Nematic phase and phase separation near saturation field in frustrated ferromagnets

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We argue the effect of quantum fluctuations on the magnetization process of the quantum frustrated ferromagnets. It is found that on general grounds in the neighborhood of the ferromagnet/antiferromagnet classical 1st-order phase boundary in zero external field, the phase separation or the non-classical phase must appear slightly below the saturation field in the quantum case, if the classical AF is not the eigenstate. Then, we study the ferromagnetic $J_1$-$J_2$ $S=1/2$ Heisenberg model ($J_1 < 0$) on the bcc lattice from the viewpoint of the magnon Bose-Einstein condensation. For $-1.50097 \leq J_1/J_2 \leq -1.389$, the nematic phase is expected and for $-1.389 < J_1/J_2 \leq -0.48$ the phase separation appears under high magnetic field.

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Quantum fluctuations (QF) in frustrated magnets can introduce a drastic change in the ground state from the classical system and induce various exotic phases. As one of quantum phases, the spin nematic has been recently paid much attention, which is nonmagnetic but exhibits the long-range order of the multiple spins.

In this paper we discuss the effect of QF on frustrated magnets which have dominant ferromagnetic (FM) couplings, studying the possibility of the spin nematic phase. We consider the $S = 1/2$ frustrated Heisenberg model described by

$$H = \sum_{i,j} J_{ij} S_i \cdot S_j - H \sum_i S_i^z. \quad (1)$$

The classical limit ($S \to \infty$) is equivalent to considering the spin operator as the vector of the fixed length $S$. In the case with one magnetic ion per unit cell, the classical ground state is obtained by minimizing the Fourier transform of the exchange interactions

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

The $J_1$-$J_2$ model, described by the nearest-neighbor coupling $J_1$ and next-nearest-neighbor coupling $J_2$, has been extensively studied in the research of frustration effects. In the one-dimensional (1D) $J_1$-$J_2$ chain with FM $J_1 < 0$, the spiral spin structure is classically stable for $|J_1/J_2| < 4$ and, is replaced by the spin nematic phase in $S = 1/2$ system in high magnetic field. In LiCuVO$_4$, the magnetic properties of which is understood as the quasi-1D $J_1$-$J_2$ model with the weak interchain couplings, a non-trivial phase is recently found slightly below the saturation field and it is expected that the spin nematic is observed for the first time.

The $J_1$-$J_2$ model on the square lattice is another system which exhibits the nematic phase. Classically, three types of orders are stabilized according to $J_{1,2}$: Néel antiferromagnetic (NAF), collinear antiferromagnetic (CAF), and FM phases. In the quantum case, the nematic phase was proposed from both the numerical and analytical approaches near the classical FM/CAF phase boundary ($J_1/J_2 = -2$ and $J_1 < 0$), though there is still debate about the stability of spin nematic order in the case of zero field. Slightly below the saturation field, the spin nematic phase is firmly induced by the two-magnon instability in the saturated FM state for the broad parameter range $-2.5 \leq J_1/J_2 \leq -0.225$. The recent experimental advance found the various compounds, e.g., BaCdVO(PO$_4$)$_2$, which are adequate to consider as the FM $J_1$-$J_2$ model. Some of them lie in the parameter range where the nematic phase is theoretically proposed. It would be useful to discuss how QF changes magnetic properties near the boundary between FM and AF phases in general frustrated FM systems.

In this paper, we first argue the quantum effects near the FM/AF phase boundary on general grounds. In the neighborhood of the FM/AF-'classical' phase boundary in zero field, there necessarily exists either a non-classical phase or a phase separation accompanied with magnetization jump below the saturation field, if the FM/AF transition is first order and the classical AF state is not the exact ground state of the Hamiltonian. As the FM/CAF phase boundary in the $S = 1/2$ square-lattice $J_1$-$J_2$ model satisfies this condition, this perspective reinforces the former proposal of the quantum spin nematic phase. As another example of this argument, we further study the three-dimensional $J_1$-$J_2$ model on the bcc lattice, which has been extensively studied as one of the simplest frustrated magnets. Recent theoretical studies concluded that in the AF case ($J_{1,2} > 0$) the classically expected AF orders persist even in the quantum case. For the FM $J_1$ case, however, the above discussion asserts the appearance of non-classical behavior. In the later half of this paper, we study the appearance of spin nematic phase or the phase separation in high field in the $S = 1/2$ $J_1$-$J_2$ model with FM $J_1$ on the bcc lattice. We employ the dilute Bose gas approach from the viewpoint of the magnon Bose-Einstein condensation (BEC).

