Effective field theory approach for the $S = \frac{3}{2}$ bilayer honeycomb antiferromagnet

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The spin-$\frac{3}{2}$ Heisenberg antiferromagnet on the bilayer honeycomb lattice is a minimal model to describe the magnetic behavior of $Bi_3Mn_4O_{12}(NO_3)$. We study this model with frustrating interlayer second-neighbor couplings taking into account quantum and thermal fluctuations. We use a path integral formulation in terms of coherent states to describe the low-energy physics of the model. We show that for a particular point in the parameter space, close to the experimental estimated couplings, a continuum classical degeneracy is lifted by both quantum and thermal fluctuations, and a collinear state is then selected by an order by disorder mechanism. Our results provide a global perspective in the understanding of the experimental observations.

I. INTRODUCTION

Frustrated magnetism is a prominent area of research with a broad range of subfields, harboring new quantum states of condensed matter [1,2], topological ordering [3–5], and addressing candidates for quantum computing [6–8]. Frustrated Heisenberg models on the honeycomb lattice have become a paradigmatic example for the search of competing spiral order, lattice nematicity, and plaquette valence bond states [9–26]. Furthermore, there are materials like the bismuth oxynitrate $Bi_3Mn_4O_{12}(NO_3)$ [27], where the Mn^{4+} ions of spin $\frac{3}{2}$ form honeycomb layers, with both nearest and next-nearest-neighbor antiferromagnetic exchange, and the Mn^{4+} ions are grouped into pairs, so the resulting structure is a bilayer honeycomb lattice. This compound has led research in bilayer honeycomb systems [28–34]. Most of these studies have focused on the stability of the semiclassical phases, extending previous work on the single-layer case. The quantum phase diagram was partially studied for the $S = \frac{1}{2}$ case, emphasizing the regions of the phase diagram where the ground state consists in a product of singlets, or quantum dimers, genuinely related to the bilayer geometry and not present in the single-layer model [32]. However, a complete understanding of the quantum phase diagram of the bilayer model is still missing.

Inelastic neutron-scattering measurements [35] have been performed in $Bi_3Mn_4O_{12}(NO_3)$ at high magnetic fields, and more recently [36] the magnetic couplings were estimated analyzing the magnetic dispersions using the linear spin-wave theory. The results for the couplings are consistent with previous results determined by *ab initio* density functional theory calculations [37], suggesting that there is no significant frustration in the honeycomb plane but frustrating interlayer interactions probably play an important role in destabilizing magnetic order. The neutron-scattering experiments show the presence of a short-range antiferromagnetic order at low temperatures and the presence of a magnetic transition in which the short-range order expands into a long-range Néel order. The experimental data indicate that the collinear Néel state becomes more stable at higher temperatures, i.e., thermal fluctuations stabilize the long-range Néel order by an order by disorder mechanism [35].

Motivated by these experimental and *ab initio* results, in this paper we study the honeycomb bilayer with in-plane first-neighbor interactions and competing interlayer first- and second-neighbor interactions. In Sec. II, we start by introducing the model and recalling some results in the bilayer geometry where the ground state can be worked out exactly and exhibits a singlet product state. In Sec. III we use a path integral formulation [38,39] in terms of coherent states to describe the spin-$\frac{3}{2}$ model in the presence of a magnetic field. We study both the weakly frustrated and strongly frustrated regimes. For the latter, we make the distinction between the dominant in-plane coupling case, and the dominant interlayer coupling case. On one hand, Sec. III B 1 is relevant for $Bi_3Mn_4O_{12}(NO_3)$, where we discuss how an order by disorder mechanism selects a staggered state as the one observed experimentally when the system is magnetized in the presence of an external magnetic field. Additionally, we argue how the system may exhibit magnetic short-range order for zero magnetization. On the other hand, Sec. III B 2 describes the formation of the valence bond solid studied in Ref. [32] within the field theory approach.

II. MODEL AND EXACT GROUND STATE

The model of interest consists of quantum spins $\frac{3}{2}$ on a bilayer honeycomb lattice, depicted in Fig. 1. The spins are coupled through Heisenberg isotropic interactions J_1, J_x, and J_0. For $Bi_3Mn_4O_{12}(NO_3)$, experimental and numeric results [36] show that J_1 is the dominant coupling, and J_0 is roughly three times the value of the J_x.

