Plaquette Order in the $J_1$-$J_2$-$J_3$ model: a series expansion analysis

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Series expansion based on the flow equation method is employed to study the zero temperature properties of the spin-$1/2$ $J_1$-$J_2$-$J_3$ antiferromagnet in two dimensions. Starting from the exact limit of decoupled plaquettes in a particular generalized $J_1$-$J_2$-$J_3$ model we analyze the evolution of the ground state energy and the elementary triplet excitations in powers of all three inter-plaquette couplings up to fifth order. We find the plaquette phase to remain stable over a wide range of exchange couplings and to connect adiabatically up to the case of the plain $J_1$-$J_2$-$J_3$ model, however not to the $J_1$-$J_2$ model at $J_3 = 0$. Besides confirming the existence of such a phase, recently predicted by Mambrini, et al. [Phys. Rev. B 74, 144422 (2006)], we estimate its extent by Dlog-Padé analysis of the critical lines that result from closure of the triplet gap.

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INTRODUCTION

The study of quasi two-dimensional (2D) materials with frustrated magnetic exchange interactions is a field of intense research. This research is driven by the quest for systems which may exhibit exotic magnetic phases instead of simple long range anti/ferromagnetic order (AFM/FM LRO) [1]. Prominent examples of such phases are spin liquids (SL), with no ordering of any type, or valence bond states. The latter may occur as solids (VBS) with no breaking of lattice symmetries but potentially other hidden order, such as eg. string ordering, moreover valence bond crystals (VBC) are frequent, where lattice symmetries are directly broken in favor of eg. columnar or plaquette ordering [1, 2, 3]. Even for simple frustrated systems a quantitative understanding of the complete phase diagram is still lacking. The AFM spin-$1/2$ $J_1$-$J_2$ model on the square lattice, is a paradigmatic case in this respect. This model corresponds to Fig. 1 for $J_1 = J_0 > 0$, $J_2 > 0$ and $J_3 = 0$, where $J_1$ (and $J_0$) is the nearest neighbor exchange interaction and frustration is induced by the next-nearest neighbor exchange interaction $J_2$. Experimentally, Li$_2$VOXO$_4$ (X = Ge, Si), which has been discovered recently, is a promising candidate to realize the $J_1$-$J_2$ model in the range $J_2/J_1 \sim 5 - 10$ [4, 5]. Two limiting cases of $J_1$-$J_2$ model are well-understood. For $J_2 = 0$ and $J_1 > 0$, the system is the 2D Heisenberg AFM, which exhibits Néel LRO. In the opposite limit, $J_1/J_2 \rightarrow 0$, the system turns into a set of two decoupled AFMs on the 2D A and B sublattices. These lock into a collinear state by the order-from-disorder effect due to the finite $J_2$. In the intermediate regime, $0.4 \approx (J_2/J_1)_{c_1} < J_2/J_1 < (J_2/J_1)_{c_2} \approx 0.6$, both, the Néel and the collinear state are known to be unstable. Here, different approaches, including exact diagonalization (ED) [4, 4, 8], quantum Monte Carlo (QMC) [1, 10], spin wave theory (SW) [11], large-$N$ expansion [12], and series expansion (SE) [13, 14, 15, 16, 17, 18], have confirmed that one or several quantum disordered phases with a singlet ground state and a gap to magnetic excitations may be present. The precise nature of the intermediate phase (S), however, is still controversial. In the simplest scenario, considering the existence of a single intermediate phase only, a plaquette VBC [9], a columnar VBC [12] and a SL [10] have been proposed. Other studies suggest that the intermediate phase could be composed of two SL-like phases [16].

The main purpose of this paper is to put the $J_1$-$J_2$
model into a broader perspective, by considering an extended version, i.e. the $J_1$-$J_2$-$J_3$ model, which is depicted in Fig. 1 for $J_1 = J_0$ and includes a third nearest-neighbor interaction $J_3$ (for clarity only some of the $J_3$ couplings are shown). Classically, the competing interactions $J_2/J_1$ and $J_3/J_1$ lead to four ordered phases of the $J_1$-$J_2$-$J_3$ model [15, 20, 21]. Among them, Néel and helicoidal phases, which are separated by a classical critical line $(J_2 = 2J_3)/J_1 = 1/2$ exist. The Néel phase remains rather stable against quantum fluctuations, although it has been conjectured that critical line, at $J_2 = 0$, should be shifted to $J_3/J_1 > 1/4$ once the quantum model is considered [21].