Let us start with a general argument on the quantum effect on the phase boundary between FM and AF phases. In the absence of magnetic field, the classical ground state usually shows a first-order phase transi-
ation between FM and AF phases. In most settings of the quantum systems, the classical AF state $|\text{AF;CL}\rangle$ is not an eigenstate of the Heisenberg Hamiltonian, $H|\text{AF;CL}\rangle = E_{\text{CL}}|\text{AF;CL}\rangle + |\alpha\rangle$, where $\langle\text{AF;CL}|\alpha\rangle = 0$ and $\langle\alpha|\alpha\rangle > 0$, albeit the FM state (FM;CL) is the eigenstate having the same energy as the classical one. The variational principle guarantees that the true quantum (qAF) ground state in the classical AF regime has the lower energy than that of the FM and AF Néel states at the classical FM/AF phase boundary. Therefore, FM/qAF phase boundary shifts from the classical one toward the FM phase by a certain amount in zero field.

The magnetization process in applied field must be compatible with this fact. However, if one considers the saturation field given by one-magnon flips, which is always the same as the classical value $H_c$, the saturation field must vanish at the classical FM/AF phase boundary. This concludes that the true saturation field is not given by single magnon instability. The AF phase described with the single magnon BEC below the saturation field is veiled by qAF near the FM/AF phase boundary. We thus conclude the appearance of a non-classical phase or the phase separation below the saturation field. An appealing alternative to the single magnon BEC in applied field is the BEC of multiple-bound magnons.

Hence, as in the square-lattice $J_1$-$J_2$ model, the QF plays a considerable role in the magnetization process in $J_1$-$J_2$ models on other lattices, even in three dimensions where the QF is believed to be weak. The $J_1$-$J_2$ model on the bcc lattice satisfies the condition discussed above. We hereafter discuss the possibility of the nematic phase and the phase separation in the magnetization process on the bcc lattice.

Before studying the quantum effect, let us briefly review the ground state in the classical case. When the external field $H$ is absent, the three types of phases appear; NAF phase for $J_1/J_2 > 3/2$ and $J_1 > 0$, CAF phase for $-3/2 < J_1/J_2 < 3/2$ and $J_2 > 0$, and FM phase for $J_1/J_2 < -3/2$ and $J_1 < 0$, as shown in Fig. 2.

In CAF phase, the lattice is divided into two sublattices, and the spins form the NAF on each sublattice, where the spin angle between two sublattices is independent and QF fixes it. When the external field is applied in the CAF phase, the spins gradually point upward from the plane perpendicular to the external field. The angle between spins on two sublattices is independent even in the magnetization process. Within the linear spin wave theory, the (deformed) CAF phase is stabilized.

We can apply the argument discussed above to this model. At the FM/CAF phase boundary $J_1/J_2 = -3/2$, the classical CAF state is not an eigenstate in the quantum Heisenberg model. Hence, near $J_1/J_2 = -3/2$, a nontrivial quantum phase or the phase separation must appear under high magnetic field. On the other hand, at the FM/NAF phase boundary $J_2 = 0$, the NAF state is an eigenstate even in the quantum case and then a quantum fluctuation is not important. We hence focus our attention on the CAF and FM phase boundary. In particular, we study the magnetic behavior slightly below the saturation field in a fully quantum manner using the method of a dilute Bose gas.

The emergent magnetic order below the saturation field can be viewed as a condensation of the magnons, which successfully explains various experimental results. In the hardcore boson language, the spin operators at site $l$ are written as $S_l^z = -1/2 + a_l^\dagger a_l$, $S_l^+ = a_l^\dagger$, and $S_l^- = a_l$, where $a_l$ represents the annihilation operator of a hardcore boson. The vacuum $|\Omega\rangle$ corresponding to the saturated FM state is defined by $a_l|\Omega\rangle = 0$. The Hamiltonian reads

$$H = \sum_q (\omega(q) - \mu) a_q^\dagger a_q + \frac{1}{2N} \sum_{q,k,k'} V_{q,k,k'} a_{q,k}^\dagger a_{q,k'}^\dagger a_{q,k'} a_{q,k}$$

with $\omega(q) = \epsilon(q) - \epsilon_{\text{min}}$, $\mu = H_c - H$, $H_c = \epsilon(0) - \epsilon_{\text{min}}$, and $V_q = 2(\epsilon(q) + U)$, where $\epsilon(q)$ is given in Eq. (2), $\epsilon_{\text{min}}$ is the minimum of $\epsilon(q = Q)$, and $U(-\infty)$ is the hard-core potential. We see that the BEC occurs when
the external field is $H < H_c \ (\mu > 0)$, which leads to $\langle S^z_i \rangle = \langle a_i \rangle \neq 0$. The induced spin-ordered phase possesses characteristic wave vector $Q$. To clearly specify the characteristics of BEC, we call the BEC induced by the magnons with the magnetic quantum number $+1$ as the 'single' magnon BEC. In large spin $S$ systems, where the theoretical method is extended by using the spin wave expansion\textsuperscript{10} the classical phase near the saturation field can be well described with the single magnon BEC. In the quantum case, the single magnon BEC can induce a different spin-ordered phase from the classical one, albeit the saturation field of the single magnon BEC is exactly the same as the classical one.

