Correlated partial disorder in a weakly frustrated quantum antiferromagnet

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Partial disorder—the microscopic coexistence of long-range magnetic order and disorder—is a rare phenomenon, that has been experimental and theoretically reported in some Ising- or easy plane-spin systems, driven by entropic effects at finite temperatures. Here, we present an analytical and numerical analysis of the $S = 1/2$ Heisenberg antiferromagnet on the $\sqrt{3} \times \sqrt{3}$-distorted triangular lattice, which shows that its quantum ground state has partial disorder in the weakly frustrated regime. This state has a $180^\circ$ Néel ordered honeycomb subsystem, coexisting with disordered spins at the hexagon center sites. These central spins are ferromagnetically aligned at short distances, as a consequence of a Casimir-like effect originated by the zero-point quantum fluctuations of the honeycomb lattice.

**Introduction**- Zero-point quantum fluctuations in condensed systems are responsible for a wide variety of interesting phenomena, ranging from the existence of liquid helium near zero temperature to magnetically disordered Mott insulator\(^{112}\). It is in the quantum magnetism arena, precisely, where a plethora of control factors are available for tuning the amount of quantum fluctuations. Among these factors, space dimensionality, lattice coordination number, spin value $S$, and frustrating exchange interactions are the most relevant\(^{33}\).

While folk wisdom visualizes zero-point quantum fluctuations like an uniform foam resulting from an almost random sum of states, in some cases these fluctuations contribute to the existence of very unique phenomena. These phenomena include semiclassical orders\(^3\), order by disorder\(^4\), effective dimensionality reduction\(^5\), and topological orders associated with quantum spin liquid states\(^6\), among others. Another role for quantum fluctuations is to allow the emergence of complex degrees of freedom from the original spins, like weakly coupled clusters or active spin sublattices decoupled from orphan spins. The latter has been proposed to explain the spin liquid behavior of the LiZn\(_2\)Mo\(_3\)O\(_6\)\(^8\). Here the system is described by a triangular spin-$\frac{1}{2}$ Heisenberg antiferromagnet which is deformed into an emergent honeycomb lattice weakly coupled to the central spins.

Besides spin liquids, the presence of weakly coupled magnetic subsystems can lead to partial disorder, that is, the microscopic coexistence of long-range magnetic order and disorder. This rare phenomenon has been experimental and theoretically reported in different Ising- or XY-spin highly frustrated systems\(^9\)\(^\text{10}\)\(^\text{11}\) and it is usually driven by thermal fluctuations. In general, it is thought that some amount of spin anisotropy is needed to get partial disorder, and that the disordered subsystem behaves as a perfect paramagnet, with its decoupled spins justifying then the calificative of orphan spins.

In this work, we present a frustrated magnetic system of Heisenberg spins with a partially disordered ground state, that originates in the zero-point quantum fluctuations. Specifically, we compute the ground state of the $S = 1/2$ antiferromagnetic Heisenberg model in the $\sqrt{3} \times \sqrt{3}$-distorted triangular lattice (Fig. 1), by means of the linear spin wave theory (LSWT) and the numerically exact density matrix renormalization group (DMRG). For the weakly frustrated $0 \leq J'/J \lesssim 0.18$ range, we find a novel partial disorder state, without semiclassical analog, that consists in the coexistence of a Néel order in the honeycomb sublattice and disordered central spins. In addition, the spins of the disordered sublattice are ferromagnetically aligned at short distances, a correlated behavior induced, as we will show, by a Casimir-like effect due to the “vacuum” quantum fluctuations inherent in the quantum Néel order of the honeycomb lattice.