To connect with previous results in this system [32], let us write the Hamiltonian as a sum of terms over the square

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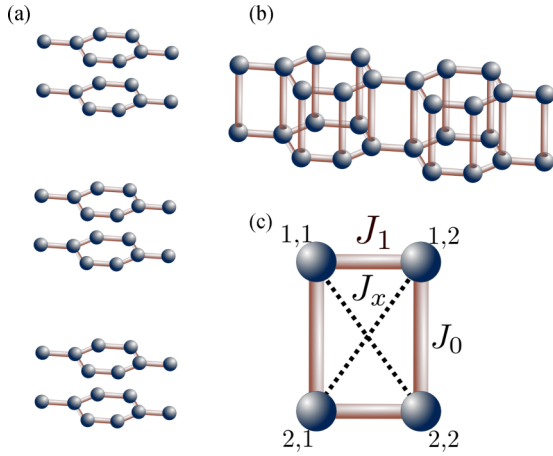[†]lamas@fisica.unlp.edu.ar

(a)　(b)

(c)

FIG. 1. The bilayer geometry considered here. Panel (a) shows the experimental structure corresponding to $Bi_3Mn_4O_{12}(NO_3)$. (b) Bilayer honeycomb lattice corresponding to model (1). (c) Four $S = \frac{3}{2}$ spins unit cell. The interactions in the Hamiltonian correspond to the in-plane nearest-neighbor coupling J_1, the interlayer nearest-neighbor coupling J_0, and the interlayer next-nearest-neighbor coupling J_x.

plaquettes schematized in Fig. 1(c).

$$
\begin{aligned}
H = \sum_{\boldsymbol{r}} \sum_{i=0}^{2} \Big\{ &\frac{J_0}{3} [\boldsymbol{S}_{1,1}(\boldsymbol{r}) \cdot \boldsymbol{S}_{2,1}(\boldsymbol{r}) + \boldsymbol{S}_{1,2}(\boldsymbol{r}) \cdot \boldsymbol{S}_{2,2}(\boldsymbol{r})] \\
&+ J_1 [\boldsymbol{S}_{1,1}(\boldsymbol{r}_i) \cdot \boldsymbol{S}_{1,2}(\boldsymbol{r}) + \boldsymbol{S}_{2,1}(\boldsymbol{r}_i) \cdot \boldsymbol{S}_{2,2}(\boldsymbol{r})] \\
&+ J_x [\boldsymbol{S}_{1,1}(\boldsymbol{r}_i) \cdot \boldsymbol{S}_{2,2}(\boldsymbol{r}) + \boldsymbol{S}_{2,1}(\boldsymbol{r}_i) \cdot \boldsymbol{S}_{1,2}(\boldsymbol{r})] \Big\} \\
&- h \sum_{\boldsymbol{r}} \left\{ S_{1,1}^z(\boldsymbol{r}) + S_{1,2}^z(\boldsymbol{r}) + S_{2,1}^z(\boldsymbol{r}) + S_{2,2}^z(\boldsymbol{r}) \right\}
\end{aligned} \quad (1)
$$

in which the z axis is oriented in the direction [40] of the external magnetic field h, $i = 0, 1, 2$ corresponds to $\boldsymbol{r}_{(0,1,2)} = \boldsymbol{r} + (\boldsymbol{0}, \boldsymbol{e}_1, \boldsymbol{e}_2)$, being $\boldsymbol{e}_1$ and $\boldsymbol{e}_2$ the primitive vectors of the triangular lattice.

When studying the low-energy theory of this model we will see that for some particular values of the couplings we find points where the theory presents zero modes. One of these points corresponds to the case $J_1 = J_x$ and J_0 large, where the ground state can be determined exactly and consists in a product of singlets. In order to see how the fine-tuning works, it is useful to introduce the bond spin operators

$$
\boldsymbol{L}_\eta = \boldsymbol{S}_{1,\eta} + \boldsymbol{S}_{2,\eta}, \quad \boldsymbol{K}_\eta = \boldsymbol{S}_{1,\eta} - \boldsymbol{S}_{2,\eta}, \quad (2)
$$

with $\eta = 1, 2$, where $[L_\eta^\alpha, L_{\eta'}^\beta] = i\epsilon^{\alpha\beta\gamma} L_\eta^\gamma \delta_{\eta,\eta'}$, $[L_\eta^\alpha, K_{\eta'}^\beta] = i\epsilon^{\alpha\beta\gamma} K_\eta^\gamma \delta_{\eta,\eta'}$, and $[K_\eta^\alpha, K_{\eta'}^\beta] = i\epsilon^{\alpha\beta\gamma} L_\eta^\gamma \delta_{\eta,\eta'}$ in the same unit cell $\boldsymbol{r}$, being $\epsilon^{\alpha\beta\gamma}$ the fully antisymmetric Levi-Civita tensor and $\epsilon^{xyz} = 1$.

The Hamiltonian written in terms of the bond operators is

$$
\begin{aligned}
H = -2N J_0 S(S+1) + \frac{1}{2} \sum_{\boldsymbol{r},i} \Big\{ &\frac{J_0}{3} \big(\boldsymbol{L}_1^2(\boldsymbol{r}_i) + \boldsymbol{L}_2^2(\boldsymbol{r}) \big) \\
&+ (J_1 + J_x) \boldsymbol{L}_1(\boldsymbol{r}_i) \cdot \boldsymbol{L}_2(\boldsymbol{r}) + (J_1 - J_x) \boldsymbol{K}_1(\boldsymbol{r}_i) \cdot \boldsymbol{K}_2(\boldsymbol{r}) \Big\} \\
&- h \sum_{\boldsymbol{r}} \left\{ L_1^z(\boldsymbol{r}) + L_2^z(\boldsymbol{r}) \right\},
\end{aligned} \quad (3)
$$

with N the number of unit cells in the system and S the spin quantum number.

