Spin nematic states in antiferromagnets containing ferromagnetic bonds

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The majority of recent works devoted to spin nematic phases deal with either frustrated magnets or with those described by Hamiltonians with large non-Heisenberg terms. We show in the present study that nonfrustrated antiferromagnets (AFs) containing ferromagnetic (FM) bonds can show nematic phases in a strong magnetic field. Among the particular spin systems discussed are a ladder with FM rungs, two AF layers coupled ferromagnetically, a chain containing alternating AF and FM bonds, and an AF anisotropic spin-1 chain.

Introduction. Frustrated spin systems have offered in recent years a wealth of opportunities for the study of a broad range of novel types of states and phase transitions. Spin nematic phases form a class of objects in this area which has received much attention. Spin nematic states are spin-liquid-like states which show a multiple-spin ordering without the conventional long-range magnetic order. The two-spin ordering can be generally described by the tensor[1] $Q_{jl}^{\alpha\beta} = \langle S_j^\alpha S_l^\beta \rangle - \delta_{\alpha\beta} \langle \mathbf{S}_j \mathbf{S}_l \rangle/3$. The antisymmetric part of $Q_{jl}^{\alpha\beta}$ is related to the vector chirality $\langle \mathbf{S}_j \times \mathbf{S}_l \rangle$ and describes a vector chiral spin liquid which can be stabilized in quantum spin models at $T = 0$ by a sizable ring exchange,[2] and can be found in classical frustrated spin systems at $T \neq 0$.[3] The symmetric part of $Q_{jl}^{\alpha\beta}$ describes a quadrupolar order which has been extensively studied both theoretically and experimentally in frustrated systems with ferromagnetic (FM) and antiferromagnetic (AF) nearest-neighbor and next-nearest-neighbor couplings, respectively, in strong magnetic field H (see, e.g., Ref. 4 and references therein) and in magnets with large non-Heisenberg spin couplings such as the biquadratic exchange $(\mathbf{S}_1\mathbf{S}_2)^2$.[5] It has been also shown recently that quantum fluctuations accompanied by a sizable single-ion easy-axis anisotropy can also stabilize a nematic phase in the kagome spin-1 antiferromagnet.[6] This study was motivated by recent experiments on $Ni_3V_2O_8$.

It is well established that the attraction between magnons caused by frustration is the origin of quadrupolar and multipolar phases in quantum magnets.[7] In particular, the bottom of the one-magnon band lies above the lowest multimagnon bound state at $H = H_s$, where H_s is the saturation field, as a result of this attraction in magnets with FM and AF couplings between nearest- and next-nearest neighbors, respectively. Then, transitions to nematic phases at $H < H_s$ in such systems are characterized by a softening of the multimagnon bound-state spectrum rather than the one-magnon spectrum.

We show in the present paper that FM bonds in AF nonfrustrated spin systems can also lead to a magnon attraction and to the stabilization of nematic phases in a strong magnetic field. To be specific, we discuss a spin-$\frac{1}{2}$ ladder with FM rungs, two AF layers coupled ferromagnetically, and a chain containing alternating AF and FM bonds, which are presented in Figs. 1(a)–1(c). The spin ladder with FM rungs has been actively discussed in recent years (Ref. 8) and numerical evedence is presented in Ref. 9 for the nematic phase appearence in strong field. Alternating chains have also received considerable interest recently which has not been related, however, to

their nematic behavior in magnetic field (see Ref. 10 and references therein). Although the interaction between spins is antiferromagnetic in the majority of compounds containing weakly coupled spin chains, ladders, or bilayers, some (mainly organic-based) materials containing FM bonds have been synthesized recently,[11] which has stimulated the theoretical activity in this field.