**Model and methods**- We study the $S = \frac{1}{2}$ Heisenberg model on the $\sqrt{3} \times \sqrt{3}$-distorted triangular lattice. Under this distortion, the original triangular lattice is split into a honeycomb subsystem and a sublattice of spins at the

![FIG. 1: (color online)$\sqrt{3} \times \sqrt{3}$-distorted triangular lattice, with two different exchange interactions $J$ and $J'$. The arrows correspond to the spin directions of the semiclassical magnetic order.](image-url)
The classical ground state of (1) is a simple three-sublattice order \( (2^{+4} J_m^2 / 2) \) that the spin directions of spins at the centers of the hexagons. The classical ground state of (1) is a simple three-sublattice order \( (2^{+4} J_m^2 / 2) \) that the spin directions of spins at the centers of the hexagons. The classical ground state of (1) is a simple three-sublattice order \( (2^{+4} J_m^2 / 2) \) that the spin directions of spins at the centers of the hexagons.

This model has two very well known limits: (i) for \( J' = 1 \), we recover the Heisenberg model on the isotropic triangular lattice, with its three equivalent sublattices and a 120° Néel ordered ground state, while (ii) for \( J' = 0 \), we have a honeycomb Heisenberg model with its 180° Néel ordered ground state and an orphan (completely decoupled) spins at the centers of the hexagons.

The classical ground state of (1) is a simple three-sublattice order, as depicted in Fig. 1, characterized by the magnetic wave vector \( \mathbf{Q} = 0 \) and by the angles \( \phi_A = -\phi_B = -\arccos(-J'/2) \) that the spin directions on sublattices \( A \) and \( B \) make with the spin direction in sublattice \( C \). This ground state evolves continuously from the honeycomb (plus orphan \( C \) spins) to the isotropic triangular classical ground states, and it is a ferrimagnet for \( 0 < J' < 1 \). The Lacorre parameter \( \frac{2}{3} \) whose departure from the unity quantifies the degree of magnetic frustration, is \( (2 + J'^2)/(2 + 4J') \), so the maximal frustrated case corresponds to the isotropic triangular lattice.

In this work, we solve (1) by means of complementary analytical and numerical techniques—the semiclassical linear spin wave theory and the density matrix renormalization group—, in order to highlight the quantum behavior without classical counterpart of the model. The DMRG calculations were performed on ladders of dimension \( L_x \times L_y \), with \( L_y = 6 \) and \( L_x \) up to 15, imposing cylindrical boundary conditions (periodic along the \( y \) direction). We use up to 1200 DMRG states in the most unfavorable case to ensure a truncation error below \( 10^{-6} \) in our results.

Linear spin wave results. LSWT yields the same ground state magnetic structure as the classical one (see Fig. 1), with a semiclassically renormalized local magnetization \( m_\alpha / (\alpha = A, B, C) \) for each sublattice displayed in the inset of Fig. 3. As the \( A \) and \( B \) sublattices are equivalent, their order parameters coincide, while they are different from the central spin local magnetization \( m_C \). For \( J' = 0 \), the central spins are decoupled from the honeycomb lattice and, consequently, \( m_C \) can take any value from 0 to 1/2. As soon as \( J' \) is turned on, the sublattice \( C \) takes a large magnetization value, more than 80% of its classical value, while \( m_A \) varies continuously.

Another interesting feature that can be seen is that the increase of the frustrating interaction \( J' \) leads to an enhancement of the local magnetization in the honeycomb lattice, up to a broad maximum around \( J' = 0.35 \) (see darker curve in the inset of Fig. 3). This (apparent) paradoxical result can be explained by the increase of the effective coordination number induced by \( J' \), that drives the system closer to its classical behavior. Alternatively, it can be thought that, as \( J' \) is turned on, the honeycomb spins feel the \( C \) subsystem as an uniform Weiss magnetic field \( B = m_C J' \) that, through the suppression of quantum fluctuations, contributes to the increase of the local magnetization \( m_A = m_B \), as it was found in other frustrated systems. It is worth to notice that, for any \( J' \), the larger order parameter belong...
to the sublattice $C$, which can be considered to be the sublattice with the smaller effective coordination number, $z_{eff}^C \approx 6J'/J \leq z_{eff}^A \approx 3 + 3J'/J$. This is in agreement with the fact that in lattices with inequivalent sites or bonds, the order parameter is lower in the sites with larger coordination numbers.\textsuperscript{22}

