Dilute-Bose-Gas Approach to ground state phases of 3D quantum helimagnets under high magnetic field

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Abstract. We study high-field phase diagram and low-energy excitations of three-dimensional quantum helimagnets. Slightly below the saturation field, the emergence of magnetic order may be mathematically viewed as Bose-Einstein condensation (BEC) of magnons. The method of dilute Bose gas enables an unbiased quantitative analysis of quantum effects in three-dimensional helimagnets and thereby three phases are found: cone, coplanar fan and an attraction-dominant one. To investigate the last phase, we extend the usual BEC approach so that we can handle 2-magnon bound states. In the case of 2-magnon BEC, the transverse magnetization vanishes and long-range order occurs in the quadrupolar channel (spin-nematic phase). As an application, we map out the phase diagram of a 3D helimagnet which consists of frustrated $J_1$-$J_2$ chains coupled by an interchain interaction $J_3$. 

Magnetic frustration introduces several competing states which are energetically close to each other and thereby destabilizes simple ordered states. One way to compromise two or more competing orders is to assume a helical (spiral) spin structure[1]. In this letter, we discuss the high-field behavior of a spin-1/2 Heisenberg model with generic interactions: $H = \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + H \sum_j S_j^z$. For the simplest case with one magnetic ion per unit cell, one can easily find the classical ground state by minimizing the Fourier transform of the exchange interactions:

$$\epsilon(q) = \sum_j \frac{1}{2} J_{ij} \cos(q \cdot (r_i - r_j)) ,$$

where the summation is taken over all $j$-sites connected to the $i$-site by $J_{ij}$. When $\epsilon(q)$ takes its minima $\epsilon_{\text{min}}$ at $q = \pm Q$, helical order with the wave number $Q$ or $-Q$ appears ($\pm Q$ are not equivalent to each other). When the external magnetic field is applied in a direction perpendicular to the spiral plane, the spiral at $H=0$ is smoothly deformed into the so-called cone state (figure 1) as $H$ is increased and this persists until all spins eventually get polarized at the saturation field $H_c$ [2].

One of the simplest models which exhibit, at least in the classical limit, the helical order is a three-dimensionally coupled Heisenberg chains with nearest-neighbor- (NN) $J_1$ and next-nearest-neighbor (NNN) $J_2$ coupling. Rise of multiferroics revives study of helimagnetism and many compounds which contain these 1D-chains as subsystems have been reported (see, for instance, TABLE I. in reference [3]). For example, a helimagnetic material LiCuVO$_4$ may be viewed as coupled quantum $S = 1/2$ $J_1$-$J_2$ chains and exhibits helical spin order, which is expected from the classical theories, and ferroelectricity simultaneously under moderate magnetic field. When the field is very high, on the other hand, this compound shows modulated collinear
order, which contradicts with the aforementioned classical prediction, and this may suggest that quantum fluctuation plays an important role \[4\]. Therefore, it would be interesting to explore the possibility that quantum fluctuation replaces the classical cone state with other stable ones.

By using dilute-Bose-gas approach, Batyev and Braginskii \[5\] investigated magnetic structures near saturation (H=H\(_c\)) and concluded that a new coplanar fan phase appears if a certain condition for the bosonic interactions is satisfied. Our aim in this letter is to determine the stable spin configurations of a specific 3D spin-1/2 helimagnet in a fully quantum-mechanical manner.

General Formalism– By taking fully polarized state as the vacuum and treating spin-flips as hardcore bosons \(S_i^\pm = -1/2 + \beta_i^\pm \), \(S_i^z = \beta_i^z\), \(S_i^- = \beta_i\), we can rewrite the spin Hamiltonian as:

\[
H = \sum_q (\omega(q) - \mu) \beta_q^\dagger \beta_q + \frac{1}{2N} \sum_{q,k,k'} V_{q,k} \beta_{k+q}^\dagger \beta_{k-q}^\dagger \beta_k \beta_{k'},
\]

where \(U(-\infty)\) has been added to insure the hard-core constraint and the minimum of \(\epsilon(q)\) is taken \(\epsilon_{\text{min}} = \epsilon(\pm Q)\) for helimagnets. In what follows, we consider a cubic lattice (we reserve \((a,b,c)\) to label the three crystal axes) and assume that helical- and ferromagnetic/antiferromagnetic order occur along the c-axis and in the ab plane, respectively (i.e. \(Q = (0,0,Q)\) or \(Q = (\pi,\pi,Q)\)). On general grounds, we may expect that magnon BEC occurs when the external field is \(H < H_c\) (\(\mu > 0\)).

