Magnon condensation with finite degeneracy on the triangular lattice

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We study the spin 1/2 triangular-lattice $J_1$-$J_2$-$J_3$ antiferromagnet close to the saturation field using the dilute Bose gas theory, where the magnetic structure is determined by the condensation of magnons. We focus on the case of ferromagnetic $J_1$ and antiferromagnetic $J_2$, $J_3$, that is particularly rich because frustration effects allow the single-magnon energy dispersion to have six-fold degenerate minima at incommensurate momenta. Our calculation also includes an interlayer coupling $J_0$, which covers both antiferromagnetic and ferromagnetic cases including negligibly small regime (two-dimensional case). Besides the spiral and fan phases, we find a new double-$q$ phase (superposition of two modes), dubbed “$Q_0$-$Q_1$” (or simply “01”) phase, that enjoys a new type of multiferroic character. Certain phase boundaries have a singular $J_0$ dependence for $J_0 \rightarrow 0$, implying that even a very small interlayer coupling drastically changes the ground state. A mechanism for this singularity is presented. Moreover, in some regions of the parameter space, we show that a dilute gas of magnons can not be stable, and phase separation (corresponding to a magnetization jump) is expected. In the $J_1$-$J_2$ model ($J_3 = 0$), formation of two-magnon bound states is observed, which can lead to a quadrupolar (spin-nematic) ordered phase. Exact diagonalization analysis is also applied to the search of bound states.

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I. INTRODUCTION

Frustrated spin systems are privileged hosts of exotic phases of matter. Quantum spin liquids, quantum spin nematics, and topological spin textures (such as skyrmion and vortex crystals) among others have recently attracted a lot of theoretical and experimental interest. In theoretical analysis, however, fully quantum mechanical treatments of these systems present in general huge difficulties, most notoriously for the sign problem of quantum Monte-Carlo simulations. It is therefore crucial to establish and develop fully quantum methods of exploring at least some parts of the magnetic phase diagram.

Since the pioneering work of Batyev and Braginskii\textsuperscript{4}, the dilute Bose gas theory of magnons near saturation field has become one of the few approaches that deal with magnetic systems in a fully quantum-mechanical fashion. In this theory, magnetic systems are mapped to interacting hard-core Bose gas of magnons\textsuperscript{5} and the magnetic state near saturation is described as a dilute condensed Bose gas. With the work of Nikuni and Shiba\textsuperscript{6} on the prototypical triangular Heisenberg antiferromagnet it was made clear that frustration can induce various types of Bose-Einstein condensation (BEC), hence new magnetic phases, thanks to the more complex low-energy structure of magnons and their interaction. These states are in general characterized by the coherent superposition of one or more spirals, from which the terminology single-$q$ and multiple-$q$ states comes.

While the triangular-lattice Heisenberg antiferromagnets (and helimagnets in general\textsuperscript{7}) near saturation can accommodate only single-$q$ (spiral) or coplanar double-$q$ (fan) phases, here we are interested in new kinds of multiple-$q$ phases that appears due to condensation of magnons at (unusual) multiple momenta. In this respect, a necessary condition is a high degeneracy of inequivalent single-magnon energy minima (in momentum space), which is typically brought about by competing exchange interactions. For this purpose, we start in this paper from the triangular-lattice $J_1$-$J_2$-$J_3$ model with ferromagnetic $J_1$ and antiferromagnetic $J_2$, $J_3$ near saturation, which in a certain range of parameters features six energy minima at $\pi/3$ rotation-symmetric momenta inside the Brillouin zone. Besides being pedagogical for our study, this model has been proposed for several materials, such as NiBr\textsubscript{2} (Ref. \textsuperscript{8} and NiGa\textsubscript{2}S\textsubscript{4} (Ref. \textsuperscript{9}), both with spin $S = 1$. Moreover, a recent classical Monte-Carlo study, for a specific choice of exchange couplings, reported the appearance of an exotic triple-$q$ state, which is accompanied by skyrmion lattice, at finite temperature in intermediate applied magnetic field\textsuperscript{10}. In this paper, to study possible magnetic phases of the $S = 1/2$ triangular-lattice $J_1$-$J_2$-$J_3$ antiferromagnet near the saturation field, we use the dilute Bose gas theory. In Sec. \textsuperscript{11} we write down the general form of ground-state energy à la Ginzburg-Landau for six complex order parameters corresponding to the condensed magnon modes in the dilute limit. We stress that the same type of effective theory can arise from very different microscopic Hamiltonians. A recent attractive example is given by the spin-dimer compound Ba\textsubscript{3}Mn\textsubscript{2}O\textsubscript{8}, which features magnetic triangular lattices with non-trivial stacking and interlayer exchange couplings\textsuperscript{12}. All of the effective coupling constants in this energy func-
tional can be calculated from the microscopic model in the dilute Bose gas approximation. This will be done in two ways. First we consider layered systems with a finite, eventually very small, interlayer coupling; while the relevant physics still comes from the triangular lattice, the three-dimensionality naturally protects the calculation from infra-red singularities. Besides, we take a purely two-dimensional (2D) approach, in which an infra-red momentum cutoff is introduced as a regularization. It should be however noted that the latter approach requires the assumption of a stable low-density single-magnon Bose gas. In the present model we find that various instabilities that may affect the existence of dilute single-magnon gas can not be captured in this approach.

Minimization of the ground-state energy leads to the phase diagrams of Figs. [1] and [2] which are the main results of this paper. In particular, besides the well-known spiral and fan phases, we find in quite extended regions a new phase ("01" phase) with a striped chiral order and new multi-ferroic properties, as described in Sec. IV C. Also, we show that the presence of ferromagnetic exchange interactions can sometimes induce an effective attraction between magnon modes, causing an instability of the dilute magnon gas for weak interlayer coupling regime. In this situation a field-induced first-order phase transition (magnetization jump) or a transition to a different quantum phase (not described by a single-magnon BEC) is typically expected. It has been discussed that ferromagnetic interactions sometimes induce formation of two-magnon bound states, which give rise to spin nematic ordering. In our model, we indeed find that two-magnon bound states are more stable than single magnons in a certain parameter region inside of the "phase separation" region. We also applied exact diagonalization analysis of finite-size systems in this parameter region, which indicates a small magnetization jump at the saturation field and a tendency toward spin nematic ordering below the jump.

