Spin nematic state for a spin $S=3/2$ isotropic non-Heisenberg magnet

Yu. A. Fridman,$^1$ O. A. Kosmachev,$^1$ and B. A. Ivanov$^2$

$^1$V.I. Vernadsky Taurida national university Vernadsky ave., 4, Simferopol, Ukraine
$^2$Institute of Magnetism, National Academy of Sciences and Ministry of Education of Ukraine, 36(b) Vernadskii avenue, 03142 Kiev, Ukraine

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Abstract

$S = 3/2$ system with general isotropic nearest-neighbor exchange within a mean-field approximation possesses a magnetically ordered ferromagnetic state and antiferromagnetic state, and two different spin nematic states, with zero spin expectation values. Both spin nematic phases display complicated symmetry break, including standard rotational break described by the vector-director $\vec{u}$ and specific symmetry break with respect to the time reversal. The break of time reversal is determined by non-trivial quantum averages cubic over the spin components and can be described by unit “pseudospin” vector $\vec{\sigma}$. The vectors $\vec{\sigma}$ on different sites are parallel for a nematic state, and $\vec{\sigma}$’s are antiparallel for different sublattices for an antinematic phase.

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Quantum spin systems have provided a wide playground for the quest of novel types of quantum ordered states and quantum phase transitions. A number of exotic states has been discovered, the most well-known examples are the famous Haldane phase in integer spin antiferromagnetic chains \cite{1} and quantum spin nematic states in the spin-1 bilinear-biquadratic isotropic magnets. A quantum spin nematic state has zero value of dipolar spin order parameter, $\langle \vec{S} \rangle = 0$, but spin rotation symmetry is spontaneously broken due to nontrivial quadrupolar spin expectation values of the type of $\langle S_i S_j + S_j S_i \rangle$, $i, j = x, y, z$. For $S = 1$ spin operators, the quadrupolar variable have uniaxial symmetry and can be written through the condition $\langle (\vec{S}, \vec{u})^2 \rangle = 0$. The order parameter can be present via the vector director $\vec{u}$, in complete analogy to nematic liquid crystals. In the last two decades, this state has been actively studied for description of crystalline magnets, see for review \cite{2, 3}, including low-dimensional magnets, see recent articles \cite{4, 5, 6}. The interest has got a considerable impact, motivated by investigation of multicomponent Bose-Einstein condensates of neutral atoms with nonzero integer spins \cite{7, 8, 9}. The investigation of spin nematic states for $S = 1$ systems has been performed for two different physical models, spin lattice system and and Bose gas of interacting particles with non-zero spin. The results obtained within both approaches are in a good agreement and complement each other.

A few novel spin nematic states are found for Bose gas of interacting atoms with spin $S = 2$, including the state with non-uniaxial symmetry \cite{10}. The question of the possibility of some kind of spin nematic states for half-integer spins (the minimal half-integer spin value allowing such state in $S = 3/2$) is of large general importance for physics of Fermi systems. As was found in Refs. \cite{11, 12, 13}, high spin fermionic systems (ultracold Fermi gases) in some parameter region also exhibit many properties common to that for spin nematic states. On the best of our knowledge, for higher spins $S > 1$, both integer and half-integer, the only approach based on the direct analysis of gas of interacting particle has been used.

The aim of this Letter is to develop a mean-field analysis of the ground state of spin $S = 3/2$ isotropic spin Hamiltonian, with special attention to the spin nematic state, and to discuss the symmetry and the excitation spectra for such states.

The most general isotropic exchange interaction for pare of spins $S = 3/2$ includes, additionally to Heisenberg bilinear interaction, non-heisenberg (biquadratic and bicubic) terms as well, which naturally leads to the model described by the following nearest-neighbor
interaction Hamiltonian:

\[ \hat{H} = -\sum_{\langle \vec{l}, \vec{\delta} \rangle} \left[ J \vec{S}_{\vec{l}} \cdot \vec{S}_{\vec{l}+\vec{\delta}} + K \left( \vec{S}_{\vec{l}} \cdot \vec{S}_{\vec{l}+\vec{\delta}} \right)^2 + L \left( \vec{S}_{\vec{l}} \cdot \vec{S}_{\vec{l}+\vec{\delta}} \right)^3 \right] \tag{1} \]

where \( \vec{S}_{\vec{l}} \) are spin-3/2 operators at the lattice site \( \vec{l} \); \( J, K, \) and \( L \) are the exchange constants, corresponding to the bilinear, the biquadratic, and the bicubic exchange interactions, respectively, and summation over pairs of the nearest neighbors \( \vec{\delta} \) is implied.

