Phase diagram of the fully frustrated transverse-field Ising model on the honeycomb lattice

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Motivated by the current interest in the quantum dimer model on the triangular lattice, we investigate the phase diagram of the closely related fully-frustrated transverse field Ising model on the honeycomb lattice using classical and semi-classical approximations. We show that, in addition to the fully polarized phase at large field, the classical model possesses a multitude of phases that break the translational symmetry which, in the dimer language, correspond to a plaquette phase and a columnar phase separated by an infinite cascade of mixed phases. The modification of the phase diagram by quantum fluctuations has been investigated in the context of linear spin-wave theory. The extrapolation of the semiclassical energies suggests that the plaquette phase extends down to zero field for spin 1/2, in agreement with the $\sqrt{12} \times \sqrt{12}$ phase of the quantum dimer model on the triangular lattice with only kinetic energy.

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I. INTRODUCTION

Quantum dimer models have emerged as one of the main paradigms in the investigation of quantum spin liquids. The Rokhsar-Kivelson (RK) quantum dimer model (QDM), which includes a potential interaction of amplitude $V$ between dimers facing each other and a kinetic term of amplitude $t$ flipping them around rhombic plaquettes, has recently attracted special attention. The main reason comes from the presence on the triangular lattice of a resonating valence bond (RVB) phase first discovered by Moessner and Sondhi and extensively studied since then using zero temperature Green’s function quantum Monte Carlo (GFQMC). Exact results have been obtained at the RK point ($V/t = 1$), where the sum of all configurations can be proven to be a ground state, and at $V > t$, where the non-flippable configurations are the ground states. Analytical results have also been obtained in the limit $V/t \to -\infty$, where columnar states have been shown to be selected. However, in the intermediate range below the RK point, most of what is known about the model is based on numerical simulations.

A closely related model for which a number of analytical results have already been obtained is the fully frustrated transverse field Ising model (FFTFIM) on the honeycomb lattice defined by the Hamiltonian:

$$H = -\frac{J}{S^2} \sum_{\langle i,j \rangle} M_{ij} S_i^z S_j^z - \frac{\Gamma}{S} \sum_i S_i^z,$$

where $\Gamma > 0$ is the transverse magnetic field, $J > 0$ is the coupling constant of the Ising interaction term, $\langle i,j \rangle$ denotes pairs of nearest neighbors on the honeycomb lattice, and $M_{ij} = \pm 1$ is such that for each hexagon of the lattice the number of antiferromagnetic bonds ($M_{ij} = -1$) is odd, different choices of $M_{ij}$ corresponding to the same model up to the rotation of some spins by $\pi$ around the $x$ axis. Transverse field Ising models have been the subject of intense investigations over the years. The relationship between the FFTFIM on a regular lattice and the QDM on the dual lattice has been first emphasized by Moessner, Sondhi and Chandra, who showed (see also Ref. 1) that, in the limit $\Gamma/J \to 0$, the FFTFIM on the honeycomb lattice maps onto the QDM on the triangular lattice with $t = \Gamma^2/J$ and $V = 0$. For the FFTFIM on the honeycomb lattice, they also carried out a Landau-Ginzburg analysis and identified four soft modes which, upon lowering $\Gamma/J$, simultaneously become gapless, leading to a surprisingly large unit cell of 48 sites. Details of this calculation have been reported later by Moessner and Sondhi in Ref. 8. These authors further conjectured that the translational symmetry breaking transition out of the paramagnetic phase of the FFTFIM at large $\Gamma/J$ provides a reasonable description of the transition between the RVB phase and the intermediate phase of the QDM on the triangular lattice.

Building on this conjecture, Misguich and one of the present authors have carried out a semiclassical investigation of the paramagnetic phase of the FFTFIM on the honeycomb lattice and have shown that the dispersion of the spin waves and their softening at the transition are in remarkable agreement with the dispersion of visons in the QDM on the triangular lattice and their crystallization transition as revealed by quantum Monte Carlo (QMC) simulations. However, the analysis of Ref. 9 has not covered the small $\Gamma/J$ parameter range.

In the present paper, we perform a systematic investigation of the FFTFIM on the honeycomb lattice in the complete parameter range $0 \leq \Gamma/J < +\infty$ with classical and semi-classical approximations. As we shall see, the classical phase diagram is much richer than expected, with an infinite number of different crystalline phases below the paramagnetic phase: a plaquette phase, a cascade of mixed phases, and a highly degenerate columnar phase. Quantum fluctuations have been treated within linear spin-wave theory, leading to a partial lifting of the
the dimer operator of the gauge theory defined by
\[ a \prod_{l \in (i)} \tau^x_{l[a]} = -1, \]  
for all \( a \), where \( a \) is a site of the triangular lattice, and the product over \( l[a] \) runs over the six links emanating from \( a \). For a compact discussion of the correspondence between the three models, see e.g. the introduction of Ref. 9.

The discussion of the ordered phases is simpler in the context of the gauge theory. Indeed, in the FFTFIM language, the actual orientation of the spins in a given state depends on the choice of the matrix \( M_{ij} \). By contrast, the dimer operator of the gauge theory defined by
\[ d_i = \frac{1}{2}(1 - \tau^x_i), \]
translates into
\[ d_{ij} = \frac{1}{2} \left( 1 - M_{ij} \frac{S^z_i S^z_j}{S^z} \right), \]
in the Ising language, and its expectation value does not depend on the choice of \( M_{ij} \). Another advantage of the gauge-invariant language is that it allows to make a direct comparison with the numerical results obtained on the QDM since they live on the same lattice and are defined in terms of the same link operators. So, while all reasonings and calculations will be performed in the context of the FFTFIM, the only formulation adapted to the semiclassical approach, the structures of different ordered phases will be also described in gauge-invariant terms. Throughout the paper, we use only gauges in which each hexagon of the lattice contains exactly one antiferromagnetic bond (with \( M_{ij} = -1 \)) and five ferromagnetic bonds (with \( M_{ij} = +1 \)). Most results will be presented for the simplest periodic arrangement of the antiferromagnetic bonds shown in Fig. 1. However, this choice of gauge does not always lead to the smallest possible unit cell in terms of the spin representation. Thus, we will also introduce other gauges whenever this is helpful.

The paper is organized as follows. In section II, we concentrate on the limit \( \Gamma/J \ll 1 \), which has not been considered in Refs. 1, 7–9, and we show that columnar phases reminiscent of the \( V \rightarrow -\infty \) limit of the QDM are stabilized. In section III, we revisit the vicinity of the RVB phase. We recover the symmetry predicted by the Landau-Ginzburg approach of Refs. 7, 8 and by the spin-wave analysis of Ref. 9, but we find that the bonds with the largest dimer density form separate 4-site rhombic plaquettes instead of having a uniform distribution inside a 12-site unit cell as reported in Ref. 9. The reasons for this discrepancy are explained in subsection III C. In section IV, we discuss the transition between the plaquette phase and the columnar phase and show that they are separated by a region of intermediate phases of mixed character. The stability of these phases with respect to quantum fluctuations and the semi-classical phase diagram are discussed in section V. The paper ends with a short conclusion in section VI.

II. COLUMNAR PHASE

In this section we discuss the properties of the model when \( \Gamma/J \) is small. The argument proceeds in three steps. First we determine the ground state manifold of the Heisenberg model with purely Ising-like interactions in the absence of magnetic field \( (\Gamma = 0) \). Then we investigate how the extensive degeneracy of these ground states is lifted by a small transverse field. Finally, we discuss the effect of quantum fluctuations in the context of linear spin-wave theory.

A. Zero transverse field

In the absence of a transverse magnetic field \( (\Gamma = 0) \), we are left with a model without quantum fluctuations in which the interaction term couples only the \( z \) components of neighboring spins on the honeycomb lattice. With our choice of gauge, one bond on each hexagon is antiferromagnetic \( (M_{ij} = -1) \) and the others are ferromagnetic \( (M_{ij} = 1) \). Frustration is present since it is clearly impossible to minimize the energy of all bonds of a given hexagon.

For Ising spins, i.e. spins which can only point up or down along the \( z \) direction, the best one can do is to satisfy five bonds leaving one unsatisfied. This can be
done in six different ways according to which bond is not satisfied ("frustrated") and the resulting energy is $-4J$. Up to a global reversal of the spins, a ground state is characterized by the distribution of frustrated bonds such that there is exactly one of them per hexagon.