From Eq. (3) we can see clearly that for $J_1 = J_x$, the last term in the Hamiltonian vanishes and the Hamiltonian depends only on the bond spin $\boldsymbol{L}_\eta(\boldsymbol{r})$. Therefore, at $J_1 = J_x$, the eigenstates of H are multiplets of the total bond spin. Among those is the *product state of bond singlets*, i.e., $|\psi\rangle = \bigotimes_{\boldsymbol{r}} |s_1\rangle_{\boldsymbol{r}} |s_2\rangle_{\boldsymbol{r}}$ with $\boldsymbol{L}_\eta(\boldsymbol{r})|s_\eta\rangle_{\boldsymbol{r}} = 0$, and $|s_\eta\rangle_{\boldsymbol{r}} = \sum_{m=-S}^{S} (-1)^{S-m} |m, -m\rangle / \sqrt{2S+1}$. Here $|m, -m\rangle$ labels a product of eigenstates of $S_{1,\eta}^z(\boldsymbol{r})$ and $S_{2,\eta}^z(\boldsymbol{r})$ on dimer η of the unit cell located at $\boldsymbol{r}$.

The preceding is valid for *any* site spin S, and for $J_0 \gg J_1 = J_x$ the valence bond solid described before is the ground state of the system both in the absence of a magnetic field or with an small magnetic field compared to the resulting magnetic gap. If we increase the value of $J_1 = J_x$ there will be a phase transition at some J_1^* where the nature of the ground state, as well as the value of J_1^* should depend on the spin S. Below we show that at the semiclassical level, the fine-tuning $J_1 = J_x$ gives rise to a zero mode in the effective theory.

III. SEMICLASSICAL EFFECTIVE FIELD THEORY

We write an effective field theory for the system using the coherent-state path integral description developed by Haldane [38] and Tanaka *et al.* [39]. The spins are represented by $O(3)$ vectors of modulus S: $\boldsymbol{S} = S(\cos\phi \sin\theta, \sin\phi \sin\theta, \cos\theta)$, and we consider quantum fluctuations on top of a lowest-energy configuration of the classical system. Our parametrization for the classical lowest-energy configuration in the presence of a magnetic field h consists in a canted Néel configuration, where for each unit cell we set

$$
\phi_{l,\eta}^0(\boldsymbol{r}) = \pi(l + \eta), \quad \theta_{l,\eta}^0(\boldsymbol{r}) = \theta^0(h, J_0, J_1), \quad (4)
$$

for $l = 1, 2$ and $\eta = 1, 2$. This configuration corresponds to the low-frustration limit $J_x \ll J_1, J_0$. The classical energy at $T = 0$ is then minimized by $\cos\theta^0 = h/2(J_0 + 3J_1)$. In the continuum limit quantum fluctuations are then added over the classical ground state changing $\phi_{l,\eta}^0(x, y) \to \pi(l + \eta) + \phi_{l,\eta}(x, y)$ and $\theta_{l,\eta}^0(x, y) \to \theta^0 + \delta\theta_{l,\eta}(x, y)$. The canonical conjugate fields of the theory are $\phi_{l,\eta}$ and $a\Pi_{l,\eta} = -S(\delta\theta \sin\theta^0 + \frac{1}{2}(\delta\theta)^2 \cos\theta_0)$, where a is the distance between the spins connected by J_1 in the unit cell. The spin operators written to quadratic order in $\phi_{l,\eta}$ and $\Pi_{l,\eta}$ are

$$
\begin{aligned}
S_{l,\eta}^z &= S\cos\theta_0 + a\Pi_{l,\eta}, \\
S_{l,\eta}^\pm &= (-1)^{l+\eta} e^{\pm i\phi_{l,\eta}} \Big(S\sin\theta^0 - \frac{m}{S\sin\theta^0} a\Pi_{l,\eta} \\
&\quad - \frac{1}{2} \frac{S^2}{S^2 - m^2} \frac{1}{S\sin\theta^0} (a\Pi_{l,\eta})^2 \Big),
\end{aligned} \quad (5)
$$

which fulfill the SU(2) algebra replacing the quantum commutator by a classical Poisson bracket. In the last equation we defined $m = S\cos\theta^0$. Replacing (5) in the Hamiltonian and keeping terms up to quadratic order, we obtain the noninteracting effective action $\mathcal{S}$ given by

$$
\mathcal{S} = \mathcal{S}_{cl} + \mathcal{S}_{BP}, \quad (6)
$$

where $\mathcal{S}_{cl}$ is the classical action of the system, which contains kinetic terms and mass terms, i.e., $\mathcal{S}_{cl} = \mathcal{S}_K + \mathcal{S}_M$. In order to simplify the notation using a single index for the fields, we rename the fields as