Comparison between purely two-dimensional analysis and quasi-two-dimensional analysis with weak interlayer coupling also reveals that the shape of a phase boundary can have strong interlayer coupling ($J_0$) dependence in the weak $J_0$ limit. The "01" phase in Fig. [1] extends to the weak $J_0$ regime, such as $J_0 \sim 10^{-4}$, but purely two-dimensional analysis concludes that this phase cannot appear in the two-dimensional system near saturation. We discuss that this singularity comes from the logarithmic correction of the effective coupling $\Gamma \sim \alpha / (\log |J_0|) + \mathcal{O}(1/(\log |J_0|)^2)$ and the phase boundary between the "01" phase and fan phase presumably has a logarithmic singularity, going rapidly down to the $J_0 = 1/4$ point in the $|J_0| \to 0$ limit.

The paper is organized as follows: In Sec. II we briefly describe the model and degeneracy in the single-magnon energy dispersion at saturation field. In Sec. III we discuss the dilute Bose gas theory for describing magnon condensation at multiple momenta, explaining how effective couplings are calculated from the microscopic model. In Sec. IV we present results of phase diagrams and characteristic of each phase. In Sec. V we conclude with a summary and discussions.

### II. MODEL

We consider the spin $S = 1/2$ $J_1$-$J_2$-$J_3$ model on the triangular lattice in applied magnetic field at zero temperature and, including an interlayer coupling, we also consider the model on the hexagonal lattice. The Hamiltonian reads

$$
\mathcal{H} = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle i,j \rangle_{2nd}} \mathbf{S}_i \cdot \mathbf{S}_j + J_3 \sum_{\langle i,j \rangle_{3rd}} \mathbf{S}_i \cdot \mathbf{S}_j + J_0 \sum_{\langle i,j \rangle_5} \mathbf{S}_i \cdot \mathbf{S}_j - H \sum_i S_i^z,
$$

(1)

where $\langle i,j \rangle$ counts nearest neighbor bonds, $\langle i,j \rangle_{2nd}$ counts next-nearest neighbor bonds, and $\langle i,j \rangle_{3rd}$ counts 3rd-nearest neighbor bonds on the triangular-lattice layers. The $J_0$ term represents the nearest-neighbor (NN) coupling between adjacent layers. In this paper, we focus on ferromagnetic (negative) $J_1$, fixing $J_1 = -1$ without loss of generality.

The saturation field is defined as the value of the applied magnetic field at which all spins are polarized. Slightly below the saturation field the magnetic excitations are interacting hard-core bosons (magnons). The bosonic vacuum corresponds to the fully polarized state. Using the hard-core boson map of spin 1/2 operators ($S_i^+ = a_i^\dagger$, $S_i^z = 1/2 - a_i^\dagger a_i$) the Hamiltonian in Fourier space becomes, modulo constant terms,

$$
\mathcal{H} = \sum_k \left[ \epsilon(k) - \mu \right] a_k^\dagger a_k + \frac{1}{2N} \sum_{k,k',q} V(q) a_k^\dagger a_{k+q} a_{k'}^\dagger a_{-q} a_k,
$$

(2)

where $N$ is the number of lattice sites and

$$
\epsilon(k) = J_1 \nu(k) + J_2 \gamma(k) + J_3 \sigma(k) + J_0 \cos k_z + |J_0|,
$$

(3)

$$
\nu(k) = \sum_{i=0}^{2} \cos a_{2i} \cdot k,
$$

(4)

$$
\gamma(k) = \sum_{i=0}^{2} \cos b_{2i} \cdot k,
$$

(5)

$$
\sigma(k) = \sum_{i=0}^{2} \cos c_{2i} \cdot k,
$$

(6)

$$
V(q) = 2 \epsilon(q) - |J_0| + U,
$$

(7)

$$
\mu = 3(J_1 + J_2 + J_3) + |J_0| - H.
$$

(8)
the triangular lattice. $U$ represents a repulsive on-site interaction, which will be eventually sent to infinity to implement the hard-core condition. The saturation field is given by $H_c = 3(J_1 + J_2 + J_3) + |J_0| - \epsilon_{\text{min}}$, where $\epsilon_{\text{min}} = \min_k \epsilon(k)$.

The single-magnon energy minima have qualitatively different structure depending on the value of the exchange couplings. For ferromagnetic $J_1$ ($J_1 = -1$), there are two interesting regions in the $J_2$-$J_3$ plane (see Fig. 1), with six degenerate minima at inequivalent (generically incommensurate) wave-vectors. In region I, they are $Q_0 = (k_0^I, 0, 0)$ (resp. $Q_0 = (k_0^I, 0, \pi)$) for $J_0 < 0$ (resp. for $J_0 > 0$) and all $\pi/3$ rotations thereof around the $k_z$ axis; $k_0^I$ is given by

$$k_0^I = 2\cos^{-1}\left(\frac{2J_3 - 3J_2 + \sqrt{(3J_2 + 2J_3)^2 + 8J_4}}{8J_3}\right).$$

In region II, we instead define $Q_0 = (0, k_0^{II}, 0)$ (resp. $Q_0 = (0, k_0^{II}, \pi)$) for $J_0 < 0$ (resp. for $J_0 > 0$), with

$$k_0^{II} = \frac{2}{\sqrt{3}}\cos^{-1}\left(\frac{1 - J_2}{2(J_2 + 2J_3)}\right),$$

and the other ones are generated by the same rotational symmetry.

In this paper we are interested in these two areas, where the system can possibly host new multiple-$q$ phases. In particular, we concentrate on two representative semi-infinite lines, namely i) $J_2 = 0, J_3 > 1/4$ for region I and ii) $J_3 = 0, J_2 > 1/3$ for region II, which correspond to the $J_1$-$J_2$ model and the $J_1$-$J_3$ model respectively. We do not expect qualitative differences for other choices of the parameters within the two regions.