For three-dimensional vectors of norm $S$, the situation is slightly more subtle because the twelve Ising configurations with all spins parallel or antiparallel to $z$ axis are not the only ground states of a single hexagon. To see this, let us consider a single hexagon and investigate the possibility of a given spin $i$ not to be directed along $z$. The variation of the energy of the hexagon

$$E_{\text{hex}} = -J \sum_{j=1}^{6} M_{i,j+1} \cos \theta_j \cos \theta_{j+1}$$  \hspace{1cm} (6)

(where the angle $\theta_j$ parameterizes the deviation of spin $j$ from the $z$ axis) with respect to $\theta_i$ leads to the condition

$$M_{i-1,i} \cos \theta_{i-1} + M_{i,i+1} \cos \theta_{i+1} = 0.$$  \hspace{1cm} (7)

If this condition is satisfied, the terms in Eq. (6) which depend on $\theta_i$ drop out, so that one is left with the energy of an open chain of five spins. In an open chain one can trivially minimize the energy of each bond by choosing $\cos \theta_{j+1} = M_{j,j+1} \cos \theta_j = \pm 1$ which leads to $E = -4J$ and to the automatic fulfillment of condition (7), leaving $\theta_i$ arbitrary. Note that this argument excludes a deviation from the $z$ axis of more than one spin, since the energy of a five-spin open chain cannot be as low as $-4J$ if not all five spins are along $z$. So, for three-dimensional spins, the energy of a single hexagon is minimal as soon as it is minimal for four consecutive bonds, and the spin at the remaining site can have any direction.

It is natural to ask whether this additional freedom increases the degeneracy of the ground state manifold of the continuous model in comparison with the case of Ising spins. To demonstrate that this is not the case, let us assume that at site $i$ the spin is not along $z$. To minimize simultaneously the energy of the three hexagons to which it belongs, three conditions of the form (7) must be fulfilled:

$$Y_1 + Y_2 = 0,$$
$$Y_2 + Y_3 = 0,$$
$$Y_3 + Y_1 = 0,$$  \hspace{1cm} (8)

where $Y_a = M_{i,a} \cos \theta_{i,a} = \pm 1$ (with $a = 1, 2, 3$) and $i_a$ are the three nearest neighbors of site $i$. It is evident that the restriction $Y_a = \pm 1$ does not allow all three equations (8) to be satisfied simultaneously. Therefore, it is impossible for any spin not to point along $z$, and the ground state manifold coincides with that of the frustrated Ising model with the same lattice, i.e. it consists of all Ising configurations with one frustrated bond per hexagon. Each of these states is a local minimum of the Hamiltonian.

### B. Classical ground states in small transverse field

Let us now switch on a small transverse field and study how the local minima of the classical Hamiltonian evolve upon increasing the field. Since the field is along $x$, the spins are expected to acquire a small $x$ component, and to describe the spin configuration evolving from a given ground state of the pure Ising case, we use the parametrization:

$$S_i^x = S \sin \theta_i,$$
$$S_i^y = \sigma_i S \cos \theta_i,$$  \hspace{1cm} (9)

where $\sigma_i = \pm 1$ is the sign of $S_i^z$ and is determined by the ground state of the pure Ising case around which we expand. In terms of the gauge-invariant bond variable

$$\tau_{ij} = M_{ij} \sigma_i \sigma_j,$$

which is equal to $-1(+1)$ if the bond $\langle i, j \rangle$ is frustrated (not frustrated), the classical energy can be rewritten as

$$E = -J \sum_{\langle i, j \rangle} \tau_{ij} \cos \theta_i \cos \theta_j - \Gamma \sum_i \sin \theta_i.$$  \hspace{1cm} (10)

In the limit $\Gamma \ll J$ the deviations from the $z$ direction are small, and the classical energy can be expanded in the variables $\theta_i$ around $\theta_i = 0$. To second order, the interaction term in Eq. (10) decouples:

$$\tau_{ij} \cos \theta_i \cos \theta_j \approx \tau_{ij} (1 - \theta_i^2/2 - \theta_j^2/2).$$

Now, for any ground state of the pure Ising case, the set $\{ \tau_{ij} \}$ is such that only one bond in each hexagon is frustrated. Therefore each site belongs at most to one frustrated bond. If we denote by $F$ (resp. $NF$) the set of what we call below frustrated (resp. non frustrated) sites, namely, the sites belonging to one frustrated bond (resp. no frustrated bond), the energy up to second order can be rewritten:

$$E^{(2)} = E_{\Gamma=0} + \sum_{i \in F} \left( \frac{J}{2} \theta_i^2 - \Gamma \theta_i \right) + \sum_{i \in NF} \left( \frac{3J}{2} \theta_i^2 - \Gamma \theta_i \right).$$  \hspace{1cm} (11)

Minimizing $E^{(2)}$ with respect to $\{ \theta_i \}$ leads to

$$\theta_i = \begin{cases} \Gamma/J & \text{for } i \in F, \\ \Gamma/3J & \text{for } i \in NF. \end{cases}$$  \hspace{1cm} (12)

Since the number of frustrated and non frustrated sites is the same for all ground states, the energy up to second order in $\theta_i$ is the same in all ground states. So, second order corrections do not lift the degeneracy. They only induce a difference in orientation between the spins which belong to a frustrated bond and those which do not.

So to lift the degeneracy we have to push the expansion in $\theta_i$ to higher orders. To fourth order, it reads:

$$E^{(4)} = E_{\Gamma=0} + \sum_{i \in F} \left[ J \left( \frac{\theta_i^2}{2} - \frac{\theta_i^4}{4!} \right) - \Gamma \left( \theta_i - \frac{\theta_i^3}{3!} \right) \right]$$
$$+ \sum_{i \in NF} \left[ 3J \left( \frac{\theta_i^2}{2} - \frac{\theta_i^4}{4!} \right) - \Gamma \left( \theta_i - \frac{\theta_i^3}{3!} \right) \right]$$
$$- \frac{J}{4} \sum_{\langle i, j \rangle} \tau_{ij} \theta_i^2 \theta_j^2.$$  \hspace{1cm} (13)
From the previous discussion, we know that the values of \( \theta_i \), minimizing the energy to order \( O(\theta^2) \) are given by Eq. (12). Injecting these solutions into the fourth order expansion of the energy, we notice that the terms \( \theta_i^3 \) and \( \theta_i^4 \) only contribute in two different ways depending on the type of site (frustrated or non frustrated). They will thus not lift the degeneracy. By contrast, the crossed terms \( \tau_{ij} \theta_i^2 \theta_j^2 \) contribute in four different ways depending on the environment of the sites \( i \) and \( j \). The four cases are illustrated in Fig. 2.

The contributions of the fourth order crossed terms to the energy for the different configurations in units of \( \Gamma^4/4J^3 \) are +1 for 2(a), \(-\frac{3}{4}\) for 2(c), \(-\frac{1}{8}\) for 2(b) and \(-1\) for 2(d). Since these energies are not equal, these crossed terms are expected to lift the degeneracy, at least partially.

The total contribution of the fourth order crossed terms of the energy can then be written as:

\[
-\frac{\Gamma^4}{4} \sum_{i,j} \tau_{ij} \theta_i^2 \theta_j^2 \approx \frac{\Gamma^4}{4} \left( \left( \frac{\beta}{J} \right)^4 \left( N_a - N_d - \frac{1}{4} N_c - \frac{1}{4} N_b \right) \right) \approx \frac{\Gamma^4}{4} \left( \left( \frac{\beta}{J} \right)^4 \left( \frac{2}{41} N_{\text{hex}} - \frac{64}{81} N_d \right) \right).
\]

This contribution is a decreasing function of \( N_d \), so the lowest energy will be reached for the largest possible value of \( N_d \). Now, since there is only one frustrated bond per hexagon, \( N_d \) cannot exceed the number of frustrated bonds, \( N_a = N_{\text{hex}}/2 \). This upper limit is reached for configurations in which all the frustrated bonds are organized into chains of alternating frustrated and non frustrated bonds (see examples in Fig. 3).

In columnar states Eqs. (14) and (15) fix both \( N_b \) and \( N_c \) to be equal to \( N_{\text{hex}} \).

