the original square lattice, the momentum sum runs over the dimerized Brillouin zone,

$$A_k = \frac{1}{4} J_1 - \mu + B_{k},$$

$$B_{k} = \frac{1}{2} N_0 \left[ -J_1 \cos(2k_x) + 2(J_1 - J_2) \cos(k_y) \right.$$

$$- J_2 \cos(2k_x + k_y) - J_2 \cos(2k_x - k_y)],$$

$$\xi_{k} = -\sqrt{N_0} \left[ J_1 \sin(2k_x) + J_2 \sin(2k_x + k_y) + J_2 \sin(2k_x - k_y) \right]$$

$$+ \frac{1}{2} [J_1 \cos(2k_x) + 2(J_1 + J_2) \cos k_y$$

$$+ J_2 \cos(2k_x + k_y) + J_2 \cos(2k_x - k_y)]$$  \hspace{1cm} (D2)

for the columnar VBS, and

$$B_{k} = \frac{1}{2} N_0 \left[ (2J_2 - J_1) \cos k_x + \cos k_y \right. - J_1 \cos(k_x - k_y)],$$

$$\xi_{k} = \sqrt{N_0} J_1 \left[ \sin k_y + \sin(k_y - k_x) - \sin(k_x) \right],$$  \hspace{1cm} (D3)

$$\gamma_{k} = -\frac{1}{2} \left[ (2J_2 + J_1) \cos k_x + \cos k_y \right.$$

$$+ J_1 \cos(k_x - k_y)]$$

for the staggered VBS. In deriving Eq. (D1), we considered the following nearest–neighbor vectors: $\tau_1 = 2a \hat{x} = a_1$ and $\tau_2 = a \hat{y} = a_2$ (columnar) and $\tau_1 = a \hat{x} = a_1$ and $\tau_2 = a \hat{y} = a_2$ (staggered), see Fig. 2 and we set $a = 1$.

Similar to the plaquette phase, the parameter $N_{1/2}$ is the average value of the singlet operator $s_i$ while $\mu$ is the Lagrange multiplier that enforce (on average) the constraint on the total number of bosons per site (dimerized lattice).

Within the harmonic approximation, the Hamiltonian (D1) can be diagonalized, and therefore one finds that the ground state energy is given by

$$E_{EGS} = \frac{3}{8} J_1 N N_0 - \frac{1}{2} \mu (N_0 - 1) + \frac{3}{2} \sum_{k} (\omega_{k} - A_{k}),$$  \hspace{1cm} (D4)
while the energy of the triplet excitations assume the form
\[ \omega_k = \sqrt{A_k^2 - B_k^2}. \]  
(5)

After self-consistently calculating \( N_0 \) and \( \mu_s \), we find the behaviour of the ground state energy [Fig. 5(b)] and the excitation gaps (Fig. 12) in terms of \( J_2/J_1 \). Recall that for the columnar VBS phase, the \( Y = (0, \pi) \) and \( \Gamma = (0, 0) \) vectors correspond to the \((\pi, \pi)\) and \((0, 0)\) vectors of the original (nondimerized) square lattice.  

**Appendix E: Details: cubic–quartic approximation**

The renormalized cubic vertices \( \Gamma_{1/2,k,p}^{abc} \) and \( \Gamma_{1s/3s,4s,k,p}^{ab} \) [see Fig. 8(a)] and Eqs. (26) and (27) in terms of the Bogoliubov coefficients \( u_{k}^{ab} \) and \( v_{k}^{ab} \) are given by
\[
\Gamma_{1s,k,p}^{abc} = \sum_{ab} 3 \xi_{ab} v_{k-p}^{ab} u_{k}^{bc} + \xi_{ab} v_{k-p}^{ab} u_{k}^{bc},
\]
\[
\Gamma_{3s,k,p}^{abc} = \sum_{ab} \xi_{ab} v_{k-p}^{ab} u_{k}^{bc} + \xi_{ab} v_{k-p}^{ab} u_{k}^{bc}.
\]

and \( \Gamma_{4s,k,p}^{ab} = \Gamma_{3s,k,p}^{abc} \) with the replacement \( u \leftrightarrow v \).