III. MAGNON CONDENSATION WITH FINITE DEGENERACY

For applied magnetic field $H$ above the saturation field $H_c$, or in other words for $\mu < \epsilon_{\text{min}}$, all spins are aligned along the direction of the field, which corresponds to the absence of magnons. When $H$ is tuned slightly below $H_c$ we expect a dilute gas of magnons, most of which occupy the lowest energy states.

A. Ground-state energy in the dilute limit

The six inequivalent single-magnon minima, denoted $\{Q_i\}_{i=0,\ldots,5}$, are arranged for region I as in Fig. 2, where we depict the appropriate section of the Brillouin zone (for region II they are rotated by $\pi/2$). We introduce the (complex) order parameters $\langle a_{Q_i} \rangle = \sqrt{N}\psi_{Q_i}$, $(i = 0, \ldots, 5)$ referring to particles condensed at the six different wave-vectors $Q_i$. In the dilute limit the ground-state energy per site can be written, by exploiting the symmetries of the system (six-fold rotation and mirror
metries, namely the product of three “chiral” symmetries), as
\[
\frac{\mathcal{E}}{N} = (\epsilon_{\text{min}} - \mu) \sum_{i=0}^{5} |\psi_{q_{i}}|^2 + \frac{1}{2} \Gamma^{(1)} \sum_{i=0}^{5} |\psi_{q_{i}}|^4 \\
+ \sum_{i=0}^{5} \sum_{j=1}^{2} \Gamma^{(2)}_{j} |\psi_{q_{i}q_{j}}|^2 |\psi_{q_{i+j}}|^2 \\
+ \sum_{i=0}^{2} \sum_{j=0}^{2} \Gamma^{(3)}_{j} |\psi_{q_{i}}\psi_{q_{i+j}}\psi_{q_{i+j}}\psi_{q_{i+j}}|^2 
\] (11)
and higher orders in the condensate amplitudes can be neglected. The coefficients \(\Gamma^{(1)}\) and \(\Gamma^{(2)}\) are the effective vertices, namely renormalized four-point functions, describing the interaction between condensed particles, that in the dilute regime can be determined by a full quantum mechanical calculation as first shown by Beliaev.\(^{2}\) The energy \(\mathcal{E}/N\) is clearly real-valued, even though not all of the quartic terms are density-density type; in particular, the last term of Eq. (11) depends on the relative phases of the condensates. This is a peculiarity of our theory, originating essentially from the presence of frustrated non-NN exchange.\(^{13}\) Note that, while there is only one global \(U(1)\) symmetry in the original spin model [Eq. (1)], the low-energy effective theory in the dilute limit enjoys an additional emergent symmetry, namely the product of three “chiral” symmetries \(U(1)_{j} (j = 0, 1, 2)\) acting as \((\psi_{q_{j}}, \psi_{-q_{j}}) \rightarrow (e^{i\alpha_{j}} \psi_{q_{j}}, e^{-i\alpha_{j}} \psi_{-q_{j}})\).

In order to find the effective couplings \(\Gamma^{(n)}\) in Eq. (11) we must calculate the renormalized scattering amplitude\(^{15}\) \(\Gamma(q; k, k')\) at low density (many-body \(T\)-matrix) for initial momenta \(k, k' \in \{Q_{i}\}\). The effective couplings are given by the following combinations:
\[
\Gamma^{(1)} = \Gamma(0; Q_{0}, Q_{0}), \\
\Gamma^{(2)} = \Gamma(0; Q_{0}, Q_{1}) + \Gamma(Q_{1} - Q_{0}; Q_{0}, Q_{1}), \\
\Gamma^{(3)} = \Gamma(0; Q_{0}, Q_{2}) + \Gamma(Q_{2} - Q_{0}; Q_{0}, Q_{2}), \\
\Gamma^{(3)} = \Gamma(Q_{1} - Q_{0}; Q_{0}, Q_{3}), \\
\Gamma^{(3)} = \Gamma(Q_{2} - Q_{0}; Q_{0}, Q_{3}), \\
\Gamma^{(3)} = \Gamma(-Q_{1} - Q_{0}; Q_{0}, Q_{3}), \\
\Gamma^{(3)} = \Gamma(-Q_{2} - Q_{0}; Q_{0}, Q_{3}) = \Gamma^{(3)}_{1}.
\] (17)
The strategy for the calculation is presented in the following Section III B.

B. Bethe-Salpeter equation

In the dilute limit \(\Gamma(q; k, k')\) satisfies the Bethe-Salpeter equation for the ladder approximation, which reads
\[
\Gamma(q; k, k') = V(q) - \frac{1}{N} \sum_{q' \in BZ} \sum_{\langle q' \rangle \sim -\epsilon_{\text{min}} > \mu} \frac{V(q - q')}{\epsilon(k + q') + \epsilon(k' - q') - 2\epsilon_{\text{min}} - E} \Gamma(q'; k, k').
\] (18)

While in three dimensions this gives a finite result that is correct also at low but non-vanishing density up to small correction of order \(\mu - \epsilon_{\text{min}}\), it is well-known that the two-body scattering amplitude vanishes logarithmically with lowering the density in two dimensions,\(^{15}\) due to the non-integrable singularities in the kernel of Eq. (18). Thus finite density (many-body) effects become important. In fact, we expect that at energies lower than \(\mu - \epsilon_{\text{min}}\) the magnon dispersion is modified à la Bogoliubov and becomes linear. In the calculation it is therefore required to cutoff the integration in the neighborhoods where \(\epsilon(k + q'), \epsilon(k' - q') \leq \mu\). This is the meaning of the cutoff introduced in Eq. (18). Whereas it is possible to work directly in momentum space, we choose the more convenient treatment of Refs. 19 (see also Ref. 20), where it is shown that calculating Eq. (18) at negative energy \(E = -C(\mu - \epsilon_{\text{min}})\) \((C\) a numerical constant of order 1) without momentum cutoff yields an equivalent result at leading order in \(\mu - \epsilon_{\text{min}}\). This procedure will be used in

\[\]
Section IV while in the following Section V we take a different approach, namely we consider the system with a non-vanishing interlayer coupling, thus avoiding the subtleties appearing in two dimensions.