Since the method is conventional and detailed in Refs.\textsuperscript{10,13,14} we mainly discuss our results on the bcc lattice. The dispersion relation is given by $\epsilon(q) = 4J_1 \cos(q_x/2) \cos(q_y/2) \cos(q_z/2) + J_2(\cos q_x + \cos q_y + \cos q_z)$ and takes minimaums at $\pm Q$ where $Q = (\pi, \pi, \pi)$ for $J_1/J_2 \geq -3/2$. The thermal potential per site $E/N$ of the dilute Bose gas is determined by the interaction among the bosons condensed at $q = \pm Q$. Then, the densities of the condensed bosons $\rho_{\pm Q}$ in the ground state are obtained by minimizing $E/N$. If we introduce the renormalizations interactions between the same bosons and that between different ones respectively as $\Gamma_1$ and $\Gamma_2$, the energy density $E/N$ in the dilute limit is given by

$$E/N = \frac{\Gamma_1}{2} (\rho_Q^2 + p_{-Q}^2) + \{G_2 + \Gamma_3 \cos 2(\theta_Q - \theta_{-Q})\} \rho_Q \rho_{-Q} - \mu (\rho_Q + \rho_{-Q}),$$

where $\langle a_i \rangle = \sqrt{N} \rho_Q \exp(i\theta_q)$ and the term with $\Gamma_3$ appears from the lattice structure. If $\Gamma_3$ is positive (negative), the phases $\theta_{\pm Q}$ are pinned as $\theta_Q = -\theta_{-Q} = m\pi \ (m = 1, 2, 3 \ldots)$ with an integer $m$. $\Gamma_\mu$ are calculated from the spin Hamiltonian later.

Various ground states can appear according to the values of $\Gamma_\mu$. If $\Gamma_2 - |\Gamma_3| > \Gamma_1 > 0$, the ground state is given by $\rho_Q = \mu/\Gamma_1$ and $\rho_{-Q} = 0$ (or vice versa), breaking the chiral symmetry. This phase is, however, not found in the present $J_1$-$J_2$ model in the other hand. If $\Gamma_1 > \Gamma_2 - |\Gamma_3| > -\Gamma_1$, on the other hand, the two modes condense simultaneously with the same density and the ground state is determined as $\rho_Q = \rho_{-Q} = \rho = \mu/(\Gamma_1 + \Gamma_2 - |\Gamma_3|)$ and $\langle a_i \rangle = \sqrt{\rho} [\exp(iQ.R_i + \theta_Q) + \exp(-iQ.R_i + \theta_{-Q})]$. In this phase, the spin expectation values are given as

$$\langle S^z_i \rangle = -\frac{\Gamma_1}{2} + 4\rho \cos^2(\theta_Q - \theta_{-Q}),$$

$$\langle S^\pm_i \rangle = 2\sqrt{\rho} \cos(\theta_Q - \theta_{-Q}) \mu^2 \frac{\Omega_{+a_{\pm}a_{\mp}}}{\mu}$$. \textsuperscript{(5)}

For positive $\Gamma_3$, the CAF appears, as expected within the linear spin wave theory. Meanwhile, for negative $\Gamma_3$, the emergent phase has a modulation of the density $\langle S^z_i \rangle$, which can be considered as the spin-supersolid phase.

If $\Gamma_1 < 0$ or $\Gamma_1 + \Gamma_2 - |\Gamma_3| < 0$, the low-energy bosons around $q = \pm Q$ attract each other and we hence expect a first order transition, or equivalently, phase separation. So far, our argument based on Eq. (4) is restricted to the magnon BEC that occurs in the single-particle channel. However, strong attraction can also induce formation of magnon bound states, which lead to the BEC of magnon bound states.

The condensed phase of the two-magnon bound states has the different properties from the single-magnon BEC. The striking one is the absence of the transverse local magnetization, and instead the existence of the low-range order in the quadratic channels $\langle S^z_i S^z_j \rangle \neq 0$ for a certain bond $(i, j)$, which corresponds to the spin nematic order.\textsuperscript{12} The stability of the two magnon bound state is implied from the divergence of the scattering amplitude of the two magnons.