Model and technique. All spin-$\frac{1}{2}$ systems under discussion are described by the Hamiltonian

$$\mathcal{H} = J \sum_{\langle i,l \rangle} (\mathbf{S}_{1,i}\mathbf{S}_{1,l} + \mathbf{S}_{2,i}\mathbf{S}_{2,l}) - j \sum_i \mathbf{S}_{1,i}\mathbf{S}_{2,i}$$
$$- A \sum_i S_{1,i}^z S_{2,i}^z - H \sum_i (S_{1,i}^z + S_{2,i}^z) + \mathcal{H}', \quad (1)$$

where $\mathbf{S}_{n,i}$ is the nth spin in the ith FM bond, j and A are values of the isotropic and anisotropic parts of the FM exchange coupling, respectively, $\langle i,l \rangle$ denote nearest-neighbor spins coupled antiferromagnetically, we set the AF coupling constant J to be equal to unity in our calculations, and $\mathcal{H}'$ describes a small interladder (interbilayer or interchain) interaction.

The ground state of Hamiltonian (1) has a collinear antiferromagnetic spin structure at small H and the magnon spectrum has a gap induced by the easy-axis anisotropy A. There is a sequence of phase transitions upon the field increasing.

If the easy-axis anisotropy A is smaller than a critical value A_c [the classical value of A_c is J_0, where $J_{\mathbf{p}} = J \sum_j \exp(i\mathbf{p}\mathbf{R}_{jl})$, that does not depend on j], the first phase transition is the first-order spin flop one that happens when the field reaches a value H_{sf}. One has, for the classical value of H_{sf},

$$H_{sf}^{cl} = \tfrac{1}{2}\sqrt{A(2J_0 - A)}. \quad (2)$$

There is a canted AF spin structure at $H > H_{sf}$ and one of the magnon branches is gapless [as a consequence of the continuous SO(2) symmetry breakdown in this phase]. Our experience suggests that there would be only one phase transition to the fully polarized phase at $H = H_s$ upon further field increasing. However, we show below that in a range of parameters the fully polarized phase is preceded by a nematic one with the order parameter $\langle S_{1,i}^- S_{2,i}^- \rangle$ [see Fig. 1(d)]. A consideration of the nematic order parameter symmetry shows that SO(2)/Z_2 symmetry is broken in the nematic phase (see, e.g., Ref. 4). Then there is a Goldstone mode in the nematic phase and the transition from the canted AF phase to the
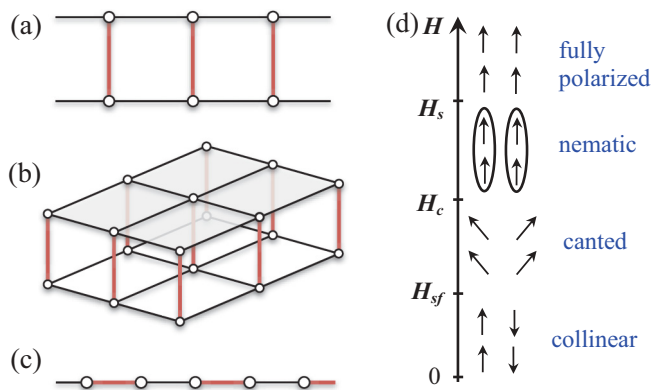
FIG. 1. (Color online) (a)–(c) Spin-$\frac{1}{2}$ systems which show a nematic phase in a strong magnetic field and which are discussed in the present paper in some detail. Antiferromagnetic and ferromagnetic bonds are shown in black and blue, respectively. (d) A sketch of the phase diagram at small T of the considered systems. The canted or the nematic phase is absent in some cases (see the text). Small interchain, interlayer, and interladder couplings are taken into account.

nematic one is apparently of a two-dimensional (2D) Ising type because the Z_2 subgroup breaks down.

If A is large enough (e.g., at $A \gg J, j$), there is no canted phase. We show below that even in this case the collinear and the fully polarized gapped phases are separated by the gapless nematic one.