The nature of the quantum phases in a selected region $J_1$, $J_2$, and $J_3$ has been considered recently by Mambrini et al. [22]. By employing ED and diagonalization in a subset of short-range valence bond singlets (SRVB method) these authors have found evidence for a VBC in a subset of short-range valence bond singlets (SR VB 3). By employing ED and diagonalization in a subset of short-range valence bond singlets (SRVB method) these authors have found evidence for a VBC in a subset of short-range valence bond singlets (SR VB 3). By employing ED and diagonalization in a subset of short-range valence bond singlets (SRVB method) these authors have found evidence for a VBC in a subset of short-range valence bond singlets (SR VB 3). By employing ED and diagonalization in a subset of short-range valence bond singlets (SRVB method) these authors have found evidence for a VBC in a subset of short-range valence bond singlets (SR VB 3). By employing ED and diagonalization in a subset of short-range valence bond singlets (SRVB method) these authors have found evidence for a VBC in a subset of short-range valence bond singlets (SR VB 3).

In this paper we will complement and extend these findings by performing SE analysis. In particular we will aim at a quantitative determination of the extension of the plaquette phase around the previously mentioned line by localizing the critical lines for a closure of the triplet gap.

Our strategy will be to analyze perturbatively the evolution of the ground state of a generalized version of the $J_1$-$J_2$-$J_3$ model. For this version $J_0 \neq J_1$. At $J_{1,3} = 0$ and $J_2 \neq 0$ only on those squares formed by the $J_0$-links the generalized $J_1$-$J_2$-$J_3$ model shown in Fig. 1 exhibits a product-state of disconnected bare four-spin plaquettes. This will be the unperturbed ground state from which we start. The local $J_0$ couplings (bold lines) will be set to unity hereafter. Therefore at $J_1 = 1$ we recover the $J_1$-$J_2$-$J_3$ model (in units of $J_1$) from the generalized model. The Hamiltonian of generalized model is

$$H = H_0 + V; \quad H_0 = \sum_{i=1}^{n} h_{0,i}; \quad V = \sum_{i=1}^{n} (V_{1,1} + V_{2,1} + V_{3,1}),$$

where $h_{0,i}$ refers to the local plaquette at site $i$, given by

$$h_{0,i} = [S_1 \cdot S_2 + S_2 \cdot S_3 + S_3 \cdot S_4 + S_4 \cdot S_1 + J_2(S_1 \cdot S_3 + S_2 \cdot S_4)]_1$$

$$= \frac{1}{2}[S_{1234}^2 - S_{13}^2 - S_{24}^2 + J_2(S_{13}^2 + S_{24}^2 - 3)],$$

in which $S_{1...n} = S_1 + \ldots + S_n$. $V_{1,1}$, $V_{2,1}$ and $V_{3,1}$ in Eq. (1) represent the inter-plaquette coupling at site $i$ via nearest $(J_1)$, next nearest $(J_2)$ and next-next nearest $(J_3)$ interactions, respectively.

$$V_{1,1} = J_1[S_{1,1} \cdot S_{2,1} + S_{1,1} \cdot S_{1,1} + S_{1,1} \cdot S_{4,1} + S_{4,1} \cdot S_1]$$

$$V_{2,1} = J_2[S_{1,1} \cdot S_{2,1} + S_{1,1} \cdot S_{1,1} + S_{4,1} \cdot S_{4,1} + S_{4,1} \cdot S_1 + S_{1,1} \cdot S_{2,1}].$$

Table I shows the eigenstates of a local plaquette Hamiltonian, $h_{0,i}$, in which each state is labeled by the ground state energy and thereafter the ground state is set to unity.

| State | $\Psi$ | $S_{1234}$ | $S_{13}$ | $S_{24}$ |
|-------|--------|------------|----------|----------|
| $|S_i\rangle$ | $\frac{1}{2}J_2$ | 0 | 1 | 1 |
| $|T_i\rangle$ | $\frac{1}{2}J_2 + 1$ | 1 | 1 | 1 |
| $|S_i\rangle$ | $2J_2 + 2$ | 0 | 0 | 0 |
| $|T_i\rangle$ | $\frac{1}{2}J_2 + 2$ | 1 | 0 | 0 |
| $|Q_i\rangle$ | $\frac{1}{2}J_2 + 3$ | 0 | 1 | 1 |