So, the fourth order contribution to the energy partially lifts the degeneracy and selects the family of columnar states. A priori, higher orders might further lift the degeneracy. That this is not the case is best seen by constructing the exact local minima that correspond to columnar states. We start by rewriting the energy:

\[
E = -\sum_{i \in \text{NF}} \left[ \frac{J}{2} \cos \theta_i \left( \cos \theta_{i_1} + \cos \theta_{i_2} + \cos \theta_{i_3} \right) + \Gamma \sin \theta_i \right] -\sum_{j \in \text{F}} \left[ \frac{J}{2} \cos \theta_j \left( -\cos \theta_{j_1} + \cos \theta_{j_2} + \cos \theta_{j_3} \right) + \Gamma \sin \theta_j \right],
\]

where \( i_1, i_2, i_3 \) (resp. \( j_1, j_2, j_3 \)) are the three neighbors of site \( i \) (resp. \( j \)), and the frustrated bond is taken to be between sites \( j \) and \( j_1 \). To minimize the energy, the set of angles \( \{ \theta_i, \theta_j \} \) must be a solution of the equations:

\[
\frac{\partial E}{\partial \theta_i} = J \sin \theta_i \left( \cos \theta_{i_1} + \cos \theta_{i_2} + \cos \theta_{i_3} \right) - \Gamma \cos \theta_i = 0,
\]

\[
\frac{\partial E}{\partial \theta_j} = J \sin \theta_j \left( -\cos \theta_{j_1} + \cos \theta_{j_2} + \cos \theta_{j_3} \right) - \Gamma \cos \theta_j = 0.
\]

Now, in columnar structures, all frustrated sites have identical environments (with exactly two frustrated neighbors) and all unfrustrated sites also have identical environments (with exactly one frustrated neighbor). So, if the angles \( \theta_1 \) and \( \theta_2 \) satisfy the equations:

\[
J \sin \theta_1 \left( 2 \cos \theta_1 + \cos \theta_2 \right) - \Gamma \cos \theta_1 = 0,
\]

\[
J \sin \theta_2 \cos \theta_1 - \Gamma \cos \theta_2 = 0,
\]

then the set of angles

\[
\theta_i = \begin{cases} 
 \theta_1 & \text{for } i \in \text{NF} \\
 \theta_2 & \text{for } i \in \text{F} 
\end{cases}
\]

is a solution of Eqs. (18). The non trivial solutions of Eqs. (19) describing the evolution of columnar states with the change of \( \Gamma/J \) are given by:

\[
\sin \theta_1 = \frac{\sin(\beta/3)}{\cos(\beta/3)} , \quad \sin \theta_2 = \frac{\sin(\beta)}{\cos(\beta/3)} ,
\]
where \( \tan \beta = \Gamma / J \).

The substitution of Eq. (20) into Eq. (10) shows that the classical energy of a columnar state is given by
\[
E_{\text{col}} = -\frac{N}{2} \left[ J \cos \theta_1 (\cos \theta_1 + \cos \theta_2) + \Gamma (\sin \theta_1 + \sin \theta_2) \right],
\]
(22)
where \( N \) is the total number of sites. Naturally, the variation of \( E_{\text{col}} \) with respect to \( \theta_1 \) and \( \theta_2 \) reproduces Eqs. (19) which we used to find the values of \( \theta_1 \) and \( \theta_2 \). In order to verify that it never becomes more advantageous to minimize \( N_d \) rather than to maximize it, we have studied also the solutions with \( N_d = 0 \) and checked that for any relation between \( \Gamma \) and \( J \) they have higher energy than the columnar states (see Appendix A).

A convenient classification of columnar states can be introduced by describing them in terms of zero-energy domain walls formed on the background of the simplest columnar state, an example of which is shown in Fig. 3(a). Below we call it the 1st columnar state. In this state all frustrated bonds have the same orientation and form straight columns shown in the figure by the shading. In terms of Fig. 3 the walls of the first type are horizontal and take place whenever the orientation of the frustrated bonds changes from left to right. The 2nd columnar state (Fig. 3(b)) corresponds to the configuration having the highest possible density of such domain walls.

The domain walls of the second type are perpendicular to the frustrated bonds, and correspond to changing the orientation not of frustrated bonds but of columns. The 3rd columnar state (Fig. 3(c)) is the configuration having the highest possible density of walls of the second type as the orientation of the columns changes at every frustrated bond. Other columnar states having the same classical energy can be obtained by introducing arbitrary sequences of parallel domain walls either of the first or of the second type separating domains of the 1st columnar state. An analogous classification of columnar states had earlier been introduced by Moessner and Sondhi in terms of the QDM.

Fig. 4(a) presents a plot of the dimer density for the 1st columnar state at \( \Gamma / J = 1.5 \). The bonds of the dual triangular lattice having the highest dimer densities are organized into a columnar pattern. Fig. 4(b) is a plot of the 1st columnar state in the classical spin model. The \( S^z \) component of the spin on frustrated sites (green arrows in Fig. 4(b)) is smaller than that on non frustrated sites.

\[
\begin{align*}
S^x_i &= \sigma_i \cos \theta_i, S^y_i = \sin \theta_i, \\
S^z_i &= -\sin \theta_i, S^z_i = \cos \theta_i.
\end{align*}
\]
(23)

in such a way that the Hamiltonian expressed in terms of the variables \( S^x \) and \( S^z \) has a ferromagnetic ground state.

Mapping the new spin operators to Holstein-Primakoff bosons in the harmonic limit:
\[
S^x_i = \sigma_i, \quad S^z_i \approx \sqrt{S / 2} (a_i + a_i^\dagger),
\]
(24)
sitions of frustrated and non frustrated bonds forming main walls of the first type interchanges only the po-
for all columnar states having the same sets of frustrated and non frustrated bonds is proportional to \(1/J\).
The vectors \(\vec{a}\) and \(\vec{b}\) are shown in Fig. 4(b). The unit cell is defined by the two vectors \(\vec{a}\) and \(\vec{b}\) with \(|\vec{b}| = \sqrt{3}|\vec{a}|\).

then yields the quadratic Hamiltonian:

\[
H = E_{\text{col}} + \gamma_1 \sum_{i \in \mathcal{F}} a_i^\dagger a_i + \gamma_2 \sum_{i \in \mathcal{F}} b_i^\dagger b_i - \frac{J}{2S} \sum_{(i,j)} M_{ij} \sin \theta_i \sin \theta_j \left[ a_i^\dagger a_j + a_j^\dagger a_i + \text{h.c.} \right],
\]

where

\[
\gamma_1 = (1/S)[J \cos \theta_1(2 \cos \theta_1 + \cos \theta_2) + \Gamma \sin \theta_1],
\gamma_2 = (1/S)[J \cos \theta_1 \cos \theta_2 + \Gamma \sin \theta_2],
\]

and \(E_{\text{col}}\) is the classical energy of a columnar state. Eq. (25) can be reduced to a gauge-invariant form (with \(M_{ij}\) replaced by \(\tau_{ij}\)) by replacing \(a_i\) by \(\sigma_i a_i\) and \(a_i^\dagger\) by \(\sigma_i a_i^\dagger\) in Eqs. (24). However, we use Eq. (25) in the following because it allows an easy proof that domain walls of the first type do not change the energy of the harmonic fluctuations.

It is evident that for \(\theta_i\) given by Eq. (20), the expression in the right-hand side of Eq. (25) is exactly the same for all columnar states having the same sets of frustrated and non frustrated sites. Since the introduction of domain walls of the first type interchanges only the positions of frustrated and non frustrated bonds forming straight columns, but does not change the positions of frustrated sites [see Fig. 3(a) and Fig. 3(b)], the expression in the right-hand side of Eq. (25) will be exactly the same for all columnar states which can be transformed one into another by the introduction of some number of domain walls of the first type. This proves that the contribution of the harmonic fluctuations to the energy is the same for all members of the family of columnar states having only domain walls of the first type.

After partitioning the honeycomb lattice into four sublattices in accordance with the structure of the unit cell shown in Fig. 4(b) and performing on each sublattice the Fourier transformation with wavevector \(\vec{q}\), the quadratic bosonic Hamiltonian of the 1st columnar state is reduced to the form

\[
H = E_{\text{col}} + N \sum_{\vec{q}} \langle \hat{a}_{\vec{q}}^\dagger \hat{H}(\vec{q}) \hat{a}_{\vec{q}} - (\gamma_1 + \gamma_2) \rangle
\]

In this expression, \(\hat{a}_{\vec{q}}\) is an eight-component vector \((a_{-\vec{q},1}, a_{-\vec{q},2}, a_{-\vec{q},3}, a_{-\vec{q},4}, a_{\vec{q},1}, a_{\vec{q},2}, a_{\vec{q},3}, a_{\vec{q},4})\), where \(a_{\vec{q},n}\) are the bosonic operators with wavevector \(\vec{q}\) acting on the \(n\)th sublattice, and \(\hat{H}(\vec{q})\) is an \(8 \times 8\) hermitian matrix given by:

\[
\hat{H}(\vec{q}) = \frac{1}{2} \begin{pmatrix}
\gamma_2 & \mu^* & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & \gamma_1 & \mu & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & \gamma_2 & \mu^* & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \gamma_1 & \mu & 0 & 0 & 0 \\
\mu & 0 & 0 & 0 & \gamma_2 & \mu^* & 0 & 0 \\
0 & \mu^* & 0 & 0 & 0 & \gamma_1 & \mu & 0 \\
0 & 0 & \mu & 0 & 0 & 0 & \gamma_2 & \mu^* \\
0 & 0 & 0 & \mu^* & 0 & 0 & 0 & \gamma_1
\end{pmatrix}
\]

where

\[
\mu \equiv \mu(\vec{q}) = -\frac{J \sin^2 \theta_2}{2S} (-1 + e^{i\vec{q}\vec{a}}),
\eta \equiv \eta(\vec{q}) = -\frac{J \sin \theta_1}{2S} (1 + e^{i\vec{q}\vec{a}}),
\delta \equiv \delta(\vec{q}) = \tau e^{i\vec{q}\vec{b}}, \quad \tau = -\frac{J \sin \theta_1 \sin \theta_2}{2S}.
\]

The vectors \(\vec{a}\) and \(\vec{b}\) are shown in Fig. 4(b).