DMRG results. For all the considered range $0 < J' \leq 1$, the computed spin correlations $(S_i \cdot S_j)$ exhibit a three sublattice pattern, in full agreement with the semiclassical approach. Thus, if a given sublattice is ordered, all its spins will point out in the same direction (ferromagnetic order) and its local magnetization can be evaluated using the expression\textsuperscript{29}

$$m_{\alpha}^2 = \frac{1}{N_\alpha(N_\alpha - 1)} \sum_{i,j \in \alpha \atop i \neq j} \langle S_i \cdot S_j \rangle,$$  \hspace{1cm} (2)

where $\alpha$ denotes the sublattice (A, B, or C), and $N_\alpha$ is its number of sites. The calculated $m_A$ and $m_C$ are shown in the main panel of Fig. [3].

The most eye-catching difference between the DMRG and the semiclassical local magnetizations appears in the weakly frustrated parameter region, close to the honeycomb phase, $0 \leq J' \leq 0.18$. There, DMRG shows a vanishing order parameter $m_C$ for the C sublattice along with an almost constant honeycomb lattice local magnetization $m_A$. This corresponds to a partially disordered phase, driven solely by quantum fluctuations (in competition with frustration), as we are working at $T = 0$. Notice that, in general, partially disordered phases are associated with entropic effects, and they appear at intermediate temperatures, between the lower and higher energy scales of the system.\textsuperscript{14,15}

There is a critical value $J'_c \approx 0.18$ where the C sublattice gets suddenly ordered, as it happens at $J' = 0^+$ in LSWT (inset of Fig. [3]). Furthermore, beyond this critical value, the DMRG calculations show a higher local magnetization in the central spin sublattice, in agreement with the semiclassical expectation.\textsuperscript{30}

The local magnetization $m_A (= m_B)$ of the honeycomb spins decreases when $J'$ increases due to the frustration introduced by the coupling with the central spins, until it reaches its minimum value in the isotropic triangular lattice, corresponding to the most frustrated case.\textsuperscript{30} In contrast with the spin wave results, $m_A$ does not exhibit a clear maximum for intermediate values of $J'$, but shows an almost constant region ranging from $J' = 0$ to 0.2. This feature signals a negligible effect of the central spins on the honeycomb ones.

As we have mentioned above, the semiclassical ground state is ferrimagnetic for $0 < J' < 1$. So, in order to further characterize the DMRG ground state magnetic structure, we calculate the lowest eigenenergy in the different $S_z$ subspaces. In the case of $J' = 0$, the spins $C$ are totally disconnected from the honeycomb subsystem and, thus, do not contribute to the total energy. This results in a perfect paramagnetic behavior of the central spin sublattice, with a high degeneracy of the ground state, as $E(S_z = 0) = E(S_z = \pm 1) = \ldots = E(S_z = S_z^{\max} = \pm 1/2 \times N_3/3)$, where $S_z^{\max}$ denotes the maximum value of $S_z$ whose subspace belongs to the ground state manifold, and $N_3$ is the number of sites of the cluster. For $J' \neq 0$ the situation changes: for $J' \leq J'_c$ there are no more orphan spins and $S_z^{\max} = 0$, indicating that the partially disordered phase is a (correlated) singlet. On the other hand, at the critical value $J'_c$, $S_z^{\max}$ jumps to a finite value, signaling a first order transition from the singlet to the ferrimagnetic state. With further increase of $J'$, $S_z^{\max}$ decreases until it vanishes for the isotropic triangular point $J' = 1$, whose ground state again is a singlet. Therefore, the DMRG magnetic order has a ferrimagnetic character for $J'_c < J' < 1$.