$$\phi_{i,j} \rightarrow \phi_{i(i-1)+j},$$

obtaining for the action the following expressions:

$$\mathcal{S}_K = \int d\tau \frac{d^2x}{v} \frac{K}{2a^2} \sum_{j=1}^{2}((\boldsymbol{e}_j \cdot \boldsymbol{\nabla}\phi_1)^2 + (\boldsymbol{e}_j \cdot \boldsymbol{\nabla}\phi_3)^2)$$

$$= \int d\tau \frac{d^2x}{v} \frac{K}{2} \frac{3}{2} (3(\partial_x\phi_1)^2 + 3(\partial_x\phi_3)^2$$

$$+ (\partial_y\phi_1)^2 + (\partial_y\phi_3)^2), \tag{7}$$

with $\tau \in [0, \beta]$ the imaginary time, β the inverse temperature (where Boltzmann constant is absorbed), $v = (9\sqrt{3}a^2)/2$, $K = (Sa)^2(J_1 - J_x)\sin^2(\theta^0)$, $\boldsymbol{e}_j = a\frac{\sqrt{3}}{2}[\sqrt{3}, (-1)^{j+1}]$, $j = 1, 2$, and

$$\mathcal{S}_M = \int d\tau \frac{d^2x}{v} \left(\frac{1}{2}\phi_i(M_\phi)_{ij}\phi_j + \frac{a^2}{2}\Pi_i(M_\Pi)_{ij}\Pi_j \right), \tag{8}$$

where Einstein's notation is being used for the repeated indices, and M_ϕ and M_Π are symmetric mass matrices. The second term in (6) is the Berry phase term, which arises from the nonorthogonality of the coherent-state basis, and is given by

$$\mathcal{S}_{BP} = -i(S - m) \int d\tau \frac{d^2x}{v} \sum_j \partial_\tau\phi_j$$

$$+ i \int d\tau \frac{d^2x}{v} \sum_j (\partial_\tau\phi_j)a\Pi_j. \tag{9}$$

The symmetric mass matrix M_ϕ is diagonalized by the transformation

$$W = \frac{1}{4} \begin{pmatrix} 1 & 1 & 1 & 1 \\ 1 & -1 & -1 & 1 \\ -1 & -1 & 1 & 1 \\ -1 & 1 & -1 & 1 \end{pmatrix}, \tag{10}$$

where $\phi'_j = \sum_{k=1}^{4} W_{jk}\phi_k$, $j = 1, \ldots, 4$ and $M'_\phi = 4WM_\phi W^{-1} = \mathrm{diag}(m_1, m_2, m_3, m_4)$, with

$$m_1 = 0, \quad m_2 = 8S^2 \sin^2(\theta^0)(J_0 + 3J_1),$$

$$m_3 = 8S^2 \sin^2(\theta^0)(J_0 - 3J_x),$$

$$m_4 = 24S^2 \sin^2(\theta^0)(J_1 - J_x). \tag{11}$$

The nullity of the first mass is known to prevail to all order in the development and reflects the protection of the ϕ'_1 field, which is the Goldstone field associated to the $U(1)$ symmetry of the Hamiltonian. The same transformation diagonalizes M_Π as well, i.e., $M'_\Pi = 4WM_\Pi W^{-1} = \mathrm{diag}(\mu_1, \mu_2, \mu_3, \mu_4)$ with

$$\mu_1 = 8(J_0 + 3J_1), \quad \mu_2 = 8(J_0 + 3J_1)\cot^2(\theta^0),$$

$$\mu_3 = 8[-J_0 + 3J_1 + (J_0 - 3J_x)\csc^2(\theta^0)],$$

$$\mu_4 = 8[J_0 - 3J_1 + 3(J_1 - J_x)\csc^2(\theta^0)]. \tag{12}$$

The complete effective action after the transformation W takes the form

$$\mathcal{S} = \int d\tau \frac{d^2x}{v} \left\{ \frac{K}{2} 3(3(\partial_x\phi'_2 - \partial_x\phi'_3)^2 + 3(\partial_x\phi'_1 - \partial_x\phi'_4)^2 \right.$$

$$+ (\partial_y\phi'_2 - \partial_y\phi'_3)^2 + (\partial_y\phi'_1 - \partial_y\phi'_4)^2)$$

$$+ \frac{1}{2}\sum_{j=2}^{4} \left(m_j\phi'^2_j\right) + \frac{1}{2}\sum_{j=1}^{4}(\mu_j(a\Pi'_j)^2)$$

$$\left. + (-4i)(S - m)(\partial_\tau\phi'_1) + 4i\sum_{j=1}^{4}(\partial_\tau\phi'_j)a\Pi'_j \right\}. \tag{13}$$