Table II shows the eigenstates of a local plaquette Hamiltonian, $h_{0,i}$, in which each state is labeled by the ground state energy: $e_0$, the total spin: $S_{1234}$, and the spin along each diagonal: $S_{13}$ and $S_{24}$. From this table it follows that for $0 \leq J_2 \leq 1$ the ground state is $|S_i\rangle$, i.e. a spin singlet along the plaquette and triplets along the diagonals. For $0 \leq J_2 \leq \frac{1}{2}$ the first excited state is $|T_i\rangle$, i.e. triplets along both the plaquette and the diagonals. At $J_2 = 1$ there is a crossover in the ground state energy and thereafter the ground state is $|S_i\rangle$, i.e. singlets along the plaquette and the diagonals. The other states are total triplets, $|T_{i2}\rangle$ and $|T_{i1}\rangle$, consisting of a triplet on one of the diagonals and a triplet on the other one.

### SERIES EXPANSION BY CONTINUOUS UNITARY TRANSFORMATION

In this Section we briefly describe the SE expansion in terms of $J_1$, $J_2$ and $J_3$. First, we rewrite the Hamiltonian (Eq. (1)) as

$$H = H_0(J_2 = 0) + J_2O^2 + \sum_{i=1}^{3} \left( \sum_{n=-N}^{N} O_i^n \right),$$

where $H_0$ has been split into the first two terms. The first one, $H_0(J_2 = 0)$, has a set of equally spaced energy levels (Table II). These are labeled with a total particle-number operator: $Q = \sum q_i$. $J_0 = 0$ corresponds to zero particle states: $|0\rangle = \prod |S_i\rangle_1$. $Q = 1$ sector corresponds to one-particle states: $|1\rangle = \prod |T_i\rangle_1 \otimes \prod |S_i\rangle_1$, i.e., a local triplet at site $i'$ created from the vacuum. $Q \geq 2$ sector of the spectrum is of multiparticle nature.

The second term in Eq. (4) refers to local contributions in $H_0$ proportional to $J_2$. The last three terms in the same Eq. (4) represent the inter-plaquette interactions,
via $J_1$, $J_2$ and $J_3$, respectively. There, $O_i^n$ operators non-locally create ($n \geq 0$) and destroy ($n < 0$) quanta within the ladder spectrum of $H_0(J_2 = 0)$. The explicit tabulation of $O_i^n$ in this model shows that $N \leq 4$ [23]. It has been shown [23] that models of type Eq. (4) allow for SE by means of Wegner’s continuous unitary transformation (CUT) method [25]. The basic idea is to map $H \rightarrow H_{\text{eff}}$, where

$$H_{\text{eff}} = H_0 + \sum_{k,m,l=1}^{\infty} C_{k,m,l} J_1^k J_2^m J_3^l. \quad (5)$$

The $C_{k,m,l}$ operators in Eq. (5) involve products of the $O_i^n$ operators of Eq. (4). However, as the main point and unlike in $H$, the effective Hamiltonian $H_{\text{eff}}$ is constructed to have a block diagonal structure, where each block has a fixed number of particles $Q$ of $H_0(J_2 = 0)$. This is achieved order by order in the expansion. We refer to Ref. [24] for further details. In the following Sections we will apply this technique to calculate the ground state energy and the one-particle excitations.

**DISPERSION OF ONE-TRIPLET EXCITATIONS**

In this Section we evaluate the dispersion of one-triplet states for different values of the coupling constants, $J_1$, $J_2$ and $J_3$. To this end, it is necessary to diagonalize $H_{\text{eff}}$ in the $Q = 1$ sector of $H_0(J_2 = 0)$, i.e., the subspace spanned by $|1\rangle$ states. Q-conservation implies that the sole action of $H_{\text{eff}}$ on the local triplet states refers to translation in real space, i.e.,

$$H_{\text{eff}} |1\rangle_0 = \sum_{c_1} c_1 |1\rangle_1, \quad (6)$$

where the $c_1$’s are the hopping amplitudes of a local triplet from origin $0$ to site $1$. Due to the lattice translational invariance Eq. (6) can be diagonalized by Fourier transformation

$$E_1(k) = \sum_{c_1} c_1 \exp(i \mathbf{k} \cdot \mathbf{l}). \quad (7)$$

From this, the dispersion $\omega(k)$ follows as

$$\omega(k) = E_1(k) - E_0, \quad (8)$$

where the ground state energy, $E_0$, is obtained by applying Q-conservation to the 0-particle sector, i.e., $E_0 = \langle 0 | H_{\text{eff}} | 0 \rangle$. It is important to note that, even without an explicit discussion of this quantity, Eq. (8) requires a full calculation of the ground state energy up to the same order as the hopping amplitudes.