IV. PHASE DIAGRAMS: QUASI-2D SYSTEMS WITH INTERLAYER COUPLING

We now consider layered systems with non-vanishing interlayer NN exchange coupling \( J_0 \). Whereas the interesting physics is essentially delivered by the triangular lattice planes, the sign of \( J_0 \) determines the relative ordering of two adjacent planes.

The solution of Eq. (18) can be obtained by expanding in lattice harmonics, that is by taking the Ansatz

\[
\psi_j = \sum \Gamma(q) e^{i q \cdot j},
\]

where \( \langle \Gamma(q) \rangle = (1/N) \sum_q \Gamma(q) \). The calculation of the effective coupling is detailed in Appendix A; in this case we do not need any cutoff procedure since all integrals are finite.

By plugging the result into and minimizing Eq. (11) we obtain the phase diagrams of the \( J_1-J_3 \) and \( J_1-J_2 \) models, which are shown in Figs. 4 and 5 respectively. It is interesting to note that in the classical limit, these models always have the spiral state in their ground state manifold in the present parameter space. However, this phase disappears in most cases due to quantum effects and is replaced with other new quantum phases.

Below we describe the characteristics of the different regions composing the phase diagrams.

A. Spiral phase

Magnon condensation at a single wave-vector, say \( \psi_{Q_0} = \sqrt{\rho e^{i \alpha}} \) and \( \psi_{Q_i} = 0 \) for \( i = 1, \cdots, 5 \), yields the so-called spiral phase, whose spin structure is

\[
\langle S_j^\alpha \rangle = \sqrt{\rho} \cos (Q_0 \cdot r_j + \alpha),
\]

\[
\langle S_j^\beta \rangle = \sqrt{\rho} \sin (Q_0 \cdot r_j + \alpha),
\]

\[
\langle S_j^\gamma \rangle = \frac{1}{2} - \rho.
\]

The magnon density is given by \( \rho = (\mu - \epsilon_{\text{min}})/\Gamma^{(1)} \). This phase breaks the \( C_6 \) rotation symmetry and reflection symmetry, and is accompanied by a vector chiral order

\[
\langle S_r \times (S_{r+a_i}) \rangle^2 = \rho \sin (Q_0 \cdot a_i).
\]

As noted already in Ref. 3 this phase shows multi-ferroic behaviour due to the spin-current mechanism which generates an electric polarization \( P_e = \eta e \times (\langle S_r \times (S_{r+a_i}) \rangle \times (S_{r+a_i}) \rangle \).

B. Fan phase

In this phase magnons condense simultaneously at two opposite wave-vectors, e.g. \( Q_0 \) and \( Q_3 \equiv -Q_0 \) with the same density. We can choose the parametrization \( \psi_{Q_0} = \sqrt{\rho e^{i \alpha_1}} \) and \( \psi_{Q_3} = \sqrt{\rho e^{i \alpha_2}} \) with \( \rho = (\mu - \epsilon_{\text{min}})/\Gamma^{(1)} + \Gamma^{(3)} \), and define \( \theta = \alpha_1 + \alpha_2, \phi = \alpha_2 - \alpha_1 \). The spin
The state is given by

\[
\langle S^z_j \rangle = 2\sqrt{\rho} \cos \left( \frac{Q_1 - Q_0}{2} \cdot r_j + \frac{\phi}{2} \right) \cos \left( \frac{Q_1 + Q_0}{2} \cdot r_j + \frac{\theta}{2} \right),
\]

\[
\langle S^y_j \rangle = 2\sqrt{\rho} \cos \left( \frac{Q_1 - Q_0}{2} \cdot r_j + \frac{\phi}{2} \right) \sin \left( \frac{Q_1 + Q_0}{2} \cdot r_j + \frac{\theta}{2} \right),
\]

\[
\langle S^z_j \rangle = \frac{1}{2} - 4\rho \cos^2 \left( \frac{Q_1 - Q_0}{2} \cdot r_j + \frac{\phi}{2} \right),
\]

This state has a coplanar spin structure; The spins oscillate within a fixed plane parallel to the z-axis and identified by the angle \( \theta \). This phase breaks only \( C_3 \) symmetry and is not accompanied by chiral symmetry breaking. The vector chirality \( \langle S_x \rangle \times \langle S_{r+e} \rangle \) always vanishes on average and no multi-ferroic property can appear.

C. “01” phase

In the regions denoted by “01-Q1” in Figs. 4 and 5, magnons equally occupy the lowest-energy states of two adjacent wave-vectors, e.g. \( Q_0 \) and \( Q_1 \). The spin structure is given by

\[
\langle S^z_j \rangle = 2\sqrt{\rho} \cos \left( \frac{Q_1 - Q_0}{2} \cdot r_j + \frac{\phi}{2} \right) \cos \left( \frac{Q_1 + Q_0}{2} \cdot r_j + \frac{\theta}{2} \right),
\]

\[
\langle S^y_j \rangle = 2\sqrt{\rho} \cos \left( \frac{Q_1 - Q_0}{2} \cdot r_j + \frac{\phi}{2} \right) \sin \left( \frac{Q_1 + Q_0}{2} \cdot r_j + \frac{\theta}{2} \right),
\]

\[
\langle S^z_j \rangle = \frac{1}{2} - 4\rho \cos^2 \left( \frac{Q_1 - Q_0}{2} \cdot r_j + \frac{\phi}{2} \right),
\]

where the condensate density is \( \rho = (\mu - \epsilon_{\text{min}})/(\Gamma^{(0)} + \Gamma^{(2)}) \). This phase somehow interpolates between the spiral and fan phases. In fact, along the direction of \( Q_1 + Q_0 \) the spins spiral with pitch vector \( (Q_1 + Q_0)/2 \), whereas along the orthogonal direction they oscillate in the fan state (see Fig. 6). The \( z \)-component of vector chiral order exists forming a stripe structure,

\[
\langle (S_z) \times (S_{z+1}) \rangle^z = 4\rho \cos^2 \left( \frac{Q_1 - Q_0}{2} \cdot r_j + \frac{\phi}{2} \right) \sin \left( \frac{Q_1 + Q_0}{2} \cdot 1 \right)
\]
For bonds in the $l$ direction ($l \parallel Q_1 + Q_0$), this expression simplifies to

$$P_1 = 4 \eta \rho \cos^2 \left( \frac{Q_1 - Q_0}{2} \cdot r_j + \frac{\phi}{2} \right) \sin \left( \frac{Q_1 + Q_0}{2} \cdot l \right) (1 \times \hat{H}).$$  \hspace{1cm} (27)$$

The main peculiarity compared to the spiral case is that the amplitude of the polarization is modulated along one direction, but does not change sign, thus yielding a striped structure with non-zero net average over a period.

