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 experimentally 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° 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.

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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 insulators [1,2]. 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, the lattice coordination number, the spin value S, and frustrating exchange interactions are the most relevant [3,4].

While folk wisdom visualizes zero-point quantum fluctuations like a 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 [5], effective dimensionality reduction [6,7], and topological orders associated with quantum spin liquid states [1], 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 $LiZn_2Mo_3O_8$ [8]. Here, the system is described by a triangular spin-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 localized or itinerant Ising- or XY-spin highly frustrated systems [9–25], and it is driven by *entropic effects* at finite temperature. In general, it is believed 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 then justifying the denomination of orphan spins.

In this work, we present an isotropic frustrated magnetic system for which the ground state exhibits partial disorder, originated by *zero-point quantum effects*, in contrast to the entropic origin of the so-far known cases. 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 of 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.

At the heart of the decoupling mechanism is the competition between the exchange energy favored by larger

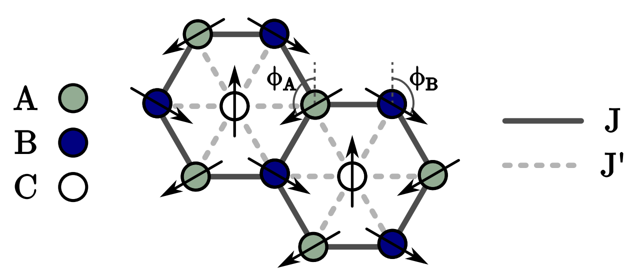

FIG. 1. $\sqrt{3} \times \sqrt{3}$-distorted triangular lattice, with two different exchange interactions J and J'. The arrows correspond to the spin directions of the semiclassical magnetic order.

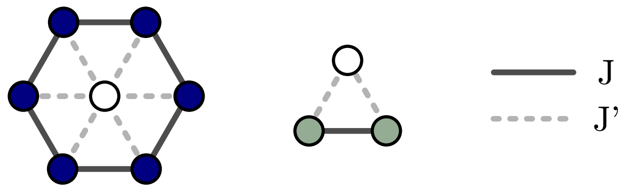

FIG. 2. Hexagon and triangle toy models with one spin connected to the remaining ones by a different exchange interaction J' (dashed lines).

coordination numbers and the zero-point quantum fluctuations. This can be roughly illustrated by simple toy models. For example, we can resort to the triangle and the hexagon with a central spin of Fig. 2, which present ground state energy level crossings at $J'/J = 1$ and $J'/J = 0.63$, respectively. For small J', the strongly connected spins form singlets, leaving the central spin completely decoupled.

Model and methods.—Under this distortion, the original triangular lattice is split into a honeycomb subsystem and a sublattice of spins at the center of each hexagon (see Fig. 1). Naturally, two different nearest-neighbor exchange interactions arise, and the Heisenberg Hamiltonian turns out

$$H = J\sum_{\langle ij\rangle}\mathbf{S}_i \cdot \mathbf{S}_j + J'\sum_{[ik]}\mathbf{S}_i \cdot \mathbf{S}_k, \qquad (1)$$

where $\langle ij\rangle$ runs over nearest-neighbor spins belonging to the honeycomb lattice (with equivalent sublattices A and B), whereas $[ik]$ links the honeycomb and central spins C that interact with energy J'. Throughout this work, we take $J = 1$ as the energy unit, whereas J' is the only varying parameter, which we consider in the range [0, 1].

Almost two decades ago, this model was proposed in the context of the honeycomb reconstruction of the metallic surface of Pb/Ge(111) [26]. On the other hand, the complementary range of $J' \geq 1$ has been considered [27,28] and, very recently, the uniform magnetization in the regime $J' \leq 1$ has been computed by exact diagonalization [29].

Related stacked triangular lattice XY antiferromagnets have been extensively studied in the context of the magnetic properties of hexagonal ABX_3 compounds with space group $P6_3cm$ [12,19,30].

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 [31,32]; whereas (ii) for $J' = 0$, we have a honeycomb Heisenberg model with its 180° Néel ordered ground state [33], and the orphan (completely decoupled) spins at the centers of the hexagons.

The classical ground state of Eq. (1) is a simple three-sublattice order [34], as depicted in Fig. 1, characterized by the magnetic wave vector $\mathbf{Q} = \mathbf{0}$ 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 [38], whose departure from the unity quantifies the degree of magnetic frustration, is $(2 + J'^2)/(2 + 4J')$, and so the maximal frustrated case corresponds to the isotropic triangular lattice.

In this work, we solve Eq. (1) by means of complementary analytical and numerical techniques–the semiclassical linear spin wave theory [34] and the density matrix renormalization group [39]–in order to highlight the quantum behavior without the classical counterpart of the model. The DMRG calculations were performed on ladders of dimension $L_x \times L_y$ [40], with $L_y = 6$ and L_x up to 15, imposing cylindrical boundary conditions (periodic along the y direction). We use up to 3000 DMRG states [34] in the most unfavorable case to ensure a truncation error below 10^{-6} in our results.