Next, we calculate the DMRG angles between the local magnetization in different sublattices. Previously, we have checked that the classical angles $\phi_{\alpha}$ are not renormalized in LSWT, in contrast to what happens in other frustrated models, like the triangular anisotropic Heisenberg model, where the pitch of its spiral magnetic order is sizable renormalized by quantum fluctuations.\textsuperscript{31} In order to estimate the angles, we take into account that the large values of the DMRG local magnetizations enable us to use a semiclassical picture of the spins. Let us think of a three-spin unit cell built by spins $A$, $B$, and $C$ of lengths $m_A$, $m_B$, and $m_C$, respectively, as seen in Fig. [1]. We can assume that the $S_z^{\max}$ subspace corresponds to the $C$ spin pointing out in the $z$-direction, while the $A$ and $B$ spins make angles $\phi_A = -\phi_B$ with it. Then, we get the equation

$$\frac{N_s}{3} (m_C + 2m_A \cos \phi_B) = S_z^{\max},$$

for $\phi_B$,\textsuperscript{31} which finally leads to the angle $\theta$ between $A$ and $B$ spins

$$\theta = 2 \arccos \left[ \frac{1}{2m_A} \left( m_C - \frac{3S_z^{\max}}{N_s} \right) \right].$$  \hspace{1cm} (3)

In Fig. [4] the angle $\theta$ is plot as a function of $J'$. It can be seen that the honeycomb 180° Néel order persists all along the partially disordered phase where $m_C$ vanishes. This result is a simple consequence of the singlet character of the ground state for $0 < J' < J'_c$ ($S_z^{\max} = 0$ in Eq. [3]). As a consequence, the canting behavior observed semiclassically for any finite $J'$ moves to the region above $J'_c$ in the strong quantum limit $S = 1/2$ calculated with DMRG. That is to say that, when $J'$ is small, $C$ spins are disordered because the system gains zero-point quantum energy from that disorder. For larger values of $J'$, the system chooses to gain (frustrated) exchange energy over zero-point quantum fluctuation, and the $C$ sublattice gets ordered, canting simultaneously the $A$ and $B$ spins.

It is worth to emphasize that, even when the ground state seems to undergo a first order transition at $J'_c$ ($S_z^{\max}$ changes abruptly and the local magnetization $m_C$ sharply rises), the angle $\theta$ between the $A$ and $B$ spins...
varies continuously from its 180° value in the partially disordered phase. This is similar to the spin wave behavior around \( J' = 0 \), where the sublattice \( C \) is disordered, but as soon as \( J' \) rises, \( m_C \) suddenly grows over \( m_A \) without any abrupt change in the magnetic order.

The quantitative agreement between the DMRG and LSWT angles for \( J' \gtrsim J'_c \), displayed in Fig. 4, is a clear evidence that, beyond the partially disordered phase present in the weakly frustrated regime, the quantum ground state of the model is very well described semi-classically.

Up to now, we have characterized the region between \( J' = 0 \) and 0.18 as a singlet partially disordered phase. To deepen the understanding of such disorder, in Fig. 3 the average nearest neighbor spin correlation \( r_{ij} \), \( i \neq j \), between the central spins is shown as a function of \( J' \). It can be seen that, even though the sum over all the correlations in the \( C \) sublattice is zero in the region of its null local magnetization (see Eq. 2), the (average) nearest neighbor correlation has an almost constant positive value, close to 1/8. This means that each central spin is ferromagnetically correlated with its first neighbor \( C \) spins. In other magnetic systems which exhibit partial order, mostly with Ising or XY-like spins, the disordered sub-system is a perfect paramagnet of orphan spins, with zero correlation between them. As there is no explicit exchange interaction between the \( C \) spins, their correlation should be mediated by the coexistent honeycomb Néel order.

In order to build up a qualitative argument about the origin of the correlated character of the partially disordered phase below \( J'_c \), we appeal to a Weiss molecular field approach for the simplest toy model (see Supplemental Material for details). We consider a 4-spin cluster, composed of two nearest neighbor honeycomb spins (1 and 2) interacting with the two closest \( C \) spins (3 and 4), with a Hamiltonian

\[ H = J S_1 \cdot S_2 + J' (S_1 + S_2) \cdot (S_3 + S_4). \]