By symmetry considerations not all the $c_1$’s are independent, which leaves only a subset of them to be calculated. Usually, in CUT applications, the $c_1$’s at $O(n)$ are obtained in the thermodynamic limit, by considering finite clusters which are large enough to embed all the paths of length $n$ [26] that connect origin $0$ with site $1$. In our model, due to the number of couplings considered and its dimensionality, this method becomes computationally very demanding. Alternatively, we have implemented a linked cluster approach, with subgraph subtraction to obtain the $c_1$’s. We refer to Ref. [24] for technical details of this method.

We have evaluated analytic expressions for the triplet dispersion, $\omega(k)$, keeping all three independent variables $J_1$, $J_2$, and $J_3$, i.e. without any parametrization, up to $O(5)$ [25].

Fig. 2 shows the dispersion obtained at $O(5)$, as a function of wave vector $k$, along high symmetry directions and for different values of the couplings. We have chosen paths in the couplings space that show the instabilities of the plaquette phase associated with triplet softening, i.e., $\omega(k) = 0$. We have selected two families of curves, parametrized according to $J_1$, $J_2 = b J_1$, and $J_3 = c J_1$, around $J_1 = 1$, the latter being the point where the $J_1$-$J_2$-$J_3$ model (in units of $J_1$) is recovered (see Fig. 1).

As shown in Fig. 2, triplet softening occurs at a critical wave vector of $k_{c_1} = (0,0)$ for the specific value of $(J_1, J_2, J_3) \approx (1, 0.2, 0.12)$, i.e. for relatively small values of $J_3$, as compared to $J_2$ (solid lines). Additionally, for larger values of $J_1$ a critical wave vector at $k_{c_2} = (\pi, \pi)$ (dashed lines) can be observed for the particular value of $(J_1, J_2, J_3) \approx (1.1, 0.15, 0.76)$ (dashed line). We have found no other values for critical wave vectors. Fig. 2 clar-
as parameter. Triplet softening occurs at $k_{c1} = (0,0)$ and $k_{c2} = (\pi,\pi)$, shown with solid and dashed lines, respectively. In all cases results from Dlog-Padé analysis are depicted. For $0 \leq J_3 \leq 0.4$, the instability at $k_{c1}$ limits the plaquette phase, projected onto $J_1 - J_2$ plane. But only for $J_3 \geq 0.05$ a plaquette phase appears in $J_1$-$J_2$-$J_3$ model (critical lines cross $J_1 = 1$). In particular, for $J_1 = 1$, $J_2$, and $J_3 = 0$, i.e. in the $J_1$-$J_2$ model, the plaquette phase is not present. For $0.7 \lesssim J_3 \lesssim 0.8$, the critical lines at $k_{c2}$ limit the plaquette phase projected onto $J_1 - J_2$ plane.

This will give us a quantitative estimate of the stability region of the plaquette phase in $J_1$-$J_2$-$J_3$ space will be studied in detail in the following Sections.

**STABILITY OF THE PLAQUETTE PHASE**

In this Section we discuss the quantum critical lines, resulting from the closure of the plaquette triplet gap, which resembles second order quantum phase transitions. This will give us a quantitative estimate of the stability region of the plaquette phase. In particular we are interested in a possible adiabatic connection of the isolated bare plaquette phase (with only local $J_3 \neq 0$) up to the value of $J_1 = 1$. This analysis is shown in Fig.3 which depicts the borders of the stability region projected onto $J_1 - J_2$ plane, taking $J_3$ as parameter.

Fig.3 displays two families of critical lines, corresponding to the closure of the triplet gap $\omega(k_{c}) = 0$ for $k_{c1} = (0,0)$ and $k_{c2} = (\pi,\pi)$, with dotted and solid lines, respectively. First, it is obvious that independently of $J_2$ and $J_3$ the plaquette phase extends from the origin, $J_1 = J_2 = J_3 = 0$ (not shown in Fig.3) up to $J_1 = 0.55$ below which there are no signals of triplet softening. Second, we focus on the $\omega(k_{c1}) = 0$ instability. In the case of $J_3 = 0$, as can be observed from the Figure, the critical line almost reaches, but does not cross the line $J_1 = 1$. In other words: the $J_1$-$J_2$ model does not show a plaquette phase. This result is consistent with the previous SE analysis on $J_1$-$J_2$ model in Ref. [13].

