Magnetic phase diagram slightly below the saturation field in the stacked $J_1$-$J_2$ model in the square lattice with the $J_3$ interlayer coupling

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We study the effect of the $J_3$ interlayer coupling on the $J_1$-$J_2$ Heisenberg model on the square lattice under high external field. Concretely, we consider the cubic lattice where the $J_1$-$J_2$ square lattices are stacked with the interlayer $J_3$ exchange coupling. It is theoretically known that, in the pure-2-dimensional model ($J_3 = 0$), for $-0.4 \leq J_2/J_1$ and $J_2 > 0$, the spin nematic phase may appear under high external field. By using the dilute-bose-gas and the Bethe-Salpeter techniques, we obtain the magnetic-phase diagram in the fully-3-dimensional model slightly below the saturation field. We find that, for $J_2/J_1 \sim -0.5$, the spin nematic phase is still expected even for the comparable-interlayer coupling $|J_3/J_1| \sim 1$. By increasing $|J_3|$, nearby the nematic phase in the phase diagram, the broad phase-separation region appears. By further increasing $|J_3|$, the semiclassically-expected collinear antiferromagnetic phase appears respecting the order-by-disorder mechanism.

KEYWORDS: frustration, spin nematic

Introduction: Frustration and quantum fluctuations introduce us various exotic phases in magnets. A spin-nematic phase is one of the appealing magnetic phases, where not the magnetization but the rank-2 tensor of the product of the spin operators characterizes the long-range order.\(^1,2\) Theoretically, the realization of the spin nematic phase is expected in various frustrated-Heisenberg models, e.g., the frustrated spin-$1/2$ $J_1 - J_2$ model on the square lattice:

$$H_{2d} = \sum_{\text{n.n.}} J_1 \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\text{n.n.n.}} J_2 \mathbf{S}_i \cdot \mathbf{S}_j + H \sum_i S^z_i,$$

where ‘n.n. (n.n.n.)’ implies (next) nearest neighbor couplings in the a-b plane, and H is applied external magnetic field. Frustration may be strong near the classical phase boundary at $J_2/J_1 = -0.5$ between the ferromagnetic phase (FM) and the collinear anti-ferromagnetic phase (CAF), as shown in Fig. 1. At zero field (H = 0), near this classical CAF/FM phase boundary, the spin nematic phase is expected, although being still under debate.\(^3\)-\(^8\) Under high external field, the stable nematic phase is found by both the exact diagonalization and the analytic approach solving the bound-magnon energy on the saturated phase.\(^3,9\) Especially, the analytic-binding-energy approach suggests the spin-nematic phase for $0.4 \leq J_2/J_1$ and $J_1 < 0$ slightly below the saturation field;\(^3,10\) on the fully polarized phase under high magnetic field, the energy of the bound magnon can be calculated exactly.\(^12,13\) If the gap of the bound magnon closes earlier than the single magnon (spinwave) during the decrement of the external field, the spin-nematic phase may appear.

There are several compounds which may be viewed as the spin-1/2 2D $J_1$-$J_2$ square-lattice model;\(^14\)-\(^17\) for example, BaCdVO(PO$_4$)$_2$, SrZnVO(PO$_4$)$_2$, Pb$_2$VO(PO$_4$)$_2$, and BaZnVO(PO$_4$)$_2$ are considered as the ferro $J_1 < 0$ compounds,\(^14\) and their estimated exchange couplings depicted in Fig. 1 suggest the nematic phase under high magnetic field. Recently, several techniques how to measure the nematic phase are proposed,\(^18,19\) and the detection of the nematic phase may not be a distant idea.

In any real compound, there is more or less a finite interlayer coupling. Naively, the interlayer coupling may tend to destabilize a nontrivial quantum phase. For example, BaCdVO(PO$_4$)$_2$, SrZnVO(PO$_4$)$_2$, Pb$_2$VO(PO$_4$)$_2$ and BaZnVO(PO$_4$)$_2$ may have finite interlayer couplings, and it
may be important to study the effect of the interlayer coupling on the nematic phase. Although there are several theoretical studies of the effect of the interlayer coupling on the classically-expected CAF and Néel antiferromagnetic phase (NAF) on the 2D $J_1$-$J_2$ square-lattice model,9,20-22) none is known for the nematic phase within our knowledge. On the other hand, as for the nematic phase in 1D $J_1$-$J_2$ chain, the effect of the interchain coupling has been studied by many groups.23-30)

In this letter, we study the effect of the interlayer coupling on the 2D $J_1$-$J_2$ model near the CAF/FM phase boundary under high magnetic field, fully including quantum fluctuations. As one of the simplest-test models including interlayer couplings, we consider the stacked 2D $J_1$-$J_2$ model with the $J_3$ interlayer coupling on the cubic lattice (see Fig. 3). We shall complete the phase diagram of this model (Figs. 5,6) slightly below the saturation by using the dilute-Bose-gas and the Bethe-Salpeter (bound-magnon) methods.11,24,31,32) As a result, we find the robustness of the spin nematic phase against the interlayer coupling on the CAF/FM phase boundary for $J_2/J_1 \sim -0.5$: the spin nematic phase can appear even if $|J_3/J_1| \sim 1$. The larger $|J_3|$ eventually destabilizes the nematic phase, and then, there appears the non-classical broad-parameter region where the phase separation at the saturation field occurs. By further increasing $|J_3|$, the semiclassically expected canted-CAF phase appears.