The components of the normal triplet \( \Sigma_a(k,\omega) \) [Eq. (30)] and singlet \( \Sigma_s(k,\omega) \) [Eq. (31)] self-energies read
\[
\Sigma_a^{(b1)}(k,\omega) = \frac{1}{N^2} \sum_{b,c} \sum_{p} \left| \frac{\Gamma_{1s,k,p}^{abc}}{3} \right|^2 \omega - \Omega_{b,p} - \Omega_{c,k-p} + i\delta,
\]
\[
\Sigma_a^{(b4)}(k,\omega) = -\frac{1}{N^2} \sum_{b,c} \sum_{p} \left| \frac{\Gamma_{2s,k,p}^{abc}}{3} \right|^2 \omega - \Omega_{b,p} - \Omega_{c,p-k} + i\delta,
\]
\[
\Sigma_a^{(c1)}(k,\omega) = \frac{1}{N^2} \sum_{b} \sum_{p} \left| \frac{\Gamma_{1s,k,p}^{abc}}{3} \right|^2 \omega - \Omega_{b,k-p} + i\delta,
\]
\[
\Sigma_a^{(c3)}(k,\omega) = -\frac{1}{N^2} \sum_{b} \sum_{p} \left| \frac{\Gamma_{2s,k,p}^{abc}}{3} \right|^2 \omega - \Omega_{b,k-p} + i\delta,
\]
with \( a = 1, 2, 3 \) and
\[
\Sigma_a^{(d2)}(k,\omega) = \frac{3}{N^2} \sum_{a,b} \sum_{p} \frac{\Gamma_{3s,k,p}^{abc}}{3} \left( \frac{\Gamma_{2s,k,p}^{abc} + \Gamma_{3s,k,p}^{abc}}{3} \right)
\]
\[
\Sigma_a^{(d3)}(k,\omega) = -\frac{3}{N^2} \sum_{a,b} \sum_{p} \frac{\Gamma_{4s,k,p}^{abc} + \Gamma_{3s,k,p}^{abc}}{3} \left( \frac{\Gamma_{4s,k,p}^{abc} + \Gamma_{3s,k,p}^{abc}}{3} \right).
\]

The coefficients of the quartic terms \( H_{40} \) and \( H_{22} \) within the Hartree–Fock approximation [Eqs. (32) and (33)] are given by
\[
A_{ab,p}^{HF} = \frac{3}{N^2} \sum_{c,a,b,c,d} \chi_{k}^{abc} \left( u_{p}^{ab} v_{k-p}^{bc} + \text{cyclic} \right),
\]
\[
B_{ab,p}^{HF} = \frac{3}{N^2} \sum_{c,a,b,c,d} \chi_{k}^{abc} \left( u_{p}^{ab} v_{k-p}^{bc} + \text{cyclic} \right),
\]
\[
A_{s,p}^{HF} = \frac{3}{N^2} \sum_{k} \chi_{k}^{abc} \left( u_{k-p}^{ab} v_{p}^{bc} + \text{cyclic} \right),
\]
\[
B_{s,p}^{HF} = \frac{3}{N^2} \sum_{k} \chi_{k}^{abc} \left( u_{k-p}^{ab} v_{p}^{bc} + \text{cyclic} \right).
\]

Finally, it is possible to show that only the cubic vertices (2) and (4s) in Fig. 8(a) contribute to the ground state energy and therefore, we have
\[
E_{EGS}^{(3)} = -\frac{1}{N} \sum_{a,b,c,k,p} \left| \frac{\Gamma_{2s,k-p}^{abc}}{3} \right|^2 \frac{\Omega_{c,k} + \Omega_{b,p} + \Omega_{a,p-k}}{3},
\]
\[
E_{EGS}^{(4)} = -\frac{3}{N} \sum_{a,b,c,k,p} \left| \frac{\Gamma_{4s,k-p}^{abc}}{3} \right|^2 \frac{\Omega_{c,k-p} + \Omega_{a,p-k}}{3}.
\]

The correction to the ground state energy due to the quartic terms read
\[
E_{EGS}^{(3)} = \frac{9}{N} \sum_{a,b,c,d} \chi_{k}^{abc} \left( u_{p}^{ab} v_{k-p}^{bc} + \text{cyclic} \right),
\]