Linear spin wave results.—The LSWT yields the same ground state magnetic structure as the classical one (see Fig. 1), with a semiclassically renormalized local magnetization m_α ($\alpha = A, B, C$) for each sublattice [34], which is displayed in the inset of Fig. 3. As the A and B sublattices are equivalent, their order parameters coincide, whereas 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, which is more than 80% of its classical value, whereas 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 the darker curve in the inset of Fig. 3). This (apparent)

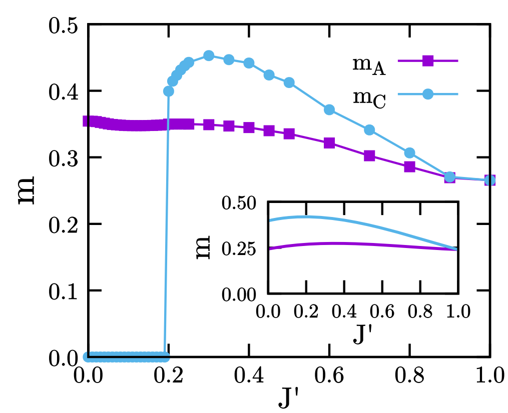

FIG. 3. Local magnetization of sublattices A and C as a function of J', calculated with the DMRG (main panel, where lines are merely a guide to the eye) and with the LSWT (inset). The DMRG results correspond to the $L_y = 6$, $L_x = 12$ cluster.

paradoxical result can be explained by the increase of the effective coordination number induced by J', which drives the system closer to its classical behavior. Alternatively, it can be thought that, as J' is turned on, the honeycomb feel the C subsystem as a 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 [41]. It is worth noticing that, for any J', the larger-order parameter belongs to the sublattice C, which can be considered to be the sublattice with the smaller effective coordination number: $z^C_{\rm eff} \approx 6J'/J \leq z^A_{\rm eff} \approx 3 + 3J'/J$. 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 [42].

DMRG results.—For all the considered range $0 \leq J' \leq 1$, the computed spin correlations $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$ exhibit a three-sublattice pattern, which is 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 [43]

$$m^2_\alpha = \frac{1}{N_\alpha(N_\alpha - 1)} \sum_{\substack{i,j \in \alpha \\ i \neq j}} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle, \qquad (2)$$

where α denotes the sublattice (A, B, or C), and N_α is its number of sites. The calculated m_A and m_C are shown in the main panel of Fig. 3 for the $N_s = 12 \times 6$ cluster (for other cluster sizes, we have obtained a similar result [34]).

The most eye-catching difference between the DMRG and the semiclassical local magnetizations appears in the weakly frustrated parameter region, which is close to the honeycomb phase: $0 \leq J' \lesssim 0.18$. 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, which is driven solely by quantum fluctuations (in competition with frustration), because 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 [11,20,22].

There is a critical value $J'_c \simeq 0.18$ where the C sublattice gets suddenly ordered because it happens at $J' = 0^+$ in the LSWT (inset of Fig. 3). Furthermore, beyond this critical value, the DMRG calculations show a higher local magnetization in the central spin sublattice, which is in agreement with the semiclassical expectation [42].

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 [44]. 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 $E(S_z = 0) = E(S_z = \pm 1) = \cdots = E(S_z = S_z^{\rm max} = \pm 1/2 \times N_s/3)$, where $S_z^{\rm max}$ denotes the maximum value of S_z whose subspace belongs to the ground state manifold, and N_s 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 $S_z^{\rm max} = 0$ [see Fig. S2(a) [34]], indicating that the partially disordered phase is a (correlated) singlet. On the other hand, at the critical value J'_c, $S_z^{\rm max}$ jumps to a finite value, signaling a first-order transition from the singlet to the ferrimagnetic state (see Fig. S2(b) [34]). With further increase of J', $S_z^{\rm max}$ 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 $J'_c < J' < 1$. This behavior shows little finite size effects [34] and it quantitatively agrees with the exact diagonalization predictions [29].

Next, we calculate the DMRG angles between the local magnetization in different sublattices. For this purpose, 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 $S_z^{\rm max}$ subspace corresponds to the C spin pointing out in the z direction, whereas the A and B spins make angles $\phi_A = -\phi_B$ with it. Then, we get the equation $(N_s/3)(m_C + 2m_A \cos\phi_B) = S_z^{\rm max}$, for ϕ_B [26], which finally leads to the angle θ between A and B spins

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

In Fig. 4, the angle θ is plotted 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 $0 < J' < J'_c$ [$S_z^{\rm max} = 0$ 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 the 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

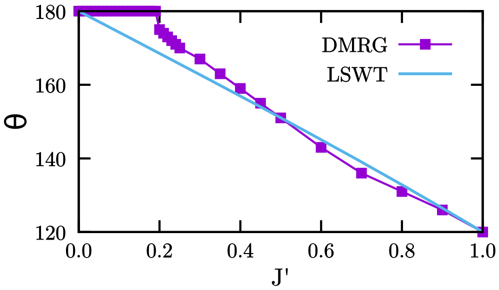

FIG. 4. DMRG and LSWT angles θ between spins in the A and B sublattices as a function of J'.