D. Phase separation (PS)

If at least one of $\Gamma^{(1)}$, $\Gamma^{(1)} + \Gamma^{(2)}_1$, and $\Gamma^{(1)} + \Gamma^{(3)}_0$ becomes negative the system suffers from instabilities as is clear from the runaway behaviour of Eq. $\text{11}$. As discussed recently in Ref. $\text{10}$, in this situation a state with low density of magnons can not be stable. The system instead undergoes a field-induced first-order phase transition, featuring phase separation between the fully polarized state and a low-magnetization state. Technically, the latter can be stabilized by including higher order terms in the ground state energy Eq. $\text{11}$ (e.g. sixth order in the $\psi$’s). By looking at which of the above three combinations of couplings is (the most) negative, one can argue about the nature of the low-magnetization state. For instance, if $\Gamma^{(1)} < 0$ and all others positive, it is reasonable to expect a low-magnetization spiral state, etc..

E. Bound states (BS)

In the $J_1-J_2$ model at relatively small interlayer coupling there exist regions where bound states of two magnons are formed (see Fig. $\text{5}$). This can be inferred from the appearance of a pole singularity in $\Gamma^{(1)}$, that is essentially a two-particle Green’s function at zero frequency. The presence of a bound state branch in the spectrum below the single-magnon states would suggest the occurrence of BEC of bound states and thus a spin nematic phase.\cite{Note2} We however do not know how the bound states interact and therefore we can not make any statement about the stability of the spin nematic phase only from this analysis. If the interaction is attractive, there will be again phase separation. Moreover, we can not rule out the existence of three magnon bound states or higher.

To examine the appearance of spin nematic phase we performed exact diagonalization study of the purely two-dimensional model ($J_0 = 0$) with $N = 36$ and 48 spins with the fixed choice of parameters $J_1 = -1$ and $J_2 = 1$. We used finite-size clusters with high space symmetry ($C_{3v}$) and under the periodic boundary condition. The magnetization process is plotted in Fig. $\text{7}$. In case of 36 spins, we find a weak signature of formation of three-magnon bound states from saturation down to low magnetization, which corresponds to the change of total magnetization by three ($\Delta S^z = 3$).\cite{Note2} However, this seems to be an artefact of a small size system, since for the larger size system ($N = 48$), the magnetization process does not posses this periodicity and, instead, it shows a tendency to the formation of two-magnon bound states; the lowest energy states in even number $S^z$ sectors have lower energy than in odd $S^z$ sectors, giving rise to wide steps at even $S^z$ in the magnetization process. Clearly the stability of this spin nematic phase below the magnetization jump remains to be studied further because finite-size effects can still be strong for $N = 48$. 

Figure 6. Spin structure of the “01” phase.

Figure 7. Magnetization process of the $S = 1/2$ triangular-lattice $J_1-J_2$ model with $J_1 = -1$ and $J_2 = 1$ for $N = 36$ and 48 spin clusters. In the smaller size system ($N = 36$), the total magnetization always changes by $\Delta S^z = 3$, but, in the larger size system ($N = 48$), it has a jump $\Delta S^z = 4$ below saturation and wide steps at even values of $S^z$ below the jump, showing a tendency to the formation of two-magnon bound states.
F. Purely 2D calculation

To compare the quasi-two-dimensional systems with purely two-dimensional systems, we analyze effective coupling $\Gamma$ in two dimensions, using the procedure described in Sec. 3.1.1 At small energy cutoff $E < 0$ of order $\mu - \epsilon_{\text{min}}$, the $\Gamma$’s in Eq. 17 have a $1/|\log |E||$ expansion that looks like

$$\Gamma^{(1)}(i) = \frac{\alpha^{(1)}(i)}{|\log |E||} + O \left( \frac{1}{|\log |E||^2} \right), \quad (28)$$

$$\Gamma^{(m)}(i) = \frac{\alpha^{(m)}(i)}{|\log |E||} + O \left( \frac{1}{|\log |E||^2} \right). \quad (29)$$

The leading coefficients can be calculated analytically as described in Appendix A. The results are shown in Fig. 8 for the case of the $J_1$-$J_3$ model, from which we can see that

$$\alpha^{(1)} = \alpha^{(3)}_0, \quad \alpha^{(2)} = \alpha^{(2)}_1, \quad \alpha^{(3)} = 0, \quad (30)$$

$$0 < \alpha^{(1)} < \alpha^{(2)}_1$$

whereby the ground-state energy becomes

$$\frac{\xi_0}{N} = (\epsilon_{\text{min}} - \mu) \sum_{i=0}^5 |\psi_i|^2 + \frac{1}{2} \frac{\alpha^{(1)}}{|\log |E||} \left( \sum_{i=0}^5 |\psi_i|^4 + 2 \sum_{j=0}^2 |\psi_i|^2 |\psi_{i+3}|^2 \right) + \frac{\alpha^{(2)}_1}{|\log |E||} \sum_{i=0}^2 \sum_{j=1}^2 |\psi_i|^2 |\psi_{i+j}|^2$$

$$+ \frac{\alpha^{(2)}_1}{|\log |E||} \left[ (|\psi_0|^2 + |\psi_1|^2)(|\psi_1|^2 + |\psi_2|^2) + (|\psi_0|^2 + |\psi_2|^2)(|\psi_2|^2 + |\psi_3|^2) + (|\psi_1|^2 + |\psi_3|^2)(|\psi_3|^2 + |\psi_4|^2) \right], \quad (32)$$