After fixing the state of honeycomb spins 1 and 2 as a “classical Néel order”, plus zero-point quantum fluctuations quantified by a parameter \( r \), we arrive at an effective Hamiltonian for the central spins 3 and 4, that consists in a Zeeman term associated with an effective uniform magnetic field \( B \propto r J' \), perpendicular to the honeycomb Néel order. Hence, this toy model helps us to understand how the ferromagnetic correlations between nearest neighbor central spins are built up under an effective interaction between them, driven by the vacuum fluctuations of the honeycomb Néel order; that is, the correlation between central spins can be considered a Casimir-like effect. This treatment is valid whenever this Néel order is unaffected by the feedback of the spins 3 and 4. This seems to be the case in the DMRG calculations for the lattice, as the local magnetization \( m_A \) changes only slightly by the coupling of the \( C \) spins in the partially disordered phase (see Fig. 5). Also, the toy model explains the almost constant correlation between central spins that can be seen, below \( J'_c \), in Fig. 5 (except for the non-monotonic behavior very close to \( J' = 0 \), probably due to numerical inaccuracies). It should be mentioned that, due to the singlet character of the DMRG ground state, the correlations between the central spins are isotropic, and not perpendicular to a given direction like in the toy model.

**Summary** - We have studied a \( S = 1/2 \) Heisenberg antiferromagnet with inequivalent exchange interactions on a distorted triangular lattice which, in the weakly frustrated regime \( 0 \leq J'/J \lesssim 0.18 \), exhibits a novel correlated partial disordered phase, driven by the competi-

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**FIG. 4**: (color online) DMRG and LSWT angles \( \theta \) between spins in the \( A \) and \( B \) sublattices as a function of \( J' \).

**FIG. 5**: Average DMRG nearest neighbor correlations between central spins as a function of \( J' \).
tion between zero-point quantum fluctuations and frustration. Due to the quantum character of the ordered subsystem, there is a non-trivial interplay between the different components of the system. While this kind of partial disordered phases has been analyzed theoretically and experimentally in hexagonal systems, at intermediate temperatures and for Ising-like spins, here we present, for the first time, the theoretical realization of such phase as the ground state of a simple Heisenberg antiferromagnet.

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Supplemental material for:
Correlated partial disorder in a weakly frustrated quantum antiferromagnet

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We sketch the linear spin wave calculation for the Heisenberg model on lattices with complex unit cells, as it corresponds to the $\sqrt{3} \times \sqrt{3}$-distorted triangular lattice. Also, we present the Weiss mean field-like treatment of a 4-spin cluster model in order to understand the role of quantum fluctuations of the honeycomb Néel order in the establishment of ferromagnetic correlations between central spins in the partially disordered phase.

I. LINEAR SPIN WAVE CALCULATION

In this section, we present the linear spin wave theory used to solve semiclassically the Heisenberg model. The Hamiltonian (1) can be rewritten in a generic way as

$$ H = \frac{1}{2} \sum_{ij\alpha\beta} J_{ij\alpha\beta} (\mathbf{R}_i - \mathbf{R}_j) \mathbf{S}_{i\alpha} \cdot \mathbf{S}_{j\beta}, $$

(S1)

where $i, j$ denote unit cells with vector positions $\mathbf{R}_i, \mathbf{R}_j$, respectively, while $\alpha, \beta$ represent the spins inside the unit cell. $J_{ij\alpha\beta}(\mathbf{R}_i - \mathbf{R}_j)$ is the exchange interaction between the spin $\alpha$ in the unit cell $\mathbf{R}_i$ and the spin $\beta$ in the unit cell $\mathbf{R}_j$. In our case, the $\sqrt{3} \times \sqrt{3}$-distorted triangular lattice is a non-Bravais lattice that can be described as a triangular lattice with basis vectors $\mathbf{a}_1 = (\frac{3a}{2}, \frac{\sqrt{3}a}{2})$ and $\mathbf{a}_2 = (0, \sqrt{3}a)$ ($a$ is the nearest neighbor distance), and a unit cell with the three sites A, B, and C (see Fig. 1). $J_{ij\alpha\beta}$'s only take non-zero values for nearest neighbors:

$$ J_{AB}(0) = J_{AB}(\mathbf{a}_1) = J_{AB}(\mathbf{a}_1 - \mathbf{a}_2) = J, $$

$$ J_{AC}(0) = J_{BC}(0) = J_{AC}(\mathbf{a}_2) = J_{BC}(\mathbf{a}_2) = J_{AC}(\mathbf{a}_1) = J_{BC}(\mathbf{a}_2 - \mathbf{a}_1) = J', $$

and the remaining ones can be obtained through the relation $J_{ij\alpha\beta}(\mathbf{R}_i - \mathbf{R}_j) = J_{ij\alpha\beta}(\mathbf{R}_j - \mathbf{R}_i)$.

In order to perform the spin wave analysis, we first need the classical ground state of the Heisenberg model. For this purpose, we consider classical spin vectors in (S1) and we minimize the corresponding energy (S2)

$$ E_{\text{clas}} = \sum_{\alpha, q} J_{ij\alpha\beta}(q) \mathbf{S}_{q\alpha} \cdot \mathbf{S}_{-q\beta}, $$

(S2)

where $J_{ij\alpha\beta}(q) = \sum_{\mathbf{R}} J_{ij\alpha\beta}(\mathbf{R}) e^{i \mathbf{q} \cdot \mathbf{R}}$ is the Fourier transform of the exchange interaction matrix $J$, and $\mathbf{S}_{q\alpha} = \frac{1}{\sqrt{N}} \sum_{i} \mathbf{S}_{i\alpha} e^{i \mathbf{q} \cdot \mathbf{R}_i}$ are classical vectors subject to the normalization condition $\mathbf{S}_{q\alpha}^2 = 1$. The sum over $\mathbf{R}$ runs over all the $N$ unit cells in the cluster. For the $\sqrt{3} \times \sqrt{3}$-distorted triangular lattice, the minimization of the classical energy (S2) gives the magnetic wave vector $\mathbf{Q} = 0$, implying that the same absolute magnetic pattern is repeated in each unit cell. If $\phi_\alpha$ is the angle between the spins in the $\alpha$ sublattice and the $z$ direction, after a little algebra we find $\phi_A = -\phi_B = -\arccos(-J'/2J)$, and $\phi_C = 0$. This three sublattice (coplanar) structure is shown in Fig. 1.

Once we know the classical ground state magnetic structure, we make a rotation to local axes in (S1). That is, at each site we define a local frame in such a way that the classical order points along the local $y$ direction, $\mathbf{H}_i$, for simplicity, we consider a planar classical magnetic structure (like in our case), lying in the $xz$ plane, then the spin operators in the local axes are

$$ \mathbf{S}_{\alpha} = \mathcal{R}_y(\Theta_{\alpha \alpha}) \mathbf{S}_{i\alpha}, $$

where $\Theta_{\alpha \alpha} = \mathbf{Q} \cdot \mathbf{R}_i + \phi_\alpha$ is the angle between the classical $\mathbf{S}_{i\alpha}$ spin and the global $z$ direction, and $\mathcal{R}_y(\Theta_{\alpha \alpha})$ is the matrix rotation of angle $\Theta_{\alpha \alpha}$ around the $y$ direction. In terms of the local spin operators $\mathbf{S}$, the Hamiltonian (S1) turns into

$$ H = \frac{1}{2} \sum_{ij\alpha\beta} \left[ \cos(\Theta_{ij\alpha\beta}) \left( \hat{S}_{i\alpha \alpha}^x \hat{S}_{j\beta}^x + \hat{S}_{i\alpha \alpha}^z \hat{S}_{j\beta}^z \right) + \sin(\Theta_{ij\alpha\beta}) \left( \hat{S}_{i\alpha \alpha}^y \hat{S}_{j\beta}^y - \hat{S}_{i\alpha \alpha}^z \hat{S}_{j\beta}^z \right) \right]. $$
In the local axis frame, the classical magnetic structure is a ferromagnetic one along the \( z \) direction. Now, we can represent the local spins by bosonic operators \( a_{i\alpha}^\dagger, a_{i\alpha} \), using the typical Holstein-Primakoff transformation:\(^{[S3]}\)

\[
\begin{align*}
\tilde{S}^z_{i\alpha} &= S - a_{i\alpha}^\dagger a_{i\alpha}, \\
\tilde{S}^+_{i\alpha} &= \sqrt{2S - a_{i\alpha}^\dagger a_{i\alpha}} a_{i\alpha}, \\
\tilde{S}^-_{i\alpha} &= a_{i\alpha}^\dagger \sqrt{2S - a_{i\alpha}^\dagger a_{i\alpha}}.
\end{align*}
\] (S3)