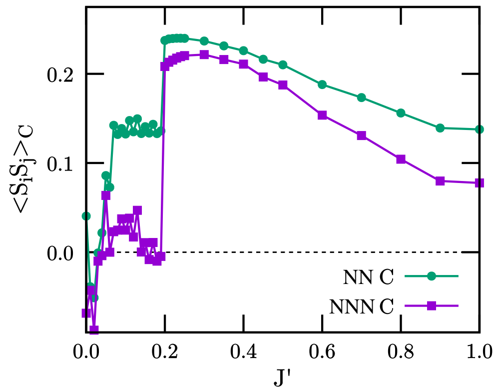

FIG. 5. Average DMRG nearest-neighbor and next-nearest-neighbor correlations between central spins as a function of J'.

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 emphasizing that, even when the ground state seems to undergo a first-order transition at J'_c ($S_z^{\max/rm}$ changes abruptly and the local magnetization m_C sharply rises), the angle θ 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 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 semiclassically.

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. 5, the average nearest and next-nearest neighbor (NN and NNN, respectively) spin correlations [45] 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 NN correlation has an almost constant positive value, close to $1/8$, whereas the NNN correlation is close to zero. This means that central spins exhibit (very) short-range ferromagnetic correlations between them, suggesting that the partial disordered phase may be thought of as a sort of a *resonating* spin-triplet valence bond state [46]. In other magnetic systems that exhibit partial disorder, mostly with Ising or XY-like spins, the disordered subsystem is a perfect paramagnet of orphan spins, with zero correlation between them [11,12]. 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 [34] for details). We consider a four-spin cluster composed of two nearest-neighbor honeycomb spins (1 and 2) interacting with the two closest C spins (3 and 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 central spins 3 and 4 that consists of a Zeeman term associated with an effective uniform magnetic field $B \propto rJ'$ 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 argument is further supported by the presence of nearest-neighbor antiferromagnetic spin–spin correlations between the orphan spins and the honeycomb ones (see Ref. [34]). This treatment is valid whenever this Néel order is unaffected by the feedback of 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. 3). Also, the toy model explains the almost constant nearest-neighbor correlation between central spins that can be seen, below J'_c, in Fig. 5 (except for the nonmonotonic behavior very close to $J' = 0$, which is 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.

The absence of a long-range ferromagnetic order of the C subsystem below J'_c [47] can be roughly explained as follows: in the previous Weiss mean-field argument, the zero-point quantum fluctuations of the honeycomb subsystem act as a magnetic field for the C spins. The direction of this effective molecular magnetic field is "random" because it depends on the phase fluctuations of the departure from the Néel state. Therefore, while the nearest-neighbor C spins are ferromagnetically correlated, the overall subsystem remains disordered due to the randomness of the zero-point quantum fluctuations.

Summary.—We have studied a $S = 1/2$ Heisenberg antiferromagnet with inequivalent exchange interactions on a distorted triangular lattice that, in the weakly frustrated regime $0 \leq J'/J \lesssim 0.18$, exhibits a novel correlated partial disordered phase driven by the competition between zero-point quantum fluctuations and frustration. Even if partial disordered phases were already known [9–25]—in anisotropic systems at finite temperatures and with disordered subsystems that behave as perfect paramagnets—here, we uncover, for the first time, a partially disordered phase as the ground state of a simple isotropic Heisenberg antiferromagnet. This phase exhibits the coexistence of two magnetic subsystems: one antiferromagnetically ordered, and the other disordered with its spins ferromagnetically correlated at short distances due to the zero-point quantum fluctuations of the ordered subsystem *via* a Casimir-like effect.

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[44] As we are working on finite size clusters, the local magnetizations are somewhat higher than the expected ones in the thermodynamic limit: $m \simeq 0.20$ for the triangular lattice [31,32], and $m \simeq 0.26$ for the honeycomb case [33].

[45] We compute this correlation averaging over all the first neighbors of the cluster, except those closest to the open edges in order to avoid border effects.

[46] R. Shindou and T. Momoi, SU(2) Slave-Boson formulation of spin nematic states in $S=1/2$ frustrated ferromagnets, Phys. Rev. B **80**, 064410 (2009).

[47] Another evidence of the absence of long-range ferromagnetic order in the central spin subsystem, below J_c', is given by its zero magnetization even under the application of a small uniform magnetic field h to this subsystem. See Ref. [34].