We examine in the present paper the possibility of nematic phase formation just below the saturation field H_s by considering the transition from the fully polarized phase. We use for this purpose the approach suggested recently by one of us in Ref. 4 for a high-field nematic phases analysis that is based on the following bond-operator spin representation:

$$
\begin{aligned}
S_{1,i}^+ &= b_i^\dagger a_i + c_i, \quad S_{2,i}^+ = c_i^\dagger a_i + b_i, \\
S_{1,i}^- &= a_i^\dagger b_i + c_i^\dagger, \quad S_{2,i}^- = a_i^\dagger c_i + b_i^\dagger, \\
S_{1,i}^z &= \tfrac{1}{2} - a_i^\dagger a_i - c_i^\dagger c_i, \quad S_{2,i}^z = \tfrac{1}{2} - a_i^\dagger a_i - b_i^\dagger b_i,
\end{aligned}
\tag{3}
$$

where $a_i^\dagger$, $b_i^\dagger$, and $c_i^\dagger$ are Bose operators which create three spin states from the vacuum $|0\rangle = |\uparrow\uparrow\rangle$ as follows: $a_i^\dagger|0\rangle = |\downarrow\downarrow\rangle$, $b_i^\dagger|0\rangle = |\uparrow\downarrow\rangle$, and $c_i^\dagger|0\rangle = |\downarrow\uparrow\rangle$, where all spins have a maximum projection on the field direction at the state $|0\rangle$. To avoid the contribution of unphysical states containing more than one particle a, b, or c on a FM bond, we add to the Hamiltonian constraint terms describing an infinite repulsion between particles on each FM bond: $U \sum_i (a_i^\dagger a_i^\dagger a_i a_i + b_i^\dagger b_i^\dagger b_i b_i + c_i^\dagger c_i^\dagger c_i c_i + a_i^\dagger b_i^\dagger a_i b_i + b_i^\dagger c_i^\dagger b_i c_i + a_i^\dagger c_i^\dagger a_i c_i)$, where $U \to +\infty$. In particular, it is shown in Ref. 4 that (i) this approach is quite convenient for a discussion of the quantum phase transition to the nematic phase, (ii) along with some different results it yields also those obtained by other methods,[7,12] and (iii) although it gives quantitatively correct results at $H \approx H_s$ when $H < H_s$, it works in the nematic phase qualitatively also when H is not very close to H_s. For the sake of self-consistency, we present below some details of this approach. We confirm below our key analytical results by numerical ones obtained using the finite cluster diagonalization technique.[13]

Substituting Eqs. (3) into Hamiltonian (1), taking into account the constraint terms, and for a moment neglecting $\mathcal{H}'$, one obtains for the ladder and the bilayer

$$
\begin{aligned}
\mathcal{H}_2 = \sum_{\mathbf{p}} \Big[&(b_{\mathbf{p}}^\dagger b_{\mathbf{p}} + c_{\mathbf{p}}^\dagger c_{\mathbf{p}})\Big(H + \tfrac{1}{2}(J_{\mathbf{p}} - J_0) + \tfrac{1}{2}(j + A) \Big) \\
&- \tfrac{1}{2} j (b_{\mathbf{p}}^\dagger c_{\mathbf{p}} + c_{\mathbf{p}}^\dagger b_{\mathbf{p}}) + a_{\mathbf{p}}^\dagger a_{\mathbf{p}}(2H - J_0) \Big],
\end{aligned}
\tag{4}
$$

$$
\mathcal{H}_3 = \frac{1}{2\sqrt{N}} \sum [a_1^\dagger b_2 c_3 (J_2 + J_3) + b_1^\dagger c_2^\dagger a_3 (J_1 + J_2)], \tag{5}
$$

$$
\begin{aligned}
\mathcal{H}_4 = \frac{1}{N} \sum \Big[&a_1^\dagger a_2^\dagger a_3 a_4 (J_{1-3} + U) + U b_1^\dagger c_2^\dagger b_3 c_3 \\
&+ (a_1^\dagger b_2^\dagger a_3 b_4 + a_1^\dagger c_2^\dagger a_3 c_4)\Big(J_{1-3} + \tfrac{1}{2} J_{1-4} + U \Big) \\
&+ (b_1^\dagger b_2^\dagger b_3 b_4 + c_1^\dagger c_2^\dagger c_3 c_4)\Big(\tfrac{1}{2} J_{1-3} + U \Big) \Big],
\end{aligned}
\tag{6}
$$

where N is the number of FM bonds, the momentum conservation laws $\sum_i \mathbf{p}_i = 0$ are implied in Eqs. (5) and (6), and we omit some indexes $\mathbf{p}$ in Eqs. (5) and (6). It is convenient to introduce the following Green's functions:

$$
G_a(k) = -i\langle a_k a_k^\dagger \rangle, \tag{7}
$$

$$
G_b(k) = -i\langle b_k b_k^\dagger \rangle, \quad G_c(k) = -i\langle c_k c_k^\dagger \rangle, \tag{8}
$$

$$
F(k) = -i\langle b_k c_k^\dagger \rangle, \quad \overline{F}(k) = -i\langle c_k b_k^\dagger \rangle. \tag{9}
$$