In the semiclassical approximation, valid for \( S \rightarrow \infty \), the (operator) square roots that appear in the Holstein-Primakoff representation \([S3]\) are replaced by the scalar \( \sqrt{2S} \), yielding the linear spin wave approximation. After performing this approximation and a Fourier transformation to momentum space, the Hamiltonian \([S1]\) is written as

\[
H_{LSW} = \left(1 + \frac{1}{S}\right) E_{\text{clas}} + \frac{S}{2} \sum_{\alpha\beta\mathbf{q}} a_{\mathbf{q}\alpha}^\dagger D_{\alpha\beta}(\mathbf{q}) \cdot a_{\mathbf{q}\beta},
\] (S4)

where the spinor \( a_{\mathbf{q}\beta} = (a_{\mathbf{q}\beta}, a_{\mathbf{q}\beta}^\dagger)^T \), the bosonic dynamical matrix is

\[
D_{\mathbf{q}} = \begin{pmatrix}
A_{\alpha\beta}(\mathbf{q}) & -B_{\alpha\beta}(\mathbf{q}) \\
-B_{\alpha\beta}(\mathbf{q}) & A_{\alpha\beta}(\mathbf{q})
\end{pmatrix},
\]

and

\[
A_{\alpha\beta}(\mathbf{q}) = \sum_{\mathbf{R}} J_{\alpha\beta}(\mathbf{R}) \cos(\mathbf{q} \cdot \mathbf{R}) \cos^2 \left( \frac{\mathbf{Q} \cdot \mathbf{R} + \phi_{\alpha} - \phi_{\beta}}{2} \right) - \delta_{\alpha\beta} \sum_{\gamma\mathbf{R}} J_{\alpha\gamma}(\mathbf{R}) \cos (\mathbf{Q} \cdot \mathbf{R} + \phi_{\alpha} - \phi_{\gamma}),
\]

\[
B_{\alpha\beta}(\mathbf{q}) = \sum_{\mathbf{R}} J_{\alpha\beta}(\mathbf{R}) \cos(\mathbf{q} \cdot \mathbf{R}) \sin^2 \left( \frac{\mathbf{Q} \cdot \mathbf{R} + \phi_{\alpha} - \phi_{\beta}}{2} \right).
\]

The quadratic Hamiltonian \([S4]\) can be diagonalized by means of a paraunitary Bogoliubov transformation. After this procedure, the order parameter at each site can be evaluated using \( m_{i\alpha} = | < \tilde{S}^z_{i\alpha} > | \).

### II. WEISS MEAN FIELD-LIKE APPROXIMATION FOR A 4-SPIN CLUSTER

In order to understand the origin of the correlated character of the partial disordered phase below \( J'_c \approx 0.18 \), we resort to a molecular Weiss-like mean field approach for a simple toy model. We consider a 4-spin cluster as it is shown in Fig. S1, composed of two central spins C (3 and 4) and two honeycomb spins, one A (1) and another B (2). Its Hamiltonian is

\[
H = JS_1 \cdot S_2 + J' (S_1 + S_2) \cdot (S_3 + S_4),
\] (S5)

and we are interested in the case \( J' \ll J \).

\[\text{FIG. S1: 4-spin cluster model. The solid line corresponds to an interaction } J, \text{ while the dashed ones correspond to } J'.\]