which exhibits an emergent $U(2)^3$ symmetry. Namely in the zero density limit, $\mu \to \epsilon_{\text{min}} \to 0$, each $U(1)_i$ is effectively enhanced to $U(2)_i$, whose elements transform the doublet $(\psi_i, \psi_{i+3})$. This is reflected in the degeneracy of a continuous family of physically distinct ground states, defined by

$$|\psi_{Q,i}|^2 + |\psi_{Q_{i+3}}|^2 = \frac{\mu - \epsilon_{\text{min}}}{\Gamma(1)} \quad \text{for a certain } i \in \mathbb{Z}, \quad (33)$$

for a certain $i$. To the level of approximation of Eq. 32 the $U(2)_i$ symmetry is spontaneously broken to $U(1)_i$ by choosing a ground state out of the space Eq. 33. The spiral phase (see Sec. 1V.A) and coplanar (fan) phase (see Sec. 1V.B) are just two states in this ground-state manifold. This symmetry enhancement is analogous to that occurring at low-energy in a mixture of two species of dilute Bose gases with equal masses in the continuum in two dimensions.24 It is worth noting, however, that experiments with cold atoms are typically done at fixed number of particles, whereas in magnetic systems the chemical potential, which is determined by the applied magnetic field, can be actually made vanishingly small.

In the phase diagrams (Figs. 4 and 5) with interlayer couplings, we however find different phases. The lower part of the phase diagrams, where $J_0$ is as small as $10^{-4}$ and the system looks almost two-dimensional, shows the fan phase only for $J_3 \gtrsim 1.5$ and for $J_2 \gtrsim 5.5$, respectively. This is because, as described in Sec. 3.1.1, the calculation in two dimensions is intrinsically based on the assumption that a stable single-magnon condensate exists as the many-body ground state and the elementary excitations are quasiparticles with Bogoliubov-like dispersion. If this assumption is violated, approximating the many-body $T$-matrix with the two-body $T$-matrix calculated with negative energy cutoff is not justified. For example, if a first-order phase transition occurs between the ferromagnetic state (magnon vacuum) and a low-magnetization state.
Note that the coefficients of the leading terms, $\alpha^{(2)}_1$ and $\alpha^{(3)}_0$, are the same as in Eq. (30). We know from Fig. 8 that these coefficients are two different regular functions of $J_3 \equiv J_3 - 1/4$ satisfying $\alpha^{(2)}_1 \geq \alpha^{(3)}_0$. Assuming that $\alpha^{(2)}_1$ and $\alpha^{(3)}_0$ are regular, we can expand Eq. (34) in powers of $J_3$ as

$$a \log |J_0| + \left( B + \frac{b}{|\log |J_0||} \right) J_3 + \ldots = 0,$$

where $a = \lim_{J_3 \to 0} (\alpha^{(2)}_1 - \alpha^{(3)}_0)$, $B = \lim_{J_3 \to 0} \partial (\alpha^{(2)}_1 - \alpha^{(3)}_0)/\partial J_3$, and we have used the relation $\lim_{J_3 \to 0} (\alpha^{(2)}_1 - \alpha^{(3)}_0) = 0$. At small $J_3$, we can retain only the first two terms in the above equation, so that there exists an approximate solution for the phase boundary

$$J_3^{sc} \approx - \frac{a}{B |\log |J_0||}.$$

Due to this logarithmic singularity the phase boundary can rapidly shift to the $J_3 = 1/4$ point with decreasing $|J_0|$. The “01”-fan phase boundary in Fig. 4 indeed appears to start this logarithmic behavior for the lower values of $|J_0|$. We thus believe that the “01” phase will eventually disappear in the pure 2D limit due to this mechanism. We note that if the two coefficients $\alpha^{(2)}_1$ and $\alpha^{(3)}_0$ were equal for $J_3 > 1/4$, we could not obtain this logarithmic singularity, and in general there is no reason to expect it. We are led to conclude that, given a microscopic model whose ground state energy is in the form of Eq. (11), the transition between 3D to 2D can be either smooth or singular, according to the value of the leading coefficients $\alpha^{(i)}$ of the effective couplings.

V. CONCLUSION AND DISCUSSION

In conclusion, we have studied, in a magnon Bose-Einstein condensation picture, the triangular $J_1$-$J_2$-$J_3$ antiferromagnet as a prototypical model where a combination of competing exchange interactions and geometrical frustration makes the single-magnon energy minima more-than-doubly degenerate. We focused on the high applied magnetic field regime, just below the saturation field, where the two-magnon interaction can be treated quantum-mechanically by means of the dilute Bose gas theory, and determined the zero-temperature phase diagram as a function of the exchange couplings. Together with the spiral and fan phases (commonly featured in the phase diagram of helimagnets) we found an interesting new phase, the “01” phase, whose physical properties are in some sense halfway between the former two. In the spiral phase, magnons are condensed to a single wave-vector, whereas in both fan and “01” phases magnons are condensed to two wave-vectors with an equal density. While the fan phase is non-chiral, the spiral and “01” phases possess chiral order. The peculiarity of the chiral order in the “01” phase is its stripe structure, which results in a novel type of multiferroic. By studying the singular behavior of the relevant phase boundary as a function of the interlayer coupling $J_3$, we explained how the “01” phase disappears in the purely 2D limit. This mechanism shows that even a very small interlayer coupling can drastically change the ground state.

Also, we elucidated the circumstances in which the dilute single-magnon Bose gas picture breaks down; this occurs quite often when competing ferromagnetic exchange is present. From the discussion in Sec. IV we can expect that the condensed state in such a case is either a low-magnetization state (with finite density of magnons) or a dilute Bose gas of multi-magnon bound states. We can identify the first case by a runaway behavior, i.e., instability, in the dilute Bose gas theory, which leads to phase separation. The second case can be captured by the appearance of stable multi-magnon bound states. In the $J_1$-$J_2$ model, we found that bound two-magnons can have a lower energy than single magnons around $J_2 = 1$ with $J_1 = -1$ for a weak interlayer coupling regime. Exact diagonalization study of the purely two-dimensional model at $J_2 = 1$ with 48 spins indeed indicates a small magnetization jump at the saturation field and a tendency to the two-magnon pairing below the magnetization jump. This would correspond to a weak first-order
phase transition to spin-nematic state, but this picture needs further confirmation since finite-size effects might be strong in our numerical calculation.