Poles of $G_a(k)$ give the spectrum of a particles, whereas poles of $G_b(k)$, $G_c(k)$, $F(k)$, and $\overline{F}(k)$, which have the same denominator, determine the spectra of two one-magnon branches.[14]

Particles b and c carrying spin 1 are of a one-magnon nature. Their spectra calculated at $H > H_s$ using Eqs. (3) coincide with those derived using the conventional approaches such as the Holstein-Primakoff transformation. It can be shown[4] that spectra of one-magnon excitations are determined solely by $\mathcal{H}_2$ [Eq. (4)] at $H \geqslant H_s$ and they have the form

$$
\epsilon_1(\mathbf{p}) = H + \tfrac{1}{2}(J_{\mathbf{p}} - J_0) + \tfrac{1}{2}A, \tag{10}
$$

$$
\epsilon_2(\mathbf{p}) = \epsilon_1(\mathbf{p}) + j. \tag{11}
$$

The lower branch $\epsilon_1(\mathbf{p})$ has a minimum at $\mathbf{p} = \pi$ [or (π,π)] and a gap which closes when H becomes equal to

$$
H_c = J_0 - A/2. \tag{12}
$$

In contrast to b and c particles, a particles carrying spin 2 are of a two-magnon nature. Their spectrum coincides with the two-magnon bound-state spectrum found using conventional methods.[14] To find the spectrum $\epsilon_a(\mathbf{p})$ of a particles at $H \geqslant H_s$ one has to take into account diagrams shown in Fig. 2(a) which contain three-particle vertices. Equations for them are presented in Fig. 2(b). The minimum of $\epsilon_a(\mathbf{p})$ is at $\mathbf{p} = 0$ and the gap in $\epsilon_a(\mathbf{p})$ closes at $H = H_c'$. If $H_c > H_c'$, the transition takes place at $H = H_c = H_s$ to the canted phase, which can be described as the Bose-Einstein condensation (BEC) of one-magnon excitations. As it is seen from Eqs. (3), $\langle \mathbf{S}_{n,i}^\perp \rangle$ becomes finite in this case, where $\perp$ denotes the projection on the xy plane. In contrast, if $H_c < H_c'$, the transition from
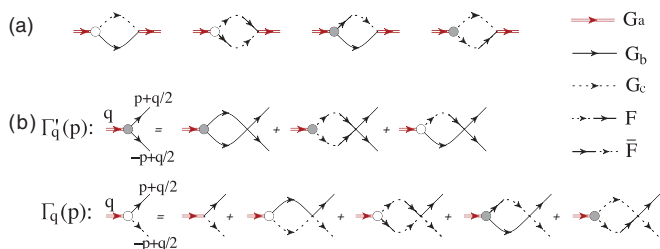
FIG. 2. (Color online) (a) Diagrams for the self-energy part $\Sigma_a(k)$ of a particles at $H \gtrsim H_s$. Green's functions of b and c particles $G_b(p)$, $G_c(p)$, $F(p)$, and $\overline{F}(p)$ are defined by Eqs. (8) and (9). Circles represent renormalized vertices. Equations for the vertices are presented in (b). Bare vertices are defined by Eqs. (5) and (6).

the fully polarized phase to the nematic one takes place at $H = H_c' = H_s$, which can be described within our approach as the BEC of a particles. It is seen from Eqs. (3) that $\langle \mathbf{S}_{n,i}^\perp \rangle = 0$ in the nematic phase and $\langle S_{1,j}^- S_{2,j}^- \rangle \equiv \langle a_j^\dagger \rangle \propto e^{-i\phi}\sqrt{H_s(T) - H}$ is the nematic order parameter (for $\mathcal{H}' \neq 0$), where ϕ is an arbitrary phase.[4] All the static two-spin correlators decay exponentially in the nematic phase. We calculate below $\epsilon_a(\mathbf{p})$ for the selected spin systems and find stability conditions of the nematic states.