The exact ground state of Hamiltonian \([S5]\) can be easily found, and it consists, for \( J'/J \leq 1/2 \)\(^{[S4]}\) in a singlet state between the 1 and 2 spins, while the remaining spins 3 and 4 can have any spin projection \( \sigma, \sigma' \):

\[
|\text{GS} > = \left( \frac{|1 \uparrow 2 \downarrow > - |1 \downarrow 2 \uparrow >}{\sqrt{2}} \right) \otimes |3\sigma 4\sigma' >.
\]
we propose to fix the spins 1 and 2 in the normalized state

\[ |\psi(1, 2)\rangle = \frac{1}{\sqrt{1 + 2r^2}} \left[ |1 \uparrow 2 \downarrow\rangle + r \left( |1 \uparrow 2 \uparrow\rangle + |1 \downarrow 2 \downarrow\rangle\right) \right], \]  

(S6)

that represent a “classical Néel order” (spin 1 \(\uparrow\) and spin 2 \(\downarrow\)) plus quantum fluctuations quantified by the real parameter \(r\). \(r\) can be related with the local magnetization of the “honeycomb spins” as

\[ m_0 = \langle \psi(1, 2)|S_1^z|\psi(1, 2)\rangle = \frac{1}{2(1 + 2r^2)}, \]

so we get the relation \(r \approx \sqrt{2} - m_0 \ll 1\) for \(m_0\) close to its classical value.

In the spirit of the Weiss molecular field theory, we approximate (S5) by an effective Hamiltonian for spins 3 and 4,

\[ H_{34} = J <\psi(1, 2)|S_1 \cdot S_2|\psi(1, 2)\rangle > + J' \langle \psi(1, 2)| (S_1 + S_2) |\psi(1, 2)\rangle \cdot (S_3 + S_4), \]  

(S7)

which results in the expression

\[ H_{34} = 4m_0 r J' \left( S_3^z + S_4^z \right) + \text{const}. \]  

(S8)

That is, the quantum fluctuations of the classical Néel order of the spins 1 and 2 act as an effective uniform magnetic field in the \(x\) direction for the central spins 3 and 4. For any \(r > 0\) the ground state of (S8) is \(3 \downarrow 4 \downarrow >_x\), where \(x\) refers to the quantization axis.

Of course, the \(x\) and \(y\) directions are equivalent, the same as the spin projection. To show this, we consider a more general “Néel state” than (S6), giving the possibility that the zero-point quantum fluctuation states \(|1 \uparrow 2 \uparrow\rangle\) and \(|1 \downarrow 2 \downarrow\rangle\) are out of phase, that is

\[ |\psi(1, 2)\rangle = \frac{1}{\sqrt{1 + 2r^2}} \left[ |1 \uparrow 2 \downarrow\rangle + e^{i\theta_1} |1 \uparrow 2 \uparrow\rangle + e^{i\theta_2} |1 \downarrow 2 \downarrow\rangle \right]. \]

(S9)

Inserting this state in (S7), we get the effective Hamiltonian

\[ H_{34} = 2m_0 r J' \left[ (\cos \theta_1 + \cos \theta_4) (S_3^z + S_4^z) + (\sin \theta_1 - \sin \theta_4) (S_3^y + S_4^y) \right], \]

that is, a Zeeman term for spins 3 and 4 with an effective magnetic field \(\vec{B} = 2m_0 r J' (\cos \theta_1 + \cos \theta_4, \sin \theta_1 - \sin \theta_4, 0)\), perpendicular to the classical Néel order in the \(z\) direction. Again, in the ground state \(|\psi(3, 4)\rangle >\) of this Hamiltonian the spins 3 and 4 are aligned along the \(\vec{B}\) direction and, straightforwardly, it results the classical ferromagnetic correlation

\[ <\psi(3, 4)|S_3 \cdot S_4|\psi(3, 4)\rangle > = \frac{1}{4} \]

between them, independently of the value of \(J'\). It is worth to notice that in the two extreme cases, when the state of spins 1 and 2 is taken as a classical Néel one or as a singlet, there is no correlation between spins 3 and 4. So, a quantum Néel order is necessary for the spins 3 and 4 to be correlated.