The thermal potential per site \(E/N\) of the dilute Bose gas is determined by the renormalized interaction (\(t\)-matrix) among the condensed bosons at \(q = \pm Q\) and the ground state densities \(\rho_{\pm Q}\) are obtained by minimizing \(E/N\). If we denote the renormalized interactions between the like bosons and that between the different ones respectively as \(\Gamma_1\) and \(\Gamma_2\), the energy density \(E/N\) is given by

\[
\frac{E}{N} = \frac{1}{2} \Gamma_1 (\rho_{Q}^2 + \rho_{-Q}^2) + \Gamma_2 \rho_Q \rho_{-Q} - \mu (\rho_{Q} + \rho_{-Q}),
\]

where \(\rho_Q = |\langle \beta_Q \rangle|^2/N\). Different phases appear according to the values of \(\Gamma_{1,2}\). When \(\Gamma_2 > \Gamma_1 > 0\), \(E/N\) is minimal for the choice \(\rho_Q = \rho/\Gamma_1\), \(\rho_{-Q} = 0\) (or vice versa) and \(E/N = -\mu^2/(2\Gamma_1)\). Then, the spin configuration is determined as:

\[
\langle \beta_i \rangle = \sqrt{\rho} \exp\{\pm i(Q R_i + \theta)\}, \quad \langle S_i^z \rangle = -\frac{1}{2} + \rho, \quad \langle S_i^z \rangle = \sqrt{\rho} \cos(Q R_i + \theta), \quad \langle S_i^z \rangle = \pm \sqrt{\rho} \sin(Q R_i + \theta).
\]
That is, the cone state (the left panel of figure 1), which exists already in the classical case [2],
is favored for $\Gamma > \Gamma_1$.

If $\Gamma_1 > \Gamma_2$ and $\Gamma \equiv \Gamma_1 + \Gamma_2 > 0$, on the other hand, the two modes condense simultaneously
and the ground state is determined as: $\rho Q = \rho_+ Q = \rho'_- = \rho'/\Gamma$, $\frac{E}{\rho} = -u^2/\Gamma$

$$
\langle \beta_1 \rangle = \sqrt{\rho} \left\{ e^{i(Q R_1 + \theta_1)} + e^{i(-Q R_1 + \theta_2)} \right\},
$$

$$
\langle S^z_1 \rangle = \frac{1}{2} + 4 \rho' \cos^2 (Q R_1 + \frac{\theta_1 - \theta_2}{2}) \quad \langle S^z_1 \rangle = 2 \sqrt{\rho} \cos (Q R_1 + \frac{\theta_1 - \theta_2}{2}) e^{i\frac{\theta_1 + \theta_2}{2}}.
$$

(6)

The existence of the two phases $\theta_1$ and $\theta_2$, which correspond respectively to the two condensates $\langle \beta Q \rangle$ and $\langle \beta - Q \rangle$, lead to two different low-energy excitations. Since $\langle S^y_1 \rangle / \langle S^x_1 \rangle = -\tan \frac{\theta_1 + \theta_2}{2}$, the spins assume a coplanar configuration (fan) shown in the right panel of figure 1. According to the standard theory [6], this phase does not exhibit ferroelectricity.

In these two Bose condensed phases, there exists two low-energy modes at $q = \pm Q$, and the
low-energy physics is described by the effective Lagrangian with $U(1)$ $\cap$ $U(1)$ symmetry; one comes from the axial (around the external field) symmetry and the other from an emergent translational symmetry. In the cone phase, only one of the two bosons condenses and there is one gapless Goldstone mode (the two $U(1)$s are no longer independent). Meanwhile, the fan phase breaks both symmetries and has two types of gapless Goldstone modes (for more detail, see reference [7]).

When $\Gamma_1 < 0$ or $\Gamma_1 + \Gamma_2 < 0$, low-energy bosons around $q = \pm Q$ attract each other. If the energy (4) is taken literally, one may expect, on general grounds, first order transitions to occur (provided that the cubic terms of $\rho$ are positive). However, eq.(4) is based on the assumption that magnon BEC occurs in the single-magnon channel and may not work when we expect magnon bound states stabilized by strong attraction. In fact, according to the standard Bethe-Salpeter method, divergingly large (renormalized) attraction implies the existence of stable 2-magnon bound states as a pole in the two-particle Green’s function corresponds to a bound state. Therefore, the simple single-magnon BECs will give way to those in multi-particle channel. To check the above scenario, we solve the 2-magnon scattering problem. We denote the ladder diagram as $\Gamma(\Delta, K; p, p')$ (see figure 2), where we take the total energy of initial state as $-2\mu - \Delta$. Then, on the fully saturated ground state, the exact scattering amplitude $M(\Delta, K; p, p') = \Gamma(\Delta, K; p, p') + \Gamma(\Delta, K; p, -p')$ is obtained by solving the following integral equation:

$$
M(\Delta, K; p, p') = V(p' - p) + V(-p' - p) - \frac{1}{2N} \sum_{p''} \frac{M(\Delta, K; p, p'') (V(p' - p'') + V(-p' - p''))}{\omega(k/2 - p''') + \Delta - i0^+}.
$$