AF ladder with FM rungs [see Fig. 1(a)]. Unfortunately, the general expression for $\epsilon_a(\mathbf{p})$ is quite complicated but it is simplified greatly in the limiting case of the Ising exchange on the FM bond (i.e., at $j = 0$). To illustrate the main properties of the nematic phase we consider here in some detail the case of $j = 0$. In particular, one finds at $\mathcal{H}' = 0$ and $H \gtrsim H_s$,

$$\epsilon_a(\mathbf{p}) = 2H - 2 + A - \sqrt{4\cos^2\frac{p}{2} + A^2}. \quad (13)$$

The gap in $\epsilon_a(\mathbf{p})$ vanishes at $H_s = 1 + \sqrt{1 + A^2/4} - A/2$, which is larger than $H_c = 2 - A/2$ [see Eq. (12)] at all positive A. Then, the nematic phase is stable at $A > 0$ when $j = 0$. Interestingly, the nematic phase arises also in the case of $A \gg 1$, when the canted phase is absent. Thus, the gapless nematic phase separates two gapped phases at large A, the fully polarized and the collinear ones.

Spectrum (13) has a quadratic dispersion near its minimum at $p = 0$: $\epsilon_a(\mathbf{p}) \approx 2H - 2 + A - \sqrt{4 + A^2} + D_\parallel p^2$, where $D_\parallel = 1/2\sqrt{4 + A^2}$. The spectrum remains quadratic also after taking into account a small $\mathcal{H}'$ that leads to the three-dimensional (3D) BEC relation $H_s(0) - H_s(T) \propto T^{3/2}$.[4]

One obtains in the first order in ρ and in the leading order in the interladder interaction, as it is done in Ref. 4 for quasi-one-dimensional (1D) frustrated systems,

$$\langle S_{1,j}^- S_{2,j+n}^- \rangle = \sqrt{\rho}\, e^{-i\phi} \frac{4}{A^2}\left(\frac{A}{2} - \sqrt{1 + \frac{A^2}{4}}\right)^n, \quad (14)$$

$$\langle S_{1,j}^+ S_{1,j+n}^- \rangle = \langle S_{2,j}^+ S_{2,j+n}^- \rangle$$
$$= \rho \frac{4}{A^2}\left(\frac{4n}{A^2} + \frac{4\sqrt{4 + A^2}}{A^3} - 2\right)$$
$$\times \left(\frac{A}{2} - \sqrt{1 + \frac{A^2}{4}}\right)^n, \quad (15)$$

$$\left\langle \left(S_{1,j}^z - \tfrac{1}{2}\right)\left(S_{2,j+n}^z - \tfrac{1}{2}\right)\right\rangle = |\langle S_{1,j}^- S_{2,j+n}^- \rangle|^2, \quad (16)$$

where $n > 0$ and $\rho = \langle a_i^\dagger a_i \rangle \propto [H_s(T) - H]$ is the "condensate" density. The rest of the two-spin static correlators containing S^+ or S^- are equal to zero and $\langle S_{1,j}^z S_{1,j+n}^z \rangle = \langle S_{2,j}^z S_{2,j+n}^z \rangle \sim \rho^2$. It is seen from Eqs. (14)–(16) that all correlators decay exponentially at $A > 0$.

One obtains for the magnetization

$$\langle S_{q,j}^z \rangle = \frac{1}{2} - \left(2 - \frac{8}{A^2} + \frac{16\sqrt{4 + A^2}}{A^5}\right)\rho, \quad (17)$$

where $q = 1, 2$. It may seem that Eqs. (14)–(17) are invalid for arbitrary small A. However, the above results are valid in the near vicinity of H_s (i.e., at very small ρ) in this case because H_s and H_c merge in the limit $A \to 0$.