To study $\Gamma_\mu$ concretely, we study the ladder diagram shown in Fig. 4 which describes the exact scattering amplitude $M$ on the saturated FM phase:

$$M(\Delta, K; p, p') = V_{p'-p} + V_{-p'-p} - \frac{1}{2} \int \frac{d^3 p''}{(2\pi)^3} \frac{M(\Delta, K; p, p'')}{(\omega(K/2 + p'') + \omega(K/2 - p'')+ \Delta - \mu)^+},$$

where $\Delta$ is the total energy and $K$ is the center-of-mass momentum. $\Gamma_\mu$ are given by $\Gamma_1 = M(0, 2Q; 0, 0)/2$, $\Gamma_2 = M(0, 0; Q, Q)$, $\Gamma_3 = M(0, 2Q; 0, q_i)/2$ with $q_i = (0, 0, 2\pi)$. The dispersion relation of the bound state $\Delta(K)$ is identified as the divergence of $M$ by scanning it with respect to $\Delta$ and $K$. By using the harmonic expansion, Eq. (6) is exactly solvable.\textsuperscript{10,13,14}

First, we discuss the result of $\Gamma_\mu$ obtained for $\Delta = 0$ to study the single magnon BEC. For $-0.48 \leq J_1/2 \leq 3/2$, the CAF appears since $\Gamma_1 > \Gamma_2 - |\Gamma_3| > -\Gamma_1$ and $\Gamma_3 > 0$ hold. For $-1.389 \leq J_1/2 \leq -0.48$, we obtain $\Gamma_1 < 0$ and the phase separation near the saturation field is expected. The resulting phase below the magnetization jump may be CAF, but we do not exclude the possibility of nontrivial phases such as the spin nematic appearing by the 1st-order transition. At $J_1/2 = -1.389$, both $\Gamma_1$ and $\Gamma_3$ diverge, implying the appearance of bound states. For $-3/2 \leq J_1/2 \leq -1.389$, we have $\Gamma_1 > \Gamma_2 - |\Gamma_3| > -\Gamma_1$ and $\Gamma_3 < 0$, which suggests the spin supersolid.

To see the possibility of the bound magnon BEC, we detail the binding energy of the bound state. The minimum of the dispersion $\Delta(K)$ is always obtained at

![FIG. 3: Scattering amplitude $M$ given by the ladder diagram.](image)
pairs closes earlier than that of the single magnons and thus spin nematic ordering veils the expected spin super-solid phase. For $-1.50097 \leq J_1/J_2 \leq -3/2$, the ground state is not the classically expected FM phase in the absence of the external field since the reference state (FM) is destabilized by the fluctuation of the bound-magnon pairs. Our finding shows that the nematic phase is the serious candidate of the ground state. From the symmetry perspective the FM/nematic phase transition in zero field may be 1st-order as in the case of the square lattice.

We note that there remains the possibility of appearance of CAF phase in very low field regime accompanied with the magnetization jump below the saturation field since the energy of the bound state is almost the same as the single magnon.

Finally, we discuss the properties of the bound magnon condensed phase. The wave function of the bound-magnon pairs with respect to the relative coordinate is given by

$$
\chi(p) \propto \frac{\cos \frac{p_z}{2} \cos \frac{p_y}{2} \cos \frac{p_x}{2}}{\omega(K_0/2 + p) + \omega(K_0/2 - p) - \Delta},
$$

(8)

where we have omitted the normalization constant. The nematic order parameter, which is dominant on the nearest neighbor bonds, has the $f_{xyz}$-wave symmetry, i.e., \( \langle S^+_i S^+_{i+\epsilon} \rangle = -\langle S^+_i S^+_{i+\epsilon} \rangle \rangle = \langle S^+_i S^+_{i+\epsilon} \rangle \rangle = \langle S^+_i S^+_{i+\epsilon} \rangle \rangle \), with the wave vector \( K_0 \).

In conclusion, we have discussed the effect of QF on the quantum frustrated ferromagnets under high magnetic field. In general, near the FM/AF classical 1st-order phase boundary, the non-classical phase or the phase separation must occur in magnetization process, if the classical AF is not an eigenstate. We stress that the phase separation occurs even in the isotropic Heisenberg model and is purely the quantum phenomena. As a concrete model satisfying the above condition, we study the \( J_1-J_2 \) model on the bcc lattice using the method of a dilute Bose gas. We found that for \(-1.389 \leq J_1/J_2 \leq -0.48\), the single magnon condensed phase is not stable near the saturation field and the phase separation occurs. For \(-1.50097 \leq J_1/J_2 \leq -1.389\), the bound magnon BEC which leads to the nematic phase is expected.

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In Ref. 4, although discussed, the region of the nematic phase is not declared explicitly. And for $-0.2 \lesssim J_1/J_2 \leq 0$, we do not comment on the possibility of the nematic phase because of the problem of the numerical precision in our calculations.