In general all mass terms of the Π'_j fields are nonzero, and as so these fields are short ranged. Since they are not expected to contribute at large scales, we can integrate them out, obtaining the effective action

$$\mathcal{S} = \int d\tau \frac{d^2x}{v} \left\{ \frac{K}{2} 3(3(\partial_x\phi'_2 - \partial_x\phi'_3)^2 + 3(\partial_x\phi'_1 - \partial_x\phi'_4)^2 \right.$$

$$+ (\partial_y\phi'_2 - \partial_y\phi'_3)^2 + (\partial_y\phi'_1 - \partial_y\phi'_4)^2)$$

$$+ \frac{1}{2}\sum_{j=2}^{4} \left(m_j\phi'^2_j\right) + \frac{1}{2}\sum_{j=1}^{4} \left(\frac{16}{\mu_j}(\partial_\tau\phi'_j)^2 \right)$$

$$\left. + (-4i)(S - m)(\partial_\tau\phi'_1) \right\}. \tag{14}$$

In the following, we will discuss some important regimes of this theory.

A. Weakly frustrated regime

For $J_x \ll J_0, J_1$ the system is weakly frustrated and we have a nonvanishing m_i, μ_j, $i = 2, \ldots, 4$, $j = 1, \ldots, 4$. As mentioned before, the field ϕ'_1 has no mass term because of the $U(1)$ symmetry, and in fact remains long ranged for $4(S - m) \notin \mathbb{Z}$ and $T = 0$.

The factor $1/4$ in the transformation (10) is chosen to give the field ϕ'_1 the correct periodicity [41]. If the fields ϕ'_2, ϕ'_3, and ϕ'_4 are gapped, they can be set to zero in the low-energy limit, meaning $\phi_1 = \phi_2 = \phi_3 = \phi_4$. In this case the action depends only on the gapless field $\phi'_1 = \phi_1$. Thus, the action (14) corresponds to a classical XY model with an additional Berry phase term. If $T = 0$, the theory is in three dimensions, whereas for finite temperature $0 \leqslant \tau \leqslant \beta < \infty$, and the theory is two-dimensional, where the Mermin-Wagner theorem forbids long-range order.

The condition $4(S - m) \notin \mathbb{Z}$ corresponds to the presence of a nontrivial Berry phase. For a spin in position (x_0, y_0) that has a phase winding of 2π when evolving in imaginary time there is a contribution to the effective action of $\delta\mathcal{S} = (-4i)(S - m)2\pi$. The Berry phase forbids vortices to contribute to the partition function because they enter in the partition function weighted with an oscillatory phase factor which leads to destructive interference [39]. For $T = 0$, these vortices are the only mechanism available to destroy the long-range antiferromagnetic order (LRAFO) for the in-plane spin
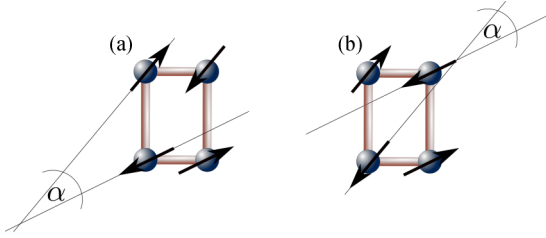
FIG. 2. Schematic representation of the zero modes corresponding to (a) $J_x = J_0/3$ and (b) $J_1 = J_x$.

components represented by the ϕ'_1 field. As vortices are forbidden for a generic value of the magnetization, the LRAFO is preserved at $T = 0$. For nonzero T, the LRAFO becomes a quasi-long-range antiferromagnetic order.

If $4(S - m)$ is an integer the Berry phase term trivializes and can be dropped. This condition is known as the Oshikawa-Yamanaka-Affleck [42] criterium, and is a well-known necessary condition for a magnetic plateau to emerge. Vortices of ϕ'_1 are able to proliferate if that is energetically favorable (i.e., if the stiffness is small enough), disordering the field ϕ'_1 and opening a gap in the system. The resulting phase has short-range antiferromagnetic order. Finally, the special case where $4(S - m)$ is a rational number can give rise to more exotic phases, as developed in Ref. [39].

B. Frustrated regime

The situation described above changes when the values of the couplings promote competition between terms of the Hamiltonian. Below we discuss those situations in two limits, where the dominant interaction is the in-plane or the interlayer coupling.

1. Dominant in-plane coupling

If J_1 is the dominant coupling [as expected for $Bi_3Mn_4O_{12}(NO_3)$] [36], increasing J_x we find that $m_3 = 0$ at $J_x = J_0/3$ [see Eq. (11)], whereas $\mu_j > 0$, $j = 1, \ldots, 4$. It is easy to see that the condition $J_x = J_0/3$ leads not only to a vanishing mass in the effective theory but it corresponds to a zero mode of the Hamiltonian using the parametrization (4). Indeed, if we take the Hamiltonian (1) evaluated in $J_x = J_0/3$, we can make a variation in the classical parametrization (4) introducing the real parameters $\alpha_{l,\eta}$ by

$$\phi^0_{l,\eta} = (l + \eta)\pi + \alpha_{l,\eta}, \qquad (15)$$

where $l = 1, 2$, $\eta = 1, 2$. If we take $\alpha_{l,1} = \alpha_{l,2}$, $l = 1, 2$, then the Hamiltonian is a function of θ^0 only. As a consequence of the U(1) symmetry of the system we can set $\alpha_{2,1} = 0$, leaving only one free parameter $\alpha_{1,1} \equiv \alpha$. The parameter α corresponds to a relative angle between spins in the two hexagonal layers as we show in Fig. 2(a), and enters in the effective field theory *only* through the kinetic term, since it is a zero mode of the theory for fluctuations that are uniform in space.