**Fig. 3.** (Color online) Three-dimensional stacked-square (cubic) lattice. The filled circles denote spins connected by Heisenberg exchange interactions. In the square lattice in the a-b plane, the (next) nearest neighbor $J_1$ ($J_2$) coupling is assumed. The interlayer coupling is given as $J_3$. The lattice constant $a_0$ is assumed to be 1.

Hamiltonian- We study the stacked $J_1$-$J_2$ Heisenberg model in the square lattice with the interlayer coupling $J_3$ (totally cubic lattice, see Fig. 3):

$$H = \sum_{\text{n.n.n in a-b}} J_1 S_i \cdot S_j + \sum_{\text{n.n.n in a-b}} J_2 S_i \cdot S_k + \sum_1 J_3 S_i \cdot S_{j+\epsilon},$$

$$+ H \sum_1 S_i^z,$$  (2)

where ‘n.n. (n.n.n) in a-b’ implies (next) nearest neighbor couplings in the a-b plane.

For convenience, we use the hardcore-boson representation:

$$S_i^+ = -1/2 + a_i^\dagger a_i, \quad S_i^- = a_i^\dagger, \quad S_i^z = a_i.$$  (3)

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

$$\omega(q) = \epsilon(q) - \mu \quad \text{and} \quad H_c = \epsilon(0) - \mu,$$

where $\epsilon(q) = 2(e(q) + U)$, $V_q = 2(\epsilon(q) + U)$, where the on-site interaction $U \to \infty$ and,

$$\epsilon(q) = J_1 (\cos k_x + \cos k_y) + J_2 (\cos (k_a + k_b) + \cos (k_a - k_b)) + J_3 \cos k_c.$$

$\epsilon_{\min}$ is the minimum of $\epsilon(q)$:

(i) For $-2 \leq J_1/J_2 \leq 2$ and $J_2 > 0$: $\epsilon_{\min} = \epsilon(Q_f^{(a)}) = -2J_2 - |J_3|$, where the labels $f$ and $a$ are respectively chosen for $Q_f^{(f)} = (\pi, 0, 0)$ and $Q_f^{(a)} = (0, \pi, 0)$.

(ii) For $J_1/J_2 \leq -2$ and $J_2 > 0$: $\epsilon_{\min} = \epsilon(Q_f^{(a)}) = 2J_1 + 2J_2 - |J_3|$, where $Q_f^{(f)} = (0, 0, 0)$ and $Q_f^{(a)} = (0, 0, \pi)$.

$H_c$ is the saturation field. If the external field lowers below $H_c$, the gap of magnon closes ($\mu > 0$), and the magnon-Bose-Einstein condensation may occur.

GL analysis- We focus on the case for $-2 \leq J_1/J_2 \leq 2$ and $J_{3,3} > 0$ since the same argument is easily applied to the $J_3 < 0$ case. Slightly below the saturation field for $\mu > 0$, Bose-Einstein condensation of magnons may occur in two momenta:

$$\langle a_{Q_f}^\dagger a_{Q_f} \rangle = \sqrt{N\rho_{Q_f}} \exp(i\theta_{Q_f}),$$

$$\langle a_{Q_f}^\dagger a_{Q_f} \rangle = \sqrt{N\rho_{Q_f}} \exp(i\theta_{Q_f}).$$  (6)

The induced spin-ordered phase is characterized by the wave vectors $Q^{(f)}$ and/or $Q^{(a)}$.

In the dilute limit, the energy density $E/N$ is expanded in the density $\rho_{Q_f}$ up to quadratic terms:

$$E/N = \frac{1}{2} \left( \rho_{Q_f}^2 + \rho_{Q_f}^2 \right) + \left( \Gamma_1 + \Gamma_3 \cos 2(\theta_Q - \theta_{Q_f}) \right) \rho_{Q_f} \rho_{Q_f}$$

$$- \mu (\rho_{Q_f}^2 + \rho_{Q_f}^2).$$

(8)

Here we introduced the renormalized interaction $\Gamma_1$ between the same bosons, $\Gamma_2$ between the different ones, and $\Gamma_3$ obtained from an umklapp scattering.