It is implied in Eqs. (14)–(17) that $\mathcal{H}' \neq 0$ so that $\rho \neq 0$. The situation is completely different in the purely 1D case because $\rho \equiv 0$ and there is no long-range nematic order. Bearing in mind the quadratic dispersion of $\epsilon_a(\mathbf{p})$ near its minimum and using the results of 1D Bose-gas discussions,[15] one obtains,[14] at $H \approx H_s$ and $T = 0$,

$$\frac{1}{2} - \langle S_{q,j}^z \rangle = \langle a_j^\dagger a_j \rangle = \frac{1}{\pi}\sqrt{\frac{2(H_s - H)}{D_\parallel}}, \quad (18)$$

$$\langle S_{q,j+n}^z(t) S_{p,j}^z(0) \rangle \approx \langle S_j^z \rangle^2 - \frac{1}{\pi}\left(\frac{1}{(n + iut)^2} + \frac{1}{(n - iut)^2}\right)$$
$$+ B_1 \frac{\cos(\pi\langle a_j^\dagger a_j\rangle n)}{n^2 + u^2 t^2}, \quad (19)$$

$$\langle S_{1,0}^+(t) S_{2,0}^+(t) S_{1,n}^-(0) S_{2,n}^-(0) \rangle \approx \frac{B_2}{\sqrt{|n + iut|}}, \quad (20)$$

where $q, p = 1, 2$, $n \to \infty$, $u = 4\pi D_\parallel \langle a_j^\dagger a_j \rangle$, and $B_{1,2}$ are constants.

An analysis of the general expression for $\epsilon_a(\mathbf{p})$ gives the following general criterion of the nematic phase stability,

$$A > \frac{4j}{j + 2\sqrt{j(j+2)}}, \quad (21)$$

which is shown graphically in Fig. 3. One concludes from Eq. (21) that only large $A \gtrsim j$ can stabilize the nematic phase if $j \sim 1$. In contrast, quite small anisotropy on FM bonds $j \gg A \sim 1$ is sufficient for this purpose if $j \gg 1$.

In the limiting case of $j \to \infty$, our model describes the spin-1 AF chain with the easy-axis single-ion anisotropy A. The nematic order parameter reads in this case as $\langle (S_i^-)^2 \rangle$, and one obtains from Eq. (21) that the nematic phase is stable at $A > 4/3$. A similar nematic phase is discussed in Ref. 6 in a spin-1 kagome AF with large single-ion easy-axis anisotropy at $H = 0$.

Our finite cluster calculations confirm that the transition takes place at $H = H_s$ to the nematic phase when inequality (21) holds. This numerical consideration is simplified by the fact that the Hamiltonian (1) commutes with the z component of the total spin $\mathcal{S}^z$ and with the Zeeman term. As a result all the Hamiltonian eigenstates can be classified by eigenvalues M of $\mathcal{S}^z$. Let us denote $E(M)$ the minimum energy in each M sector at $H = 0$. The ground state energy of a cluster with
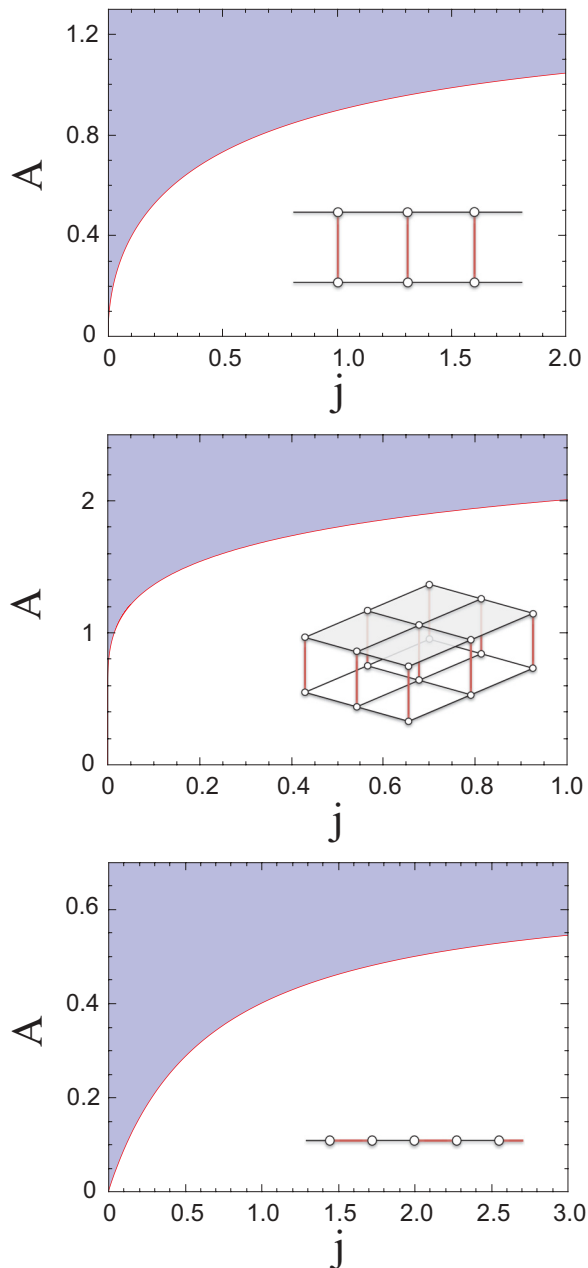
FIG. 3. (Color online) Regions of the nematic phase stability in the ladder with FM rungs [inequality (21)], in the bilayer with FM coupling [inequality (22)], and in the alternating chain.