We also note that the boundary between a dilute Bose gas of magnons (spiral, fan or “01”) and phase separation in Figs. 4 and 5 corresponds to a tricritical point on the \( J_3-M \) (or \( J_2-M \)) phase diagram. When phase separation appears, it is accompanied by two first-order phase transitions in the magnetization (\( H-M \)) curve. These two first-order transition lines merge with a line of second-order phase transition at this phase boundary. In fact, following the discussion in Sec. IV D, if we assume that the sixth-order terms are continuous and non-vanishing in that neighborhood, the tricritical point is located at \( (J_3^*, M_s) \) [or \( (J_2^*, M_s) \)], where \( J_3^* \) (\( J_2^* \)) denotes the value at which the relevant quartic term (\( \Gamma^{(1)} \) for the spiral state, etc.) changes sign and \( M_s \) the saturated magnetization.

Lastly, let us stress that the model of the ground-state energy of the dilute magnon gas in Sec. III is quite general for spin exchange models enjoying a finite degeneracy of single-magnon energy minima. For example, it is straightforward to introduce XXZ-type spin anisotropy in our model (see Appendix A). We performed the analysis for several values of anisotropy, but did not find qualitative modification of the phase diagrams. While we have found only single-\( q \)- or double-\( q \)-states near saturation in our simple microscopic model, there is the possibility to find higher-\( q \) states with this method by starting from a more complex Hamiltonian. We expect, in some complex systems including frustrated interlayer couplings, that spin anisotropy can drive the transition to interesting multiple-\( q \) phases. In fact we note that this is essentially what happens in the spin model for \( \text{Ba}_3\text{Mn}_2\text{O}_8 \), which can accommodate various new phases including magnetic vortex crystals.

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Appendix A: Details on the solution of the BS equation

A common method of solving Eq. \( \text{(A1)} \) is to make an expansion in lattice harmonics. Here we briefly describe the method. First note that, integrating over the Brillouin zone, one has

\[
\langle \Gamma(q; k, k') \rangle = 2U \left( 1 - \frac{1}{N} \sum_{q' \in BZ} \frac{\Gamma(q'; k, k')}{\epsilon(k + q') + \epsilon(k' - q') - 2\epsilon_{\text{min}} - E} \right),
\]

where \( \langle \cdots \rangle = (1/N) \sum_{q' \in BZ} \cdots \) and we have used \( \langle \epsilon \rangle = 0 \). For \( U \to \infty \) we obtain

\[
1 - \frac{1}{N} \sum_{q' \in BZ} \frac{\Gamma(q'; k, k')}{\epsilon(k + q') + \epsilon(k' - q') - 2\epsilon_{\text{min}} - E} = 0.
\]

(A2)

Using Eq. (A1), Eq. (18) becomes

\[
\Gamma(q; k, k') = 2\epsilon(q) + \langle \Gamma(k, k') \rangle
\]

\[
- \frac{1}{N} \sum_{q' \in BZ} \frac{2\epsilon(q - q')}{\epsilon(k + q') + \epsilon(k' - q') - 2\epsilon_{\text{min}} - E} \Gamma(q'; k, k').
\]

(A3)

At this stage one would like to take a suitable Ansatz for \( \Gamma \) and transform the problem, namely Eqs. (A2) and (A3), to a linear algebraic system. The most general expansion of lattice harmonics would contain both harmonics of type sine and cosine. However, one can get rid of harmonics of the type sine by considering even functions of \( q \) to obtain the \( \Gamma \)-s, namely

\[
\hat{\Gamma}^{(1)}(q) = \Gamma(q; Q_0, Q_0),
\]

\[
\hat{\Gamma}^{(2)}_1(q) = \Gamma((Q_1 - Q_0)/2 + q; Q_0, Q_1) + \Gamma((Q_1 - Q_0)/2 - q; Q_0, Q_1),
\]

\[
\hat{\Gamma}^{(2)}_2(q) = \Gamma((Q_2 - Q_0)/2 + q; Q_0, Q_2) + \Gamma((Q_2 - Q_0)/2 - q; Q_0, Q_2),
\]

\[
\hat{\Gamma}^{(3)}(q) = \Gamma((Q_3 - Q_0)/2 + q; Q_0, Q_3) + \Gamma((Q_3 - Q_0)/2 - q; Q_0, Q_3). \quad (A4)
\]

Then the couplings in the ground-state energy \( \text{(H)} \) will be given by

\[
\Gamma^{(1)} = \hat{\Gamma}^{(1)}(0),
\]

\[
\Gamma^{(2)}_1 = \hat{\Gamma}^{(2)}_1((Q_1 - Q_0)/2),
\]

\[
\Gamma^{(2)}_2 = \hat{\Gamma}^{(2)}_2((Q_2 - Q_0)/2),
\]

\[
\Gamma^{(3)}_0 = \hat{\Gamma}^{(3)}((Q_3 - Q_0)/2),
\]

\[
\Gamma^{(3)}_1 = \hat{\Gamma}^{(3)}(Q_1 - (Q_3 + Q_0)/2),
\]

\[
\Gamma^{(3)}_2 = \hat{\Gamma}^{(3)}(Q_2 - (Q_3 + Q_0)/2). \quad (A5)
\]

It is possible to verify \( \Gamma^{(1)}_1 = \Gamma^{(2)}_1 \), as argued previously from symmetry considerations.

In order to solve Eq. (A4) we then take the Ansatz Eq. \( \text{(19)} \). For simplicity let us restrict ourselves to \( J_2 = 0 \) and define
\[ T(q) = \left(1, \cos a_0 \cdot q, \cos a_2 \cdot q, \cos a_4 \cdot q, \cos 2a_0 \cdot q, \cos 2a_2 \cdot q, \cos 2a_4 \cdot q, \cos q_2\right)^T, \]  

\[ \tau_{ij}(k, k'; E) = \frac{1}{N} \sum_{q' \in \mathcal{B}} \epsilon \left(\frac{k + k'}{2} + q'\right) + \epsilon \left(\frac{k + k'}{2} - q'\right) - \epsilon(k) - \epsilon(k') - E, \]  

For the 3D calculation in Sec. IV we can safely set E to its "on-shell" value E = 0. However, for the 2D calculation in Sec. IV.B (J_0 = 0), a small but finite E will be used to regularize the integrals in Eq. (A7), that typically suffer from logarithmic divergences, as explained in Sec. III.B.