$\Gamma_{1,2,3}$ are concretely given by considering the scattering amplitude shown in Fig. 4:

$$\Gamma(\Delta, K; p, p') = V(p' - p) + V(-p' - p)$$

$$- \frac{1}{2} \int \frac{d^3 p''}{(2\pi)^3} \Gamma(\Delta, K; p, p'') [V(p' - p'') + V(-p' - p'')] \omega(K/2 + p'') + \Delta - i0^+.$$  (9)

where the integral is taken for the region $p''_{x,y,z} \in (0, 2\pi)$. $K$ and $\Delta$ respectively are the center-of-mass momentum of the two magnons and the binding energy. This integral equation is exactly solved.11,24,31,32) As a result, we obtain

$$\Gamma(\Delta, K; p, p') = \left[ \begin{array}{c} \frac{K}{2} + p \\frac{K}{2} + p' \\frac{K}{2} + p' \end{array} \right]$$

Fig. 4. (Color online) Scattering amplitude $\Gamma$ given by the ladder diagram.
Phase separation

When the magnon of the wavevector $q$ induced by the order-by-disorder mechanism is given by

$$\langle a \rangle = \sqrt{\rho} \exp[i(Q^{(a)}_l R_l + \theta_{Q^{(a)}})]$$

the spin-expectation values are explicitly described as

$$\langle S_i^z \rangle = -\frac{1}{2} + \rho$$

$$\langle S_i^1 \rangle = \sqrt{\rho} \cos(Q^{(a)}_l R_l + \theta_{Q^{(a)}})$$

$$\langle S_i^2 \rangle = -\sqrt{\rho} \sin(Q^{(a)}_l R_l + \theta_{Q^{(a)}})$$

This phase is the canted-CAF one, which may be expected within the large-$S$ linear spinwave theory respecting the order-by-disorder mechanism. If $\Gamma_1 > \Gamma_2 - |\Gamma_3|$, $\Gamma_1 > 0$ and $\Gamma_1 + \Gamma_2 - |\Gamma_3| > 0$, $\rho_{Q^{(a)}} = \rho_{Q^{(a)*}} = \rho' = |\mu|^2$. In this case, we expect the nontrivial multiple-$Q$ (double-$Q$) phase, which is observed in other several models. However, we abbreviate the detailed characters since the concrete calculation did not imply the existence of this phase in our model. When $\Gamma_1 < 0$ or $\Gamma_1 + \Gamma_2 - |\Gamma_3| < 0$, the dilutely-condensed phase is unstable, and the jump of the magnetization curve (phase separation) at $\mu < 0$ is expected. This is because, for example, if $\Gamma_1 < 0$, $E/N$ of eq. (8) goes to $-\infty$ if the density of magnon $\rho_{Q^{(a)}} \rightarrow \infty$; this divergence is due to the lack of higher-order interaction terms.

Bound magnon- We have discussed the magnetic phases induced by the single magnon slightly below the saturation field. Besides, there is the other possibility that magnons form the stable-bound state, and the gap of the bound magnon closes earlier than that of the single magnon by reducing the external field. Then, the bound magnon condenses, accompanied by not the magnetization but the spin-nematic order parameter. This phase is semiclassically expected. Especially for $0 < J_1/J_2 < 2$ and $J_3 > 0$, there is only the canted-CAF phase (no nematic, phase separation, double-$Q$ phase) slightly below the saturation field, even near $J_1/J_2 \sim 2$ where frustration is considered very large.

**Conclusion-** We have studied the effect of the interlayer coupling $J_3$ on the magnetic phases in the $S = 1/2$ stacked-square-lattice $J_1$-$J_2$ model under high external field by using the dilute Bose-gas technique. The main result of this letter is the phase diagram slightly below the saturation field shown in Figs. 5, 6. In contrast to the (semi-)classical case only with the canted-CAF phase, in addition to the CAF, there appear the spin-nematic phase and the phase separation. The spin nematic phase, the existence of which has been already naively use the $\Gamma_{1,2,3}$ neglecting effects of the finite density, $\Gamma_1 < \Gamma_2 - |\Gamma_3|$ may suggest the canted-CAF phase after the jump. However, we cannot exclude the possibility of the nematic phase or the double-$Q$ phase due to effects of finite density. On the boundary of the (i) nematic and the (ii) phase separation, the s-wave scattering amplitude $\Gamma_1$ diverges, and the Efimov effect is expected on and near this boundary. In the (iii) red region, the single magnon condenses so that the canted-CAF phase appears. We didn't find another phase for $-2 < J_1/J_2 < 2$ and $J_3 > 0$, where the canted-CAF phase is semiclassically expected. Especially for $0 < J_1/J_2 < 2$ and $J_3 > 0$, there is only the canted-CAF phase (no nematic, phase separation, double-$Q$ phase) slightly below the saturation field.
known in the pure 2D model ($J_3 = 0$),\(^{39}\) is quantitatively robust against the interlayer coupling near the FM/CAF phase boundary for $J_2/J_1 \sim -0.5$. For larger $|J_3|$, the broad phase-separation region appears. The phase separation is the magnetization jump (the first-order-phase transition) at the saturation field, and, after the jump, the canted-CAF may appear, although the possibility of the spin nematic or the double-Q phase still remains. On the boundary between the nematic phase and the phase separation, the s-wave scattering amplitude $\Gamma_1$ diverges and the Efimov effect is expected.\(^{43}\) In conclusion, this phase diagram suggests that, in a quasi-2D $J_1$-$J_2$ compound for the ferromagnetic $J_1 < 0$ and $J_2/|J_1| > 0.5$ even with a non-negligible interlayer coupling, it is expected that quantum fluctuations introduce, under high external field, the appearance of the nematic phase or the phase separation.

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