$M_{\rm sat} = L/2$. Thus, one can expect that a condensation takes place in the thermodynamic limit of elementary excitations carrying spin 2. Then, the lowest state in each even $(M_{\rm sat} - M)$ sector has zero momentum (if the periodic boundary conditions are applied), which is also in agreement with the bound-state condensation scenario. Values of H_s obtained numerically as a result of an analysis of clusters with $L = 8$–48 are in excellent agreement with the corresponding values found analytically. A numerical consideration of clusters with $L = 8$–26 similar to that performed in Ref. 16 confirms also the validity of Eq. (18). For example, one obtains for $j = 0$ and $A = 2$ that $\frac{1}{2} - \langle S_{q,j}^z \rangle = \alpha(H_s - H)^{1/\delta}$, where $\alpha = 1.04 \pm 0.11$ and $\delta = 2.05 \pm 0.05$, which is in good agreement with the analytical result $\langle a_j^\dagger a_j \rangle \approx 1.07\sqrt{H_s - H}$. Our numerical results are also fully consistent with those of Ref. 9.

AF bilayer with FM coupling [see Fig. 1(b)]. The analytical analysis of this system is much more complicated than that carried out above for the ladder. Then, we restrict ourselves by a discussion of the region of the nematic phase stability that is defined by the inequality (see Fig. 3)

$$A > \frac{8 - 8jf(j)}{(8 + j)f(j) - 1}, \qquad (22)$$

$$f(j) = \frac{1}{(2\pi)^2} \int_0^{2\pi} \frac{dxdy}{j + 2 + \cos x + \cos y}. \qquad (23)$$

One concludes from Eq. (22) that, similar to the ladder with FM rungs, only large $A \gtrsim j$ can stabilize the nematic phase if $j \sim 1$. In contrast, quite small anisotropy on FM bonds $j \gg A \sim 1$ is sufficient for this purpose if $j \gg 1$.

Alternating chain [see Fig. 1(c)]. Particular expressions are quite cumbersome in this case. Then we restrict ourselves by a graphical representation of the nematic phase stability region which is also shown in Fig. 3. It is seen that the graphic resembles those for the ladder and the bilayer. A numerical consideration confirms the existence of the nematic phase and the validity of Eq. (18). Numerical values of H_s are in excellent agreement with analytical results.

Conclusion. We demonstrate that FM bonds in nonfrustrated antiferromagnets can lead to nematic spin states in a strong magnetic field. The uniaxial anisotropy on the FM bond is necessary for nematic phase stabilization in all the considered systems. The present study should stimulate further theoretical and experimental activity in nematic phase discussions both in the considered systems and in other ones containing FM bonds.

L spins in a magnetic field is given by the minimum value of $E(M) - HM$. An important observation is that the values $E(M) - HM$ at even $M_{\rm sat} - M$ are smaller than those with odd $M_{\rm sat} - M$ when H is close to its saturation value, where

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