As discussed above, the harmonic Hamiltonian is the same for the whole family of columnar states constructed by introducing an arbitrary number of domain walls of the 1st type. This family includes for instance the 2nd columnar state. In the harmonic approximation, all these states have the same quantum corrections to the energy, therefore to order \(1/S\) the degeneracy is not lifted. Note however that the absence of degeneracy lifting for this family of states at the harmonic level is not related to a symmetry of the original Hamiltonian. So we expect this degeneracy to be removed if one goes beyond the harmonic approximation, and higher order terms are expected to select either the 1st or the 2nd columnar state depending on whether the energy of a domain wall of the first type is positive or negative. However the effect of anharmonicities has not been investigated in this
work. Note that a similar effect, namely the incapacity of harmonic fluctuations to fully lift a well-developed accidental degeneracy of the ground states, has already been reported for various other models (in particular, with kagomé,\textsuperscript{12–14} honeycomb,\textsuperscript{15} dice\textsuperscript{16} and pyrochlore\textsuperscript{17} lattices).

By contrast, the 3\textsuperscript{rd} columnar state is described by a different harmonic Hamiltonian which is not written down here explicitly because the number of sites per unit cell, hence the linear dimension of the matrix $H(\vec{q})$, is twice as large, so that the matrix $H(\vec{q})$ is $16 \times 16$. The energy of zero point fluctuations in this state turns out to be higher than in the 1\textsuperscript{st} columnar state (see Fig. 5). This suggests that domain walls of the second type have a positive energy.

To support this statement, we have applied the same reasoning as used in Ref. 18 for the investigation of the frustrated XY model on a triangular lattice and have considered the 4\textsuperscript{th} columnar state (Fig. 3(d)) which differs from the 3\textsuperscript{rd} one in that the density of domain walls of the second type is exactly half as large. Fig. 5 compares the numerically calculated differences between the value of the quantum corrections to the energies of the 2\textsuperscript{nd}, 3\textsuperscript{rd} and 4\textsuperscript{th} columnar states and its value for the 1\textsuperscript{st} columnar state. In particular, the inset in Fig. 5 presents the ratio of these quantities for the 3\textsuperscript{rd} and 4\textsuperscript{th} states. This ratio is very close to two, supporting the suggestion that the fluctuation induced corrections to the energy are essentially proportional to the density of domain walls of the second type.

Upon increasing $\Gamma / J$, the classical states remain locally stable until soft-modes appear in the spin-wave dispersion. For all columnar states without domain walls of the second type this takes place at $\Gamma / J \approx 2.004$, and for the 3\textsuperscript{rd} columnar state at $\Gamma / J \approx 2.373$. To summarize, harmonic fluctuations partially lift the degeneracy of the classical ground state manifold in favor of the columnar states having only domain walls of the 1\textsuperscript{st} type.

## III. PLAQUETTE PHASE

### A. Soft modes and the ground state periodicity

In the limit $J = 0$ the Hamiltonian consists simply of a coupling to the transverse magnetic field $\Gamma$, and the classical ground state is completely polarized with all spins aligned along the magnetic field in the $x$ direction. The same state minimizes the classical energy for sufficiently large ratio $\Gamma / J$. With the choice of gauge of Fig. 1, the unit cell of this state contains 4 sites (see Fig. 6).

The analysis of Refs. 7–9 indicates that the polarized phase becomes unstable at $\Gamma = \Gamma_c = \sqrt{6}J$. At this value of the field, soft modes appear in the dispersion relation at momenta $(q_x, q_z) = \pm (\frac{\pi}{6\sqrt{3}}, \frac{\pi}{2})$ and $(q_x, q_z) = \pm (\frac{5\pi}{6\sqrt{3}}, \frac{\pi}{2})$, triggering a second order transition to a new phase whose periodicity can be determined from the $\vec{q}$ points corresponding to the soft modes.

Since any linear combination of these four modes is invariant under translations by vectors $(3\vec{a} - \vec{b})$ and $4\vec{b}$, a state associated with them should have the periodicity in real space imposed by these two vectors that define a unit cell containing 48 sites of the honeycomb lattice (Fig. 7(b)). Moreover, since any linear combination of the four soft modes under the translation by $2\vec{b}$ just changes sign, this cell should allow a division into two halves which in the spin representation differ from each other only by the reflection of all spins about the $x$ axis but in terms of gauge-invariant variables are identical.

There exists a possibility to make these two halves really identical in terms of spin representation as well just by choosing a different gauge shown in Fig. 7(c). In this gauge a state related to the soft modes listed above is periodic with a 24-site unit cell defined, for example, by vectors $3\vec{a} - \vec{b}$ and $2\vec{b}$. However, if one uses the simplest gauge of Fig. 1 and imposes periodic boundary conditions along the $x$ and $z$ directions, the periodicity dictated by the wave vectors of the soft modes requires to use a cell

![Fig. 6: (Color online) In the polarized state all spins are aligned along the magnetic field. The unit cell of this state is the same as that of the 1\textsuperscript{st} columnar state: it is defined by the vectors $\vec{a}$ and $\vec{b}$.](image-url)
of size $12\vec{a} \times 4\vec{b}$ that contains 192 sites of the honeycomb lattice.  

B. Numerical minimization of energy

The minimization of the classical energy using Mathematica minimization routines for the 192-site system with periodic boundary conditions have confirmed that the real periodicity of the classical ground state in the gauge of Fig. 1 is determined by a 48-site unit cell which can be divided into two halves in such a way that the second half differs from the first one by the reflection of all spins about the $x$ axis. Inside the cell one finds a pattern of six different orientations of the spins as well as their reflections about the direction of the field.

The structure of the state minimizing the classical energy is shown in figure 7(b). The radii of the circles are proportional to the absolute value of the $z$ component of the spins $|S^-|$, and the different signs of $S^z$ are kept track of by plotting full and empty circles. $S^z$ is not plotted but is always positive since the spins tend to align with the magnetic field. The size of the elementary cell can be reduced to 24 sites by choosing the gauge depicted in Fig. 7(c) by zigzagged bonds. In this gauge the sign of $S^z$ is the same for all spins and the spin pattern is centered on one of the sites of the honeycomb lattice.

Naturally, it is even more convenient to discuss the structure of an ordered state in terms of gauge-invariant dimer density $d_{ij}$ defined by Eq. (5). In the polarized phase (at $\Gamma/J > \sqrt{6}$), $S^- = 0$ for all sites $i$, so that the dimer density is uniform and equal to $\frac{1}{2}$ on all bonds. Below the critical magnetic field, $\Gamma_c = \sqrt{6}J$, the dimer density on many bonds becomes smaller than $\frac{1}{2}$. For the pattern of $d_{ij}$, the two halves of the 48-site elementary cell are identical because the dimer pattern is conserved when reversing the sign of $S^z$ for all spins. Accordingly, the elementary cell corresponds to 24 sites of the honeycomb lattice or to 12 sites of the triangular lattice dual to it. In other terms, the periodicity of the dimer density pattern is the same as in the $\sqrt{12} \times \sqrt{12}$ phase found around $V/t = 0$ in the QDM on the triangular lattice.1,4

In Fig. 7(a) the elementary cells are represented by the large hexagons. Since inside an elementary cell the dimer density plot displays a pattern of four-site plaquettes having the highest dimer density (see Fig. 7(a)), following the convention adopted in the QDM literature19 we refer to this phase as the plaquette phase. This phase is the analog of the $\sqrt{12} \times \sqrt{12}$ phase found around $V/t = 0$ in the QDM.1

Note that the dimer density plot obtained below $\Gamma/J = \sqrt{6}$ in our calculation (Fig. 7(a)) differs significantly from the one presented in Ref. 9. The two plots have the same symmetry, $P31m$, but the pattern of Ref. 9 does not reveal four-site plaquettes. In fact, the difference can be traced back to the fact that the solution of Ref. 9 was obtained by a variational calculation in the subspace of linear combinations of the four soft modes (which minimize the sum of the second and fourth order contributions to the classical energy), whereas the present solution was obtained by assuming that the soft modes dictate only its periodicity. The reason why the two solutions do not have the same asymptotic form when $\Gamma/J$ tends to $\sqrt{6}$ from below is detailed below in Sec.