Here we make the low-energy approximation $\phi'_2 = \phi'_4 = 0$ for the massive fields, which implies $\phi_2 = \phi_1$ and $\phi_4 = \phi_3$,

and we study the massless theory

$$
\begin{aligned}
\mathcal{S} = \sqrt{3} \int d\tau\, d^2x \Big\{ &\frac{\tilde{K}}{2}\big((\nabla\phi_a)^2 + (\nabla\phi_s)^2\big) \\
&+ \frac{1}{2\tilde{\mu}_s}(\partial_\tau\phi_s)^2 + \frac{1}{2\tilde{\mu}_a}(\partial_\tau\phi_a)^2 + (-4i)\frac{(S-m)}{v}(\partial_\tau\phi_s) \Big\},
\end{aligned}
\tag{16}
$$

where $\tilde{K} = \frac{3}{v}(Sa)^2(J_1 - J_x \cos\alpha)\sin^2\theta^0$, $\tilde{\mu}_{s,a} = \frac{v}{16}\mu_{s,a}$, $\phi_s = (\phi_1 + \phi_3)/2$, $\phi_a = (-\phi_1 + \phi_3)/2$. Notice that all the α dependence is in the parameter $\tilde{K}$ and we have made a rescaling $x \to x/3$.

In the following we drop the factor $\sqrt{3}$ outside the integral in the last equation. The notations ϕ_s and ϕ_a are chosen to emphasize the presence of a symmetrical and an antisymmetrical combination of the fields. At this order the partition function is factorized as

$$
Z = \left(\int \mathcal{D}\phi_s e^{-\mathcal{S}[\phi_s]} \right)\left(\int \mathcal{D}\phi_a e^{-\mathcal{S}[\phi_a]} \right) = Z_s Z_a,
$$

and can be computed analytically. Here we follow the approach from Ref. [43], and we express ϕ_b, $b \in \{s, a\}$ in terms of the crystalline momentum k and the Matsubara frequencies $\omega_n = \frac{2\pi n}{\beta}$, $n \in \mathbb{Z}$, by

$$
\phi_b(\mathbf{r}, \tau) = \sum_{n=-\infty}^{\infty} \frac{1}{2\pi\beta} \int d^2k\, e^{i\mathbf{k}\mathbf{r}} e^{-i\omega_n\tau} \phi_b(\mathbf{k}, \omega_n). \tag{17}
$$

This representation is valid only for fields satisfying $\phi_b(\mathbf{r}, 0) = \phi_b(\mathbf{r}, \beta)$, i.e., without vorticity, and yields $S_{BP} = 0$.

The complete action reads

$$
S = \frac{1}{\beta}\sum_b \sum_{n=-\infty}^{\infty} \frac{1}{2} \int d^2k\, \phi_b^*(\mathbf{k}, \omega_n)\phi_b(\mathbf{k}, \omega_n)\left(\tilde{K}k^2 + \frac{\omega_n^2}{\tilde{\mu}_b} \right).
$$

We calculate the Gaussian functional integral in terms of a dimensionless rescaled field $\phi'(k, \omega_n) = \phi(k, \omega_n)\Gamma/\beta$, where $\Gamma \propto 1/a^2$, obtaining

$$
\log(Z_b) = \frac{-1}{2} \int \frac{d^2k}{\Gamma} \sum_{n=-\infty}^{\infty} \log\left[\frac{\beta}{\Gamma}\left(\tilde{K}k^2 + \frac{\omega_n^2}{\tilde{\mu}_b} \right) \right].
$$

To sum the series, we make use of the identity

$$
\int_1^{\beta\tilde{K}k^2/\Gamma} \frac{dt^2}{\frac{\omega_n^2}{\tilde{\mu}_b\Gamma/\beta} + t^2} = \log\left(\frac{\beta}{\Gamma}\tilde{K}k^2 + \frac{\omega_n^2}{\tilde{\mu}_b\Gamma/\beta} \right) - \log\left(1 + \frac{\omega_n^2}{\tilde{\mu}_b\Gamma/\beta} \right).
$$