(7)

Even in the presence of the boson condensate, as long as it is dilute, we can safely use this scattering amplitude. Hence, one obtains the renormalized interaction $\Gamma_1$ and $\Gamma_2$ as $\Gamma_1 = (1/2) M(0, 2Q; 0, 0)$, $\Gamma_2 = M(0, 0; Q, Q)$ [8]. Moreover, the pole of $M(\Delta, K; p, p')$ gives the stable bound state.

Coupled $J_1$-$J_2$ model-- Having established the formalism, we proceed to investigating a specific model--a frustrated spin-1/2 model on a simple cubic lattice whose Hamiltonian is given by

$$
H = \sum_{r, i=a, b} \{ J_1 S_r \cdot S_{r+e_i} + J_2 S_r \cdot S_{r+2e_i} + J_3 S_r \cdot S_{r+e_i} \} + H \sum_j S^z_j.
$$

(8)

In (8), the $J_1$-$J_2$ chains are running in the $c$-direction and $J_3$ controls the coupling among adjacent chains. The wave number $Q$ characterizing the condensate is given either by $Q = (0, 0, Q)$ ($J_3 < 0$) or by $Q = (\pi, \pi, Q)$ ($J_3 > 0$) where $Q = \arccos(-J_1/4 J_2)$. Hence the spiral
occurs in the c-direction. To determine the spin structure of our $J_1$-$J_2$-$J_3$ model, we solved eq. (7) by assuming the following form [5]: $M(\Delta, \mathbf{K}, \mathbf{p}, \mathbf{p}') = \langle M \rangle + A_1 \cos p'_a + A_2 \cos 2p'_c + A_3 \cos p'_a + A_4 \cos p'_b$, where $\langle M \rangle$ and $A_i$ are functions of $\Delta, \mathbf{K}$ and $\mathbf{p}$. After some numerical calculations, we obtained the phase diagram shown in figure 3. We show only the frustrated region $-4 \leq J_1/J_2 \leq 4$, where cone structure with incommensurate $\mathbf{Q}$ is expected classically.

**Figure 3.** Phase diagram slightly below saturation ($H \lesssim H_c$) mapped out in $(J_1, J_3)$-plane ($J_2(>0)$ is used to set the energy unit). (i) cone phase and (ii) coplanar fan phase are one-magnon condensed phases. In the phase-(iii), attractive interaction may imply instabilities toward other phases e.g. conventional ferromagnetic one or more exotic multipolar ones. The phase (iv) (nematic) is characterized by the condensation of the 2-magnon bound state and leads to the nematic order in the transverse direction. The region $|J_3|/J_1 \ll 1$ is omitted for a reason described in the text. Inset: The same phase diagram for the large negative interchain coupling ($-J_3 > 0.1$).

For $J_3 \to 0$, low-energy quantum fluctuation destabilizes $\Gamma$ and our approach cannot be extended to $J_3 = 0$ continuously ($\Gamma$ becomes $O(J_3^2)$ and $\Gamma_1 \to \Gamma_2$ at the leading order in $J_3$).

Finally, we compare the phase diagram shown in figure 3 with that of the 1D $J_1$-$J_2$ chain ($J_3 = 0$). Let us begin with the case $J_1 > 0$. Near saturation, two dominant phases are found in 1D [9]: (i) ‘chiral phase (VC)’ with finite vector chirality parallel to the magnetic field and (ii) ‘TL2’ phase where the system is described by two Tomonaga-Luttinger (TL) liquids. Obviously, the former turns, after switching on $J_3$, into the cone phase. A close inspection of the two gapless TL modes near saturation tells us that the TL2 phase should be identified with the fan phase here.

For the ferromagnetic case $J_1 < 0$, BECs of $n$-bound magnon states ($n \geq 2$) are expected in 1D chain [10]. We found how strong inter-chain coupling destabilizes the 2-bound magnon BEC. The study of the stability of higher $n$-bound magnon state ($n \geq 3$) in 3D is a future problem.

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