FIG. 7: (Color online) (a) The dimer density $d_{ij}$ in the plaquette phase at $\Gamma/J = 2$. The thick blue bonds corresponding to the highest density ($d_{ij} = \frac{1}{2}$) are organized into 4-site rhombic plaquettes. On all other bonds the dimer density satisfies $0 < d_{ij} < \frac{1}{2}$, the thickness of the bonds being proportional to $d_{ij}$.
(b) The spin configuration in the same state in the gauge of Fig. 1. The radii of the circles are proportional to $|S^-|$, while positive and negative values of $S^z$ are represented as full and empty circles. The green dashed rectangle shows the 48-site unit cell ($3\vec{a} - \vec{b}) \times 4\vec{b}$. It can be split into two halves which differ from each other by the sign of $S^z$.
(c) The same spin configuration in the gauge that leads to a 24-site unit cell (large hexagon). As before antiferromagnetic bonds fixing the gauge are depicted as zigzag bonds. The sites at which the classical spins have the same values of $S^z$ are labelled with the same number. Note the existence of 6 sites with $S^z = 0$.\n
The degeneracy of the plaquette phase is equal to 48 in terms of the spin representation and to 24 in terms of the dimer representation. Each of the 24 equivalent dimer patterns [one of which is shown in Fig. 7(a)] corresponds to two spin configurations which can be transformed one into the other by changing the sign of $S^z_i$ for all spins.

The local stability of the plaquette phase with respect to quantum fluctuations has been investigated within the gauge of Fig. 7(c) to reduce the hermitian matrix of the quadratic bosonic Hamiltonian to a $48 \times 48$ matrix. The plaquette phase has been found to be stable in the domain $1.64 < \frac{J}{\Gamma} < \sqrt{6}$ with soft modes appearing at $\vec{q} = 0$ when $\frac{J}{\Gamma} \approx 1.64$.

C. Analytical study of the critical region below $\Gamma_c$

In this subsection it will be convenient instead of Eq. (9) to use a different parametrisation of the classical spins of norm $S$,

$$
S^x_i = S \sqrt{1 - \rho^2_i},
S^z_i = S \rho_i.
$$

(31)

In the asymptotic regime where the transverse field $\Gamma$ dominates over nearest-neighbor interactions, we are in the polarized phase with $S^x_i = 0$ ($\rho_i = 0$). Upon decreasing the transverse field the components $S^x_i$ are expected to deviate from zero. To sixth order in the $\rho_i$’s, the classical energy of the model is given by

$$
E = -J \sum_{\langle i,j \rangle} M_{i,j} \rho_i \rho_j - \Gamma \sum_i \left(\frac{\rho_i^2}{2} - \frac{\rho_i^4}{8} - \frac{\rho_i^6}{16} - \ldots\right),
$$

(32)

Let us denote by $\rho_{\vec{r},n,n'} = \sum_{\vec{q}} \rho_{\vec{q},n} e^{i\vec{q}\cdot\vec{r}}$ with $n = 1, \ldots, 4$ the values of $\rho_i$ on the four sublattices (see Fig. 6). Since $\rho_{\vec{r},n,n}$ is real, $\rho_{\vec{q},n} = \rho_{-\vec{q},n}^*$. The energy per site $\mathcal{E}$ is then given by

$$
\mathcal{E} = \mathcal{E}_{0} - \frac{J}{8} \sum_{n,n',\vec{q}} \rho_{-\vec{q},n} \left[ \hat{M}(\vec{q}) - \frac{\Gamma}{\frac{J}{2}} \right] \rho_{\vec{q},n'}
+ \frac{\Gamma}{32} \sum_{n,q_1,q_2} \left( \prod_{i=1}^{4} \rho_{\vec{q}_i,n} \right) \delta_{\vec{q}_1 + \vec{q}_2 + \vec{q}_3 + \vec{q}_4, \vec{q}}
+ \frac{\Gamma}{64} \sum_{n,q_1,q_2,q_3} \left( \prod_{i=1}^{6} \rho_{\vec{q}_i,n} \right) \delta_{\vec{q}_1 + \vec{q}_2 + \vec{q}_3 + \vec{q}_4 + \vec{q}_5 + \vec{q}_6, \vec{q}} + \ldots,
$$

(33)

where $\vec{q}$ is a vector belonging to the reciprocal lattice of the lattice defined by the vectors $\vec{a}$ and $\vec{b}$, and $\hat{M}(\vec{q})$ is the Fourier transform of the interaction matrix. The analysis of the second-order terms in (33) shows$^{7,8}$ that the paramagnetic solution $\rho_i = 0$ becomes unstable at $\Gamma / J = \sqrt{6}$ at the wavevectors $\vec{q}_A = \left( \frac{\pi}{\sqrt{3}|a|}, \frac{\pi}{2|b|} \right)$, $\vec{q}_B = \left( \frac{\pi}{6|a|}, \frac{\pi}{2|b|} \right)$, $-\vec{q}_A$, and $-\vec{q}_B$, indicating a transition to a phase of periodicity $(3a - \vec{b}) \times 4b$.

The approach of Ref. 9 consists in keeping in the energy functional (33) only the critical modes with $\vec{q} = \pm \vec{q}_A$ and $\vec{q} = \pm \vec{q}_B$ whose amplitudes are described by Fourier coefficients

$$
\hat{\rho}_{\vec{q}_A} = |\rho_A| e^{i\phi_A} \hat{u}_A, \quad \hat{\rho}_{\vec{q}_B} = |\rho_B| e^{i\phi_B} \hat{u}_B,
$$

(34)

where

$$
\hat{u}_A = \begin{pmatrix} 1 & e^{i\frac{\pi}{6}} & F e^{i\frac{\pi}{12}} & F e^{-i\frac{\pi}{12}} \end{pmatrix},
\hat{u}_B = \begin{pmatrix} F & F e^{i\frac{\pi}{12}} & e^{i\frac{\pi}{12}} & e^{-i\frac{\pi}{12}} \end{pmatrix}
$$

(35)

are the eigenvectors of $\hat{M}(\vec{q}_A)$ and $\hat{M}(\vec{q}_B)$ associated to the eigenvalue $\sqrt{6}$ and

$$
F = 2 \sin \frac{5\pi}{12} = \frac{1 + \sqrt{3}}{\sqrt{2}}.
$$

(36)

In the framework of this approach, $\mathcal{E}_0^{(4)}$, the sum of the second and fourth order contributions to Eq. (33), is given by:

$$
\mathcal{E}_0^{(4)} = -\frac{1}{4} (\Gamma_c - \Gamma)(1 + F^2) \left[ |\rho_A|^2 + |\rho_B|^2 \right]
+ \frac{4 \pi^2}{J} F^2 \left[ |\rho_A|^2 + |\rho_B|^2 \right]^2
$$

(37)

and depends only on $|\rho_A|^2 + |\rho_B|^2$. $^{7,8}$

The minimum of $\mathcal{E}_0^{(4)}$ is achieved when

$$
|\rho_A|^2 + |\rho_B|^2 = \frac{1 + F^2}{6 F^2} \Gamma_c, \quad \Gamma,
$$

(38)

from which it follows that, to leading order, $|\rho_A| \sim |\rho_B| \sim (\Gamma_c - \Gamma)^{\frac{1}{2}}$ and $\mathcal{E}_0^{(4)} \sim (\Gamma_c - \Gamma)^2$. However, condition (38) leaves both the ratio $|\rho_B|/|\rho_A|$ and the phases $\phi_A$ and $\phi_B$ completely undefined. To find them one has to consider also the sixth order term in Eq. (33),$^{8,9}$ which for the critical modes reduces to

$$
\mathcal{E}_0^{(6)} = \frac{5 \pi^2}{8} (1 + F^6) \left[ |\rho_A|^2 + |\rho_B|^2 \right]^3
+ \frac{4 \sqrt{2}}{J} F^3 \left[ |\rho_A|^2 |\rho_B| \cos (5 \phi_A - \phi_B) \right]
+ |\rho_B|^5 |\rho_A| \cos (5 \phi_B - \phi_A).
$$

(39)
The general structure of Eq. (39) has been derived in Ref. 8 from the symmetries of the problem. According to the previous discussion, to leading order, \( \varepsilon_0^{(6)} \sim (\Gamma_c - \Gamma)^3 \). For all values of the amplitudes \(|\rho_A|\) and \(|\rho_B|\), the expression in the right-hand side of Eq. (39) is minimal when both cosines are equal to \(-1\). This selects the phases:

\[
\phi_A = \frac{\pi}{6} + \frac{5\pi}{12}p, \quad \phi_B = -\frac{\pi}{6} + \frac{5\pi}{12}p, \tag{40}
\]

where \(p\) is an integer, yielding 24 independent sets \((\phi_A, \phi_B)\). The variation of \( \varepsilon_0^{(6)} \) with respect to \(|\rho_A|\) and \(|\rho_B|\) under the constraints (38) and (40) then selects either \(|\rho_B|/|\rho_A| = F\) or \(|\rho_B|/|\rho_A| = F^{-1}\). All 48 solutions thus generated correspond to the same dimer pattern (shifted or/and rotated) found in Ref. 9 and thus we recover the 48-fold degeneracy discussed in Ref. 8.