Hence, the Helmholtz free energy is

$$
F = -\frac{1}{\beta}\log(Z) = \frac{1}{\beta}\int \frac{d^2k}{\Gamma} \sum_b \log\left[\sinh\left(\frac{1}{2}k\beta\sqrt{\tilde{K}\tilde{\mu}_b} \right) \right],
$$

where we have dropped vacuum contributions, i.e., terms independent of the momentum k and the angle α. The last equation can be rewritten as

$$
F = \int \frac{d^2k}{\Gamma} \sum_b \left\{ \frac{1}{2}k\sqrt{\tilde{K}\tilde{\mu}_b} + \frac{1}{\beta}\log\left(1 - e^{-k\beta\sqrt{\tilde{K}\tilde{\mu}_b}} \right) \right\},
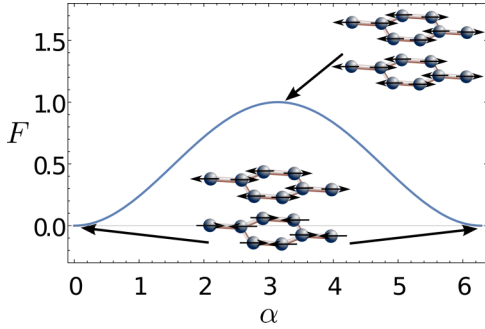$$

FIG. 3. Free energy corresponding to Eq. (18), with $J_1 = 1$, $J_x = 1/10$, $J_0 = 3J_x$, $\theta^0 = \pi/4$, and $\beta = 5$. The free energy presents minimums at $\alpha = 0, 2\pi$ corresponding to antiparallel configuration in the vertical bonds. The maximum at $\alpha = \pi$ corresponds to a parallel configuration in the vertical bonds. For the presentation, we have normalized the free energy such that $F(\alpha = 0) = 0$ and the maximum value of F is 1

where first term is the quantum contribution to the free energy, F_Q, and the second one is the thermal contribution, F_β. The former is integrated directly, giving

$$F_Q = \frac{\pi}{3\Gamma}\sqrt{\tilde{K}}\Lambda^3 \sum_b \sqrt{\tilde{\mu}_b},$$

where $\Lambda = 2\pi/a$ is the momentum cutoff. For F_β we have

$$F_\beta = \frac{2\pi}{\beta^3\Gamma\tilde{K}} \sum_b \frac{1}{\tilde{\mu}_b} \int_0^{\Lambda\beta\sqrt{\tilde{K}\tilde{\mu}_b}} dx\, x \log(1 - e^{-x}),$$

where we define the dimensionless variable $x = k\beta\sqrt{\tilde{K}\tilde{\mu}_b}$. Here we take the low-temperature limit $\beta\Lambda\sqrt{\tilde{K}\tilde{\mu}_b} \gg 1$ and we obtain

$$F_\beta = [-\zeta(3)]\frac{2\pi}{\beta^3\Gamma\tilde{K}} \sum_b \frac{1}{\tilde{\mu}_b},$$

where $-\zeta(3) \equiv \int_0^\infty dx\, x \log(1 - e^{-x}) \approx -1.2$, being $\zeta(s)$ the Riemann zeta function, for $\text{Re}(s) > 1$.

The complete Gaussian (noninteracting) free energy in terms of the bare microscopic parameters and the angle α is

$$F = 6\sqrt{2}\pi^4\left(\sqrt{J_1 - J_x} + \sqrt{J_1 + J_x}\right)\sin(\theta^0)\sqrt{J_1 - J_x\cos(\alpha)}$$
$$- \frac{32\pi J_1\zeta(3)\csc^2(\theta^0)}{81\beta^3(J_1^2 - J_x^2)[J_1 - J_x\cos(\alpha)]}. \tag{18}$$

Both the thermal and the quantum contribution to the free energy have a single minimum at $\alpha = 0$ (Fig. 3), so the Néel configuration is selected by both thermal and quantum fluctuations. Additionally, we see in Eq. (18) that in this approximation, for $J_1 = J_x$ the quantum contribution vanishes and the thermal contribution diverges, suggesting that near $J_1 = J_x = J_0/3$ the thermal contributions dominate against the quantum ones (Fig. 3). A classical analysis at finite temperature was done for this system with $J_1 = J_x = J_0/3$ in Ref. [44]. For this particular choice of magnetic couplings our effective field theory is unstable, given that $m_3 = m_4 = \mu_3 = \mu_4 = 0$. Nonetheless, the presence of an anisotropy term $H_D = \sum_{r,l,\eta} D(S_{l,\eta}^z)^2$, with $D > 0$, would provide a finite contribution to all four μ_j, $j = 1, \ldots, 4$, stabilizing the

magnetization fluctuations. This scenario is beyond the scope of our present work because the point $J_1 = J_x = J_0/3$ is far away from the experimental and numerical estimations of the magnetic interactions in $Bi_3Mn_4O_{12}(NO_3)$, where J_1 is the dominant coupling [36]. In addition, our classical ground state corresponds to the limit of low frustration whereas for $J_1 = J_x = J_0/3$ frustration is dominant and leads to more general classical ground states [44].