Third, we consider the simultaneous effect of $J_1$, $J_2$ and $J_3$. As can be observed in Fig.3 increasing the values of $J_3$ enlarges the region of stability of the plaquette phase in the $J_1$-$J_2$ plane in terms of the critical line $k_{c1} = (0,0)$ (solid lines). Most important, finite $J_3$ helps to stabilize the plaquette phase at $J_1 = 1$. In fact, already for $J_3 \approx 0.05$ the critical line crosses $J_1 = 1$. For $J_3 \approx 0.4$ the solid critical line merges with the lower righthand corner of Fig.3 and the plaquette phase extends over all of the $J_1 - J_2$ plane shown. These results are consistent with Ref. [22].

Now we turn to the plaquette phase stability region, projected onto $J_1 - J_2$ plane, limited by the critical lines $\omega(k_{c2}) = 0$ (dotted lines in Fig.3). We find a similar tendency as for $k_{c2}$, i.e. the region of stability of the plaquette phase in the $J_1$-$J_2$ plane is enlarged by increasing $J_3$. In this case however the impact of $J_3$ is somewhat less significant as compared to $J_2$. 

![FIG. 3: Critical lines ($\omega(k_{c}) = 0$) in $J_1$-$J_2$ plane and $J_3$ as parameter. Triplet softening occurs at $k_{c1} = (0,0)$ and $k_{c2} = (\pi,\pi)$, shown with solid and dashed lines, respectively. In all cases results from Dlog-Padé analysis are depicted. For $0 \leq J_3 \leq 0.4$, the instability at $k_{c1}$ limits the plaquette phase, projected onto $J_1 - J_2$ plane. But only for $J_3 \geq 0.05$ a plaquette phase appears in $J_1$-$J_2$-$J_3$ model (critical lines cross $J_1 = 1$). In particular, for $J_1 = 1$, $J_2$, and $J_3 = 0$, i.e. in the $J_1$-$J_2$ model, the plaquette phase is not present. For $0.7 \lesssim J_3 \lesssim 0.8$, the critical lines at $k_{c2}$ limit the plaquette phase projected onto $J_1 - J_2$ plane.](image1)

![FIG. 4: Comparison between the triplet gap at $k_{c} = (0,0)$ obtained by means of plain series and a particular Dlog-Padé approximant along selected straight line paths in couplings space, for a case in which the triplet gap closes at $J_1 = 1$. Results from reintegrated Dlog-Padé(3,1), plain series at $O(4)$ and $O(5)$ are shown with solid, dot-dashed and dashed lines, respectively. For $J_1 \lesssim 0.5$ the agreement is very good in all cases. Closer to $J_1$-$J_2$-$J_3$ model, i.e. to $J_1 = 1$, clear differences between the Dlog-Padé and the plain series arise.](image2)
Technically, the critical lines of Fig.3 have been obtained using Dlog-Padé analysis, rather than the plain series. This is known to improve the accuracy of locating the critical points significantly. For details on this technique we refer the reader to the literature [29]. In order to work with single variable Dlog-Padés we have scanned the exchange coupling space by means of straight lines, parametrized according to \((J_1, J_2 = b J_1, J_3 = c J_1)\). For fixed values of \(b\) and \(c\) this amounts to a single variable, i.e. \(J_1\).

To assess the impact of the Dlog-Padé analysis, we show its result for \(\omega(k_{c1})\), for a particular Dlog-Padé approximant and a case in which the triplet gap closes at \(J_1 = 1\) (Fig.4). A similar analysis has been done for all the critical lines calculated, including several Dlog-Padé approximants in each case. In this Figure, the solid line refers to the reintegrated Dlog-Padé \((3, 1)\), and the dot-dashed and dashed lines show the plain series at \(O(4)\) and \(O(5)\), respectively. From there it is clear that for \(J_1 \lesssim 0.5\) the agreement between the reintegrated Dlog-Padé and the SE at \(O(4)\) and \(O(5)\) is very good. In fact, all plots are indistinguishable on the scale used. This provides a qualitative measure of the convergence of the series. For \(J_1 \gtrsim 0.5\) and closer to criticality (at \(J_1 = 1\) in this case) however, we rely on the Dlog-Padé technique in order to describe the closure of the gap.