The approach described above is based on the assumption that all other modes would only contribute to the energy expansion to higher order. We shall now show that, since when considering only the critical modes one has to push the expansion to order 6, this assumption is not valid because some second- and fourth-order terms involving noncritical modes also make contributions of order \((\Gamma_c - \Gamma)^3\) which are essential for determining \(\phi_A\) and \(\phi_B\).

The dominant terms coupling the critical modes with \(\vec{q} = \pm \vec{q}_A\) and \(\vec{q} = \pm \vec{q}_B\) with extra modes are expected to be linear in the amplitudes of these extra modes and of the third order in the amplitudes of critical modes. The conservation of the total momentum then imposes on the wavevectors of these extra modes the condition:

\[
\vec{q} = m_A \vec{q}_A + m_B \vec{q}_B, \tag{41}
\]

where \(m_A\) and \(m_B\) are integers and \(m_A + m_B = \text{odd}\). In the first Brillouin zone there are only two wavevectors compatible with this condition: \(\vec{q}_C = 2\vec{q}_A - \vec{q}_B\) and \(-\vec{q}_C\). Let us denote the Fourier coefficients associated with the modes with \(\vec{q} = \vec{q}_C\) by \(\bar{\rho}_n = |\bar{\rho}_n|e^{i\phi_n}\), where \(n = 1, \ldots, 4\) refers to the number of the sublattice. The terms in the energy functional which are linear and harmonic in \(\bar{\rho}_n\) are

\[
\mathcal{E}_1^{(4)} = -\frac{J}{4} \sum_{n,n'} \bar{\rho}_n^* \left( \hat{M}(\vec{q}_C) - \frac{\Gamma}{J} \right)_{n,n'} \bar{\rho}_{n'} + \frac{\Gamma}{8} \sum_{n=1}^4 \left( R_n \bar{\rho}_n + \text{c.c.} \right), \tag{42}
\]

with

\[
R_n = \rho_A^3(u_A)_n^3 + 3(\rho_A^*)^2(u^*_A)_n^2 \rho_B(u_B)_n + 3\rho_A(u_A)_n(\rho_B^*)^2(u^*_B)_n^2 + \rho_B^3(u_B)_n^3. \tag{43}
\]

The variation of Eq. (42) with respect to \(\bar{\rho}_n^*\) gives

\[
\bar{\rho}_n = \frac{\Gamma}{2J} \sum_{n'} \left[ \hat{M}(\vec{q}_C) - \frac{\Gamma}{J} \right]_{n,n'}^{-1} R_n. \tag{44}
\]

Injecting Eq. (44) into Eq. (42) we obtain

\[
\mathcal{E}_1^{(4)} = -\Gamma h(\Gamma/J) \left[ |\rho_A|^2 + |\rho_B|^2 \right]^3 -\Gamma g(\Gamma/J) \left[ |\rho_A|^5 |\rho_B| \cos(5\phi_A - \phi_B) + |\rho_A|^3 |\rho_B|^3 \cos(5\phi_B - \phi_A) \right], \tag{45}
\]

where we have introduced the notation

\[
h(\gamma) = \frac{\gamma}{8(\gamma^2 - 3)} \left\{ \gamma(1 + F^6) + 6\sqrt{2}(3F^2 - 1) \right\}, \quad g(\gamma) = \frac{3\gamma F^6}{4(\gamma^2 - 3)} \left\{ 4\gamma F^2 + 3\sqrt{2}(2F^2 - 1) \right\}.
\]

Eq. (44) proves that \(\bar{\rho}_n\) scales as

\[
|\bar{\rho}_n| \sim |\rho_A|^3 \sim |\rho_B|^3 \sim (\Gamma_c - \Gamma)^{\frac{3}{2}}, \tag{46}
\]

leading to \(\mathcal{E}_1^{(4)} \sim (\Gamma_c - \Gamma)^3\). So it is clear that this contribution cannot be neglected since it is of the same order as \(\varepsilon_0^{(6)}\), and that other contributions involving non-critical modes such as e.g. sixth order terms will be of higher order. This means that the phases of the critical modes have to be determined by minimizing the sum of \(\varepsilon_0^{(6)}\) and \(\mathcal{E}_1^{(4)}\). The contribution to this expression depending on the phases reads:

\[
-\Gamma \left[ g \left( \frac{\Gamma}{J} \right) - \frac{3}{2} F^6 \right] \left[ |\rho_A|^5 |\rho_B| \cos(5\phi_A - \phi_B) + |\rho_A|^3 |\rho_B|^3 \cos(5\phi_B - \phi_A) \right].
\]

Now \(g(\Gamma/J) - (3/2)F^3\) is positive for \(\Gamma/J > \sqrt{3}\). Therefore, since we are interested in the domain just below \(\Gamma/J = \sqrt{6}\), the energy is minimal when both cosines are equal to \(+1\). This selects the phases:

\[
\phi_A = \frac{\pi}{12} p, \quad \phi_B = \frac{5\pi}{12} p, \tag{47}
\]

where \(p\) is an integer. This leads again to 24 independent sets \((\phi_A, \phi_B)\). In addition, minimizing \(\varepsilon_0^{(6)} + \mathcal{E}_1^{(4)}\) with respect to the amplitudes \(|\rho_A|\) and \(|\rho_B|\) under the constraint (38) selects, as before, either \(|\rho_B|/|\rho_A| = F\) or \(|\rho_B|/|\rho_A| = F^{-1}\). The 48 resulting solutions correspond to the 24 equivalent dimer patterns which can be obtained from the one shown in Fig. 7(a). The difference between Eq. (40) and Eq. (47) explains the qualitative difference between the structures of the plaquette phase found in this work and the solution of Ref. 9, which does not disappear even when the amplitudes of the \(\vec{q} = \pm \vec{q}_C\) modes become negligible as compared to those of the critical modes.

IV. INTERMEDIATE MIXED PHASES

During the numerical minimization of the classical energy for the 192-site system with periodic boundary conditions an additional intermediate phase was found to exist between the columnar and the plaquette phases. We
refer to this intermediate phase as the mixed phase because in the dimer representation the bonds with larger dimer densities, $d_{ij} \geq \frac{1}{2}$, are arranged in an alternating pattern of plaquettes and columns (see Fig. 8(a)). The mixed and plaquette phases have the same translational symmetries. However, the point group symmetries of the gauge-invariant dimer patterns in the two phases are different: $P31m$ for the plaquette phase (see Figs. 7(a)) and $Cmm$ for the mixed phase (see Fig. 8(a)). The phase transition between these two phases has to be of the first order, since the symmetry groups are not such that one is a subgroup of the other.

The mixed and plaquette phases have the same translational symmetries. However, the point group symmetries of the gauge-invariant dimer patterns in the two phases are different: $P31m$ for the plaquette phase (see Figs. 7(a)) and $Cmm$ for the mixed phase (see Fig. 8(a)). The phase transition between these two phases has to be of the first order, since the symmetry groups are not such that one is a subgroup of the other.

![Fig. 8](image1.png)

FIG. 8: (Color online) (a) The dimer density $d_{ij}$ in the mixed phase at $\Gamma/J = 1.72$. The thickness of the bonds is proportional to $d_{ij}$. The dimer densities are also emphasized by the colors of the bonds ranging from red (the lowest densities) to dark blue (the highest densities, $d_{ij} > \frac{1}{2}$). The bonds with $d_{ij} \geq \frac{1}{2}$ are organized in an alternating pattern of plaquettes and columns. (b) The spin configuration in the same state in the gauge that leads to a 24-site unit cell (large hexagon). Note the existence of 2 sites at which $S_z^i = 0$.

As in the case of the plaquette phase, the size of a unit cell of the mixed phase can be reduced from 48 sites for the standard gauge shown in Fig. 1 to 24 sites in the gauge of Fig. 7(c), see Fig. 8(b). In this gauge the spin pattern consists of spins with the same sign of $S_z^i$ having seven different orientations, one of which is in the direction of the field. In contrast to the spin pattern in the plaquette phase, which is centered on one of the sites of the honeycomb lattice (Fig. 7(c)), in the mixed phase this pattern is centered on one of the bonds of the lattice (Fig. 8(b)), which explains the difference in symmetry between the two states. The degeneracy of the mixed phase is equal to 36 in terms of the dimer representation and to 72 in terms of the spin representation. Each of the 36 equivalent dimer patterns corresponds to two spin configurations which can be transformed one into the other by changing the sign of $S_z^i$ for all spins. The stability of the mixed state with respect to small fluctuations has been investigated with LSWT in the gauge producing a 24-site unit cell, and this phase has been found to be stable in the range $1.394 \lesssim \Gamma/J \lesssim 1.774$.