Upon plugging Eq. (A9) into Eq. (A2) and Eq. (A3) with

\[ M = \begin{pmatrix} \tau_{11} & J_1 \tau_{12} & J_1 \tau_{13} & J_3 \tau_{14} & J_3 \tau_{15} & J_3 \tau_{16} & J_3 \tau_{17} & J_0 \tau_{18} \\ 2 \tau_{21} & 1 + 2J_1 \tau_{22} & 2J_1 \tau_{23} & 2J_3 \tau_{24} & 2J_3 \tau_{25} & 2J_3 \tau_{26} & 2J_3 \tau_{27} & 2J_3 \tau_{28} \\ 2 \tau_{31} & 2J_1 \tau_{32} & 1 + 2J_1 \tau_{33} & 2J_3 \tau_{34} & 2J_3 \tau_{35} & 2J_3 \tau_{36} & 2J_3 \tau_{37} & 2J_3 \tau_{38} \\ 2 \tau_{41} & 2J_1 \tau_{42} & 2J_1 \tau_{43} & 1 + 2J_1 \tau_{44} & 2J_3 \tau_{45} & 2J_3 \tau_{46} & 2J_3 \tau_{47} & 2J_3 \tau_{48} \\ 2 \tau_{51} & 2J_1 \tau_{52} & 2J_1 \tau_{53} & 2J_1 \tau_{54} & 1 + 2J_1 \tau_{55} & 2J_3 \tau_{56} & 2J_3 \tau_{57} & 2J_3 \tau_{58} \\ 2 \tau_{61} & 2J_1 \tau_{62} & 2J_1 \tau_{63} & 2J_1 \tau_{64} & 2J_3 \tau_{65} & 1 + 2J_1 \tau_{66} & 2J_3 \tau_{67} & 2J_3 \tau_{68} \\ 2 \tau_{71} & 2J_1 \tau_{72} & 2J_1 \tau_{73} & 2J_1 \tau_{74} & 2J_3 \tau_{75} & 2J_3 \tau_{76} & 1 + 2J_1 \tau_{77} & 2J_0 \tau_{78} \\ 2 \tau_{81} & 2J_1 \tau_{72} & 2J_1 \tau_{73} & 2J_1 \tau_{74} & 2J_3 \tau_{75} & 2J_3 \tau_{76} & 2J_3 \tau_{77} & 2J_0 \tau_{78} \end{pmatrix}, \]

\[ n = \begin{pmatrix} \left(1, 2, 2, 2, 2, 2, 2, 2\right)^T \\ 2, 4 \cos k' \cdot k, 4 \cos k' \cdot a_0, 4 \cos k' \cdot a_2, 4 \cos k' \cdot a_4, 4 \cos(k' - k) \cdot a_0, \ldots, 4 \cos(k' - k_z) \end{pmatrix}^T \]

for \( \hat{\Gamma}^{(1)} \), otherwise.

The matrix elements can be calculated numerically in three dimensions.

In two dimensions, we recognize that the integration in Eq. (A3) is dominated at small E by the neighborhoods of the solutions of \( \epsilon(k + q') + \epsilon(k' - q') = 0 \) (one, two or six solutions depending on the choice of \( k, k' \in \{Q_i\} \)). The quantities in Eq. (A5) therefore have the singular behavior

\[ \hat{\Gamma}^{(1)}(q) = \frac{\hat{\alpha}^{(1)}(q)}{\log |E|^2} + \mathcal{O}\left(\frac{1}{\log |E|^2}\right) \]  

(\( \text{and similarly for the others} \)) at small E. By plugging into Eq. (A3) and keeping only the leading terms we obtain a set of coupled algebraic equations whose solution is given in the form

\[ a^{(1)}(0) = \frac{8\pi}{\sqrt{3}} \sqrt{\text{det}(h^{(1)}/2)}, \]

\[ a_{1}^{(2)} = \frac{8\pi}{\sqrt{3}} \sqrt{\text{det}(h^{(2)}/2)}, \]

\[ a_0^{(3)} = \frac{8\pi}{\sqrt{3}} \sqrt{\text{det}(h^{(3)}/2)}, \]

\[ a_1^{(3)} = \frac{8\pi}{\sqrt{3}} \sqrt{\text{det}(h^{(3)}/2)} \]  

Here we have introduced the relevant Hessian matrices
\[ h^{(1)}_{ij} = \partial_k \partial_{k'} [\epsilon(Q_0 + k) + \epsilon(Q_0 - k)]_{k=0}, \]
\[ h^{(2)}_{1,ij} = \partial_k \partial_{k'} \left[ \epsilon \left( \frac{Q_0 + Q_1}{2} + k \right) + \epsilon \left( \frac{Q_0 + Q_1}{2} - k \right) \right]_{k=Q_1 - Q_0}, \]
\[ h^{(2)}_{3,ij} = \partial_k \partial_{k'} \left[ \epsilon \left( \frac{Q_0 + Q_2}{2} + k \right) + \epsilon \left( \frac{Q_0 + Q_2}{2} - k \right) \right]_{k=Q_2 - Q_0}, \]
\[ h^{(3)}_{ij} = \partial_k \partial_{k'} [2\epsilon(k)]_{k=Q_0} = h^{(1)}_{ij}. \]

It is easy to verify that \( \det h^{(1)} = \det h^{(3)} \) and \( \det h^{(2)} = \det h^{(2)} \). Equations (30) and (31) follow.

Note that in order to include a generic XXZ-type spin anisotropy, namely to change the various terms in the Hamiltonian Eq. (11) from \( J_\alpha S_i \cdot S_j \) to \( J_\alpha(S_i^x S_j^x + S_i^y S_j^y + \Delta_\alpha S_i^z S_j^z) \), one just need the replacement \( J_\alpha \rightarrow J_\alpha \Delta_\alpha \) in Eqs. (10) and (A9).

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