So far the results in this section apply for vorticity-free fields ϕ_s, ϕ_a. If instead vortices of ϕ_s proliferate the system enters a short-ranged gapped phase. For this to be allowed the theory must not have a Berry phase term, i.e., the condition $4(S - m) \in \mathbb{Z}$ must hold. In this scenario the symmetric field ϕ_s disorders and consequently the field ϕ_a disorders as well, because both combinations of fields are not independent.

2. Dominant interlayer coupling

If instead in Eq. (14) J_0 is the dominant coupling and we increase the magnitude of J_x, the theory becomes singular at $J_1 = J_x$. In this case both the mass m_4 and the stiffness K simultaneously vanish. In contrast μ_j remains positive for $j = 1, \ldots, 4$ [see Eq. (12)]. As before, the condition $J_1 = J_x$ corresponds to a zero mode of the Hamiltonian parametrized by Eq. (4). In this case we can again make a variation in the parametrization by adding the real parameters $\alpha_{l,\eta}$, $l = 1, 2$, $\eta = 1, 2$ from Eq. (15). If we now set $\alpha_{1,\eta} = \alpha_{2,\eta}$, $\eta = 1, 2$, the classical energy at $T = 0$ is a function of θ^0 only. Again, because of the $U(1)$ symmetry of the system we can take $\alpha_{2,1} \equiv \alpha$ and $\alpha_{1,2} = 0$. This zero mode is presented graphically in Fig. 2(b). The parameter α does not enter in the effective theory this time because the kinetic term vanishes.

For $J_1 = J_x$, in the low-energy limit the two remaining massive fields ϕ_2' and ϕ_3' in Eq. (14) may be set to zero, meaning $\phi_1 = \phi_3$ and $\phi_2 = \phi_4$. If we consider the case $m = 0$, then there is no Berry phase and the vortices in ϕ_1' may proliferate if energetically favorable, leading the system to a gapped short-range phase. In this scenario the gapless field ϕ_4' gets delocalized, and its canonical-conjugate field Π_4' gets localized. The vanishing of the stiffness produces a flat band in the dispersion relation of the magnetic excitations, which imply that they are localized in coordinate space, i.e., do not propagate through the lattice. Similar magnon-crystal ground states are present in other frustrated quantum spin systems in one-, two-, and three-dimensional systems [45]. Our results are the semiclassical description of the exactly factorized ground state for general spin S, as has been done in some frustrated quantum spin chains [46,47]. As a final remark we can mention a rough estimation for the value of $J_1 = J_x$ that corresponds to the end of the dimer phase. It is easy to see from Eqs. (12) and (11) that for $J_1 = J_x > J_0/3$ the masses m_3, μ_3, and μ_4 become simultaneously negative. This means that our noninteracting field theory is unstable with respect to fluctuations ϕ_3, Π_3, and Π_4, and the system should be in a ground state described by another theory.

IV. CONCLUSIONS

In this work we use a path integral approach to study the Heisenberg model on the bilayer honeycomb lattice with

in-plane first-neighbor (J_1) and interlayer first-neighbor (J_0) and second-neighbor (J_x) interactions as a model to describe some of the observed features of $Bi_3Mn_4O_{12}(NO_3)$. Although we have explored different interesting regimes of field theory, we have paid particular attention to the case where the values of the magnetic couplings are close to those obtained experimentally [36] in $Bi_3Mn_4O_{12}(NO_3)$, where the dominant coupling is J_1. In this case, we find that the classical model presents a zero mode parametrized by the relative angle between the spins of each plane. The low-energy effective theory of quantum fluctuations can be written in terms of a symmetric field, related to global magnetization, and an antisymmetric field related to the spin imbalance between layers. We show that the presence of quantum and thermal fluctuations selects the collinear state from the degenerate manifold of classical ground states. This result holds for vorticity-free configurations, and coincides with the magnetic order observed experimentally [35] when a high enough magnetic field is applied, validating the scenario that the experimental observations can be explained with no significant frustration in the honeycomb plane but with frustrating intrabilayer interactions and also showing that this mechanism is only present in the range of parameters corresponding to the material.

If the magnetization of the system is such that $4(S - m) \in \mathbb{Z}$, then vortices may proliferate if the renormalized stiffness is small enough, driving the system to a gapped phase with only short-range antiferromagnetic ordering. This mechanism could explain the short-range correlations observed in $Bi_3Mn_4O_{12}(NO_3)$ at low temperatures and low magnetic fields.

Additionally, we show that if J_0 is the dominant coupling, the known [32] factorized ground state of a valence-bond solid composed by a singlet array over the J_0 bonds is interpreted in the field theory as a vanishing stiffness and spin-wave velocity. As was shown for the factorized valence-bond solid ground state, our approach allows one to also show that the behavior predicted in the presence of a nonzero magnetization for spin $\frac{3}{2}$ remains true for a generic value of the spin. We hope that our results will encourage further experimental investigations on $Bi_3Mn_4O_{12}(NO_3)$, in particular the ordering transition triggered by an order by disorder mechanism.

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