**PLAQUETTE PHASE AT \(J_1 = 1\)**

Here we analyze the extent of the plaquette phase on the \(J_2-J_3\) plane at \(J_1 = 1\), i.e. for the actual \(J_1-J_2-J_3\) model, written in units of \(J_1\). As it was mentioned in the Introduction, ED calculations for \(J_1-J_2-J_3\) model, using the complete Hilbert space and a restricted space of short-range dimer singlets, provide strong evidence for the existence of a plaquette phase around the line \(J_2 + J_3 = 1/2\) at \(J_1 = 1\), in particular for \(J_3 \geq J_2\) [22]. In this Section, we will extend this study by specifying the extension of this phase as obtained from SE. To this end, we proceed as in the previous Section, i.e. the critical lines are obtained by analyzing the closure of the triplet gap, i.e. solutions of \(\omega(k) = 0\).

In Fig.5 we show the corresponding results. The lower and upper critical lines mark the triplet softening at \(k_{c1}\) and \(k_{c2}\), respectively, and enclose the region of a finite triplet gap. I.e. this region refers to the plaquette phase, labeled by ‘P’. In this Figure, Dlog-Padé approximants are depicted by solid lines, and the results obtained by employing the \(O(5)\) plain series by dashed lines. We note that the critical lines shown from Dlog-Padé approximants in Fig.4 are consistent with the pairs of \((J_{2c}, J_{3c})\) at \(J_1 = 1\) shown in Fig.3.

Although, as in the previous Section, we base our results on the Dlog-Padé analysis, the agreement between the plain series and the Dlog-Padé approximants can be used to assess the convergence of the series. From Fig.5, it is clear that the best agreement for the lower critical line is found in the intermediate region, i.e where \(0.1 \lesssim J_2 \lesssim 0.2\).

For the special case of \(J_2 = 0\), i.e. for the pure \(J_1-J_3\) model, it has been conjectured that the classical critical line to Néel phase at \(J_3/J_1 = 0.25\) \((J_1 = 1\) in our case) should be shifted to larger values in the quantum model [21]. For all Dlog-Padé analyzed, our results confirm this conjecture, as e.g. for the \((3, 1)\) Dlog-Padé approximant of the lower critical line shown in Fig.5.

In conclusion we find that the plaquette phase extends considerably around the straight line of maximal frustration, connecting \(J_2 = 0, J_3 = 0.5\) with \(J_2 = J_3 \approx 0.25\), which was studied in Ref. [22]. In particular, as it can be seen in Fig.5 the upper critical line is rather far from the line of maximal frustration. Additionally, in the limiting case \(J_3 = 0\), we remain with the \(J_1-J_2\) model. For the latter, and as shown in the right lower corner of Fig.5 and unlike the plain series, the Dlog-Padé analysis suggests that the critical line does not intersect the \(J_2\) axis. I.e. we find no stability of the plaquette phase. This is in agreement with the SE results of Ref. [14]. Yet, the proximity between the critical line and the \(J_2\) axis calls for caution on this finding with respect to the convergence of the SE in this parameter range.
CONCLUSIONS

To summarize, using series expansion, based on flow equations we have analyzed the zero temperature properties of the 2D spin-1/2 $J_1$-$J_2$-$J_3$ AFM. Starting from the limit of decoupled plaquettes of a generalized $J_1$-$J_2$-$J_3$ model we have evaluated three-parameter series up to $O(5)$ in the interplaquette exchange couplings $J_1$, $J_2$ and $J_3$ for the ground state energy and for the triplet dispersion.

We find a rather large range of $J_{1,2,3}$ couplings which adiabatically connects to the state of isolated plaquettes and hosts a plaquette phase which is stable against second order quantum phase transitions into magnetic states. Our findings corroborate and enhance related predictions of Mambrini et al. 22 on the location of a stable plaquette phase at $J_1=1$.

For the particular case of the $J_1$-$J_2$ model at $J_3=0$, and consistently with results obtained in Ref. 14, our calculation predicts that the plaquette phase is not stable in the parameter range which we have investigated. However, higher order series expansions seem very desirable to render such results more reliable. In particular, from our series we are reluctant to draw any definite conclusions about the controversial region $J_3=0$ and $0.4 \lesssim J_2 \lesssim 0.6$.

Finally, we emphasize that our analysis has been focused on the stability of the plaquette phase with respect to second order transitions driven by one-particle (triplet) excitations. Further instabilities, like first order transitions or level crossings of excited states, other than elementary triplets, could give rise to further reduction of the plaquette regime and have not been considered here. Along this line, the two-particle sector, which includes singlet excitations, may play a role that can be analyzed using our SE technique. This deserves future investigation.

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