The existence of the mixed state whose structure is shown in Fig. 8 suggests that there can also exist states in which the straight rows of plaquettes are still equidistant but separated not by single columns but by a larger number of columns which below is denoted by $n$ (see Fig. 9). From now on we number such mixed states by the index $n$ and call the simplest mixed state discussed above the first mixed state.

![Fig. 9](image2.png)

FIG. 9: (Color online) Dimer patterns in the mixed states with $n \leq 3$, where $n$ denotes the number of columns separating the plaquette patterns. The notation is the same as in Figs. 7(a) and 8(a). The bonds with $d_{ij} \geq \frac{1}{2}$ are organized in an alternating pattern of plaquettes and columns. Thin lines show the boundaries between unit cells.

It is not hard to understand that the unit cell of the second mixed state (in the optimal gauge in which the sign of $S_z^i$ is the same for all spins) has exactly the same symmetry as the unit cell of the first mixed state and can be obtained from it by adding on each side eight more sites. The successive repetition of this procedure allows one to construct the unit cell for any integer $n$ and to find that it contains $8(2n + 1)$ sites. However, due to the symmetry of the unit cell, the number of non-equivalent sites only increases by four when $n$ increases by one, which leads to $4n + 3$ non-equivalent sites.

For $n \leq 7$ we have performed a numerical minimization of the energy for the unit cells corresponding to such structures, and we have found that, upon decreasing $\Gamma/J$, the energy of the second mixed state first becomes lower than that of the first mixed state, after what the energy of the third mixed state becomes lower than that of the second mixed state, and so on. Tab. I summarizes the values of $\Gamma/J$ at which the transition between the $n$th and $(n + 1)$th mixed states takes place and reports the width of the region in which the $n$th mixed state has the
lowest energy. It can be seen that for \( n > 1 \) this width is scaled down by a factor of the order of 50 each time \( n \) increases by 1. This means that \( \Gamma_c^{n,n+1} \) approaches a finite limit exponentially fast. The extrapolation shows that the accumulation point of \( \Gamma_c^{n,n+1} \) at \( n \to \infty \) is \( \Gamma_c^\infty / J = 1.67612786261 \). Below this field columnar states have the lowest classical energy.

| \( n \) | \( \Gamma_c^{n,n+1} / J \) | \( \Delta \Gamma_c^n / J \) |
|-------|------------------|------------------|
| 1     | 1.69372479498    | 4.3 \times 10^{-2} |
| 2     | 1.67655449242    | 1.7 \times 10^{-2} |
| 3     | 1.67613666486    | 4.2 \times 10^{-4} |
| 4     | 1.67612802659    | 8.6 \times 10^{-6} |
| 5     | 1.67612786551    | 1.6 \times 10^{-7} |
| 6     | 1.67612786267    | 2.8 \times 10^{-9} |
| 7     | \ldots           | \ldots           |

TABLE 1: Critical field \( \Gamma_c^{n,n+1} \) of the transition between the \( n \)th and \((n+1)\)th mixed states. The last column shows \( \Delta \Gamma_c^n = \Gamma_c^{n-1,n} - \Gamma_c^{n,n+1} \), the field range in which the \( n \)th mixed state has lower classical energy than the \((n-1)\)th and the \((n+1)\)th states (\( \Gamma_c^0 \) refers to the transition between the plaquette and the first mixed state).

Note that it was impossible to discover any of the mixed states with \( n > 1 \) during the minimization of the energy for the 192-site cell (with periodic boundary conditions and the standard gauge of Fig. 1) which was instrumental in discovering the \( n = 1 \) mixed state. The reason is very simple - the periodicity of all the states with \( n > 1 \) is incompatible with the periodic boundary conditions implemented in this 192-site cell.

The existence of such a sequence of phase transitions suggests that the main contribution to the energy of the \( n \)th mixed phase (counted off from the energy of a columnar state) is proportional to the density of linear defects (vertical rows of plaquettes) whose energy can be considered as linearly dependent on \( \Gamma \), whereas the main correction to this energy comes from the repulsion of nearest defects, which decreases exponentially fast with the distance between them. This was checked at \( \Gamma = \Gamma_c^\infty \) where the proper energy of a linear defect changes sign, and indeed we have found that the energies of different states are compatible with an interaction of linear defects that is exponential in the distance between them. This makes us confident that the narrow region above \( \Gamma_c^\infty \) has to contain an infinite sequence of mixed phases with all integer indices \( n \).

It is well known that in a system consisting of a sequence of linear defects there can also exist phases with more complex structures, in which the linear defects are not equidistant. In terms of our problem such phases would correspond to a regular alternation of, for example, \( n \) and \( n + 1 \) columns, or of \( n \), \( n \) and \( n + 1 \) columns, etc., leading to what is known as a devil’s staircase. Usually such phases appear in a phase diagram if the interaction of more distant defects is also repulsive, whereas the interaction between next-to-nearest defects is attractive, one gets a direct transition from the \( n \)th to the \((n+1)\)th phase without the presence of an intermediate \((n,n+1)\) phase.

We have verified numerically that in our system the energy of the \((1,2)\) mixed state is never lower than either the energy of the first state or that of the second mixed state, which means that it cannot be present in the phase diagram. Quite surprisingly, the situation with the \((2,3)\) phase is different, and in a narrow interval around \( \Gamma_c^{2,3} \) [from \( \Gamma_c^{2,3} - 1.3 \times 10^{-9} \) to \( \Gamma_c^{2,3} + 1.9 \times 10^{-9} \)] its energy is lower than the energies of the second and third mixed states. One can estimate that even if some other complex phases do exist, the field range where any of them minimizes the energy will be at least a couple of orders of magnitude smaller than the already extremely narrow interval of the existence of the \((2,3)\) state, so we decided not to pursue the investigation of this point any further since it cannot be of much relevance.

A more important question is whether the plaquette and the first mixed states may be separated by a region where there appear mixed states of a different type, in which the density of columns is lower than in the first mixed state, so that the neighboring columns are separated by domains of plaquette state. Such a scenario seems to us to be impossible however for the following reasons.

The comparison of Fig. 7(c) with Fig. 8(b) suggests that the structure of the first mixed state is very close to what one would obtain by constructing the superposition of two plaquette states centered on neighboring sites of the lattice (and letting this superposition relax). Therefore one can interpret these two states as different manifestations of a unique state which can move around in a complex periodic potential with minima both at the positions corresponding to lattice sites and at the positions corresponding to the middles of lattice bonds. For \( \Gamma > \Gamma_c^{0,1} = 1.73600830184 J \), the minima located at lattice sites are the lowest, whereas for \( \Gamma < \Gamma_c^{0,1} \), the minima located at the middle of lattice bonds are the lowest. Exactly at \( \Gamma = \Gamma_c^{0,1} \) all these minima have equal depths. This picture can be confirmed by constructing a family of states which continuously interpolates between the plaquette and the first mixed state, which allows a numerical analysis of the effective potential discussed above. This analysis reveals that at \( \Gamma = \Gamma_c^{0,1} \) the barrier separating inequivalent (but equal) minima is very low (~ \( 1.07 \times 10^{-5} J \) per site). Nonetheless, any attempt to construct a state which somewhere looks like the plaquette state and elsewhere like the first mixed state would force the system to overcome this barrier in some places. This will increase its energy in comparison with that of the plaquette or of the first mixed state.

The numerical evidence in favor of this conclusion comes from observing that the state which would differ from the first mixed state by having half its density of
columns has a periodicity which is compatible with the 192-site cell used in our numerical energy minimization. Therefore, if at $\Gamma = \Gamma_{c}^{0.1}$ the energy of this state was lower than that of the plaquette and of the first mixed states, this state would be accessible during this minimization procedure. To be on the safe side, we have also performed a minimization of the energy for the cell whose periodicity in addition to the formation of the plaquette and of the first mixed states allows for the appearance of the states which differ from the first mixed state by keeping only one column out of three (or two out of three), but this has not allowed us either to find any state with energy lower than that of the plaquette or of the first mixed state. This gives an additional evidence in favor of our conclusion that the phase transition between the plaquette and the first mixed states should be a direct one without any intermediate phases with a more complex structure.

V. PHASE DIAGRAM

A. Classical phase diagram

The classical phase diagram consists of 4 regions: (i) The columnar phase, which is highly degenerate since all columnar states have the same energy. It extends up to $\Gamma/J \approx 1.676$; (ii) The region of mixed states with columnar patterns separated by straight rows of plaquettes in the interval $1.676 \lesssim \Gamma/J \lesssim 1.737$; (iii) The plaquette phase, with a 24-site unit cell, in the range $1.737 \lesssim \Gamma/J \leq \sqrt{6} \approx 2.45$; (iv) The fully polarized phase with all spins pointing in the direction of the field for $\Gamma/J > \sqrt{6}$. The transition from the fully polarized phase to the plaquette phase is a second-order one, all other transitions being of the first order. These results are summarized in Fig. 10.

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B. Quantum fluctuations

Quantum fluctuations can a priori modify this phase diagram in two main ways. First of all, if the degeneracy of the classical ground states is accidental (that is, not related to symmetry), they can select some of these states. This is indeed the case in the columnar phase, where the columnar states with domain walls of only the first type are selected already at the level of harmonic fluctuations.

Secondly, quantum fluctuations can shift the phase boundaries. When one takes into account only the harmonic fluctuations, this applies only to first-order transitions. Indeed, at a first-order transition, the classical energy is the same for the two competing configurations, but the spectra of harmonic fluctuations are different, and one phase will in general be stabilized over the other by zero point fluctuations. A convenient way to keep track of the stability of the various phases with respect to quantum fluctuations is to draw a phase diagram in the $(\Gamma/J,1/S)$ plane (see Fig. 11) showing which phase has the lowest total energy.

The resulting phase diagram can be quite involved when there are many phases in competition, and this is clearly the case here since, for $1/S = 0$, there exists an infinite sequence of mixed phases. However, it turns out that for $1/S$ above $10^{-3}$ only three of them survive, as is shown in Fig. 11. All other mixed phases exist only for $1/S \lesssim 10^{-4}$ in a very narrow range of transverse magnetic field of width $\lesssim 10^{-4} J$. They are thus invisible on the scale of Fig. 11, which has been adjusted to properly describe the competition between the two
main phases (plaquette and columnar). On that scale, the phase diagram consists of six phases: the polarized phase, the plaquette phase, the first, second and fourth mixed states, and the columnar phase. The general trend is that the plaquette phase is stabilized by quantum fluctuations over the mixed phases as well as the columnar phase.

Note that the transition between the plaquette and the columnar phases cannot be followed below $\Gamma/J = 1.64$ at this level of approximation because the plaquette phase is no longer locally stable with respect to harmonic fluctuations. The continuation of this boundary by a dashed line in Fig. 11 is just a guide to the eye. To follow this line further would require to go beyond the harmonic approximation. The transition between the plaquette and polarized phases being of the second order, the boundary has to start vertically since, at the transition, both states have the same quantum corrections in the harmonic approximation. This is indicated by a vertical dashed line in Fig. 11. To find the curvature of this line would require to go beyond the harmonic approximation.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{phase_diagram.png}
\caption{Semiclassical corrections to the phase diagram, $M_n \in \{1, 2, 4\}$ denote 1st, 2nd and 4th mixed states, zoommed for values of the field close to the accumulation point of mixed states.}
\end{figure}

In view of the very strong modification of these phase boundaries upon decreasing $S$, it is legitimate to wonder about the fate of the columnar and mixed phases for $S = 1/2$, for which the model can be mapped onto the QDM in the limit $\Gamma/J \rightarrow 0$. The results presented above suggest that the mixed phases have absolutely no chance to extend to $S = 1/2$.

Regarding the competition between the columnar and the plaquette phases, we can get an estimate of the critical value of the spin at which the boundary between them crosses the axis $\Gamma = 0$ by looking at the linear $1/S$ corrections starting from the point where the two phases have the same classical energy, a point that does not appear on the phase diagram of Fig. 11 since it lies inside the 1st mixed phase. This leads to the conclusion that the columnar phase disappears above $1/S \approx 0.67$, i.e. below $S \approx 1.49$. Note that this should probably be considered as a lower bound in terms of $S$ since the boundary is slightly concave. So, for $S = 1/2$, the semiclassical calculation at the harmonic level predicts only two phases: a plaquette phase up to $\Gamma/J = \sqrt{6}$, and a polarized phase above. The fact that we find the point $\Gamma/J = 0$ to be in the region of stability of the plaquette phase is in good agreement with the QDM, which has been found by QMC to be in the $\sqrt{12} \times \sqrt{12}$ phase at $V/t = 0.1, 2$.

VI. CONCLUSIONS

In conclusion, we have investigated the classical phase diagram of the FFTFIM on the honeycomb lattice and how it is modified by the semiclassical corrections induced by harmonic fluctuations. As compared to what has been already known about the model, namely that the paramagnetic phase is unstable at $\Gamma/J = \sqrt{6}$ towards a crystalline phase with a large unit cell, the classical phase diagram turns out to be surprisingly rich, with a multitude of additional phases: a columnar phase at small transverse field and an infinite cascade of phases of mixed columnar and plaquette character. The phase towards which the paramagnetic phase is unstable at $\Gamma/J = \sqrt{6}$ has been found to have the same symmetry and periodicity as the state proposed in Ref. 9, but a different structure. Both are characterized by a 24-site unit cell in the spin language, and by a 12-site cell on the dual lattice in the dimer language, but the state we have found has a plaquette structure. At the classical level, the columnar phase is fully degenerate, all columnar states having rigorously the same classical energy.

Quantum fluctuations have been found to modify this phase diagram in two important respects. First of all, harmonic fluctuations have been shown to partially lift the degeneracy of the columnar phase in favor of the columnar states with only one type of domain walls. Since the remaining degeneracy is not related to a symmetry of the model, anharmonic corrections are expected to lift further this degeneracy. Secondly, they modify strongly the phase boundaries, and for the ultra quantum limit, $S = 1/2$, they predict that the plaquette phase survives down to $\Gamma \rightarrow 0$.

Going back to the original motivation of this investigation, namely the properties of the QDM on the triangular lattice, these results deserve a number of comments. First of all, our semiclassical approximation predicts that the phase which is the analog of the $\sqrt{12} \times \sqrt{12}$ phase of the QDM has a 4-site plaquette structure. This reopens the issue of the nature of the $\sqrt{12} \times \sqrt{12}$ phase of the QDM. According to the results of GFQMC simulations, possible structures are constrained by a quasi-extinction of the dimer density correlation function at the corner of the Brillouin zone. This has been shown to be consistent with a uniform distribution of dimer density inside the interior part of the 12-site hexagonal unit cell, a conclusion somehow supported by the conclusions of Ref. 9 regarding the nature of the phase close to the paramagnetic phase. Now that we know that this phase is in fact a plaquette phase, it would be interesting to revisit the
GFQMC results to see to which extent a plaquette phase of this type might be consistent with the quasi-extinction at the zone corner.

It is also inspiring that a columnar phase appears in the classical solution of the FFTFIM since a similar phase is present in the QDM for attractive interactions between dimers. We did not manage to find a convincing connection between large $S$ in the FFTFIM and negative $V$ in the QDM, but since we found intermediate phases between the columnar phase and the plaquette phase in the FFTFIM, it is tempting to speculate that such phases may also exist in the QDM.

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Appendix A: Comparison of columnar and staggered states

Columnar states are the states which maximize $N_d$, the number of pairs of frustrated bonds situated at the smallest possible distance from each other [as shown in Fig. 2(d)]. In this Appendix we want to compare the classical energy of these states with the energy of the states in which $N_d$ is minimal, that is, is equal to zero. In terms of dimer models such states are usually called staggered or nonflippable states, because they do not contain flippable pairs of dimers.

Since in a staggered state all frustrated sites have identical environments (with exactly one frustrated neighbor) and all non-frustrated sites also have identical environments (with exactly two frustrated neighbors), such a state can be described by the same two variables $\theta_1$ and $\theta_2$ introduced in Sec. II B for the description of a columnar state. In terms of $\theta_1$ and $\theta_2$ the energy of a staggered state can be written as

$$E_{st} = -\frac{N}{2} \left[ J + \frac{4}{2} (\cos^2 \theta_1 + 4 \cos \theta_1 \cos \theta_2 - \cos^2 \theta_2) + \Gamma(\sin \theta_1 + \sin \theta_2) \right].$$

(A1)

Even without minimizing $E_{st}$ with respect to $\theta_1$ and $\theta_2$ one can notice that for any $\theta_1$ and $\theta_2$

$$E_{st}(\theta_1, \theta_2) - E_{col}(\theta_1, \theta_2) = (JN/4)(\cos \theta_1 - \cos \theta_2)^2 \geq 0$$

and therefore the energy of a staggered state [the minimum of $E_{st}(\theta_1, \theta_2)$] has to be higher than the energy of a columnar state [the minimum of $E_{col}(\theta_1, \theta_2)$ achieved when $\cos \theta_1 \neq \cos \theta_2$]. This proves that the maximization of $N_d$ is always a better strategy than its minimization, even when the ratio $\Gamma/J$ is not small.

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