The spin-3/2 state \( |\psi_{\vec{l}}\rangle \) is a superposition of four basis states \( S_z |\psi_s\rangle = s |\psi_s\rangle \), with coefficients \( z_s \), \( s = 3/2, 1/2, -1/2 \) and \(-3/2\). The essential quantities are the ratios of these complex numbers, and the system parameter manifold is three-dimensional complex projective space \( \mathbb{CP}^3 \). It is convenient to present the ratios of \( z_s \) as,

\[
\begin{align*}
\frac{z_{-3/2}}{z_{3/2}} &= e^{i\varphi} \cdot \tan \frac{\theta}{2}, \quad \frac{z_{-1/2}}{z_{1/2}} = e^{i\beta} \cdot \tan \frac{\alpha}{2}, \\
\frac{z_{1/2}}{z_{3/2}} &= e^{i\gamma} \cdot \tan \mu \cdot \cos \frac{\alpha}{2} \cdot \sec \frac{\theta}{2}.
\end{align*}
\tag{2}
\]

It is easy to check that the resolution of identity \( \int D[\psi] |\psi\rangle \langle \psi| = \hat{1} \), with the proper measure \( D[\psi] \), is satisfied, and this six-parameter state has the properties of SU(4) coherent states.

Using the states \( |\psi\rangle \), one can construct the coherent state path integral and find the effective Lagrangian \( \mathcal{L} \)

\[
\begin{align*}
\mathcal{L} = \hbar \sum_{\vec{l}} & \left\{ \left[ \partial_t \gamma + (\partial_t \beta) \sin^2 \frac{\alpha}{2} \right] \sin^2 \mu + \\
& + (\partial_t \varphi) \sin^2 \frac{\theta}{2} \cos^2 \mu \right\} - W, \quad W = \langle \hat{H} \rangle,
\end{align*}
\tag{3}
\]

where \( \partial_t = \partial / \partial t \) and \( W \) is the “classical” (mean-field) energy of the system, which equals to the quantum mean value of the Hamiltonian with the states \( |\psi\rangle \).

The analysis of the ground state can be considerably simplified by making a rotation in the spin space to the principal axes of the spin-quadrupolar ellipsoid, i.e. by the use of condition \( \langle S^i S^j + S^j S^i \rangle = 0 \), if \( i \neq j \). This condition yields the relations \( \theta + \alpha = \pi, \beta = -\gamma, \varphi = \gamma \), and we can operate with the three-parameter state

\[
|\psi\rangle = \cos \mu \left[ \sin \frac{\alpha}{2} \left\{ \frac{3}{2} \right\} + e^{i\gamma} \cos \frac{\alpha}{2} \left\{ \frac{-3}{2} \right\} \right] + \\
+ \sin \mu \left[ \sin \frac{\alpha}{2} \left\{ \frac{1}{2} \right\} + e^{i\gamma} \cos \frac{\alpha}{2} \left\{ \frac{1}{2} \right\} \right].
\tag{4}
\]
FIG. 1: (Color online) The domains of stability for different phases present through “angular variables” \( \Pi \). For this state, diagonal biquadratic expectation values are independent on \( \gamma \) and \( \alpha \), they can be written as

\[
4 \langle S_x^2 \rangle = 3 + 2\sqrt{3} \sin 2\mu + 4 \sin^2 \mu,
\]
\[
4 \langle S_y^2 \rangle = 3 - 2\sqrt{3} \sin 2\mu + 4 \sin^2 \mu. \tag{5}
\]

The mean values of the spin operators are determined by all three parameters,

\[
\langle S_x + iS_y \rangle = \sin \mu \sin \alpha \left[ e^{-i\gamma} \sin \mu + e^{i\gamma} \sqrt{3} \cos \mu \right],
\]
\[
2 \langle S_z \rangle = \cos \alpha \left[ 1 - 4 \cos^2 \mu \right], \tag{6}
\]

and the angular variable \( \gamma \) determines the orientation of the mean value of spin operator \( \langle \vec{S} \rangle \) in the \( xy \)-plane.

Assuming a uniform ground state, one arrives to the following expression for the energy of the system \( W \)

\[
W = -\left( J - \frac{K}{2} + \frac{103}{16} L \right) \left[ \frac{\cos^2 \alpha}{4} \left( \sin^2 \mu - 3 \cos^2 \mu \right)^2 + \sin^2 \mu \sin^2 \alpha \left( 1 + 2 \cos^2 \mu + \sqrt{3} \sin 2\mu \cos 2\gamma \right) \right]. \tag{7}
\]

In fact, Eq. (7) gives us the free energy of the non-heisenberg \( S = 3/2 \) magnet with the Hamiltonian \( H \) at zero temperature \( T \). Minimizing \( W \) over parameters \( \mu, \alpha \) and \( \gamma \), one can...
find possible states of the system. Our analysis shows, at $\Lambda_1 > 0$, where

$$
\Lambda_1 = 16J - 8K + 103L,
$$

the minimum corresponds to the ferromagnetic phase with the saturated spin. For this state $\alpha = \pi, \mu = 0$, and

$$
|\Psi_{\text{ferromagnet}}\rangle = \frac{3}{2}, \langle S^z \rangle = \frac{3}{2}, \langle S^x \rangle = \langle S^y \rangle = 0.
$$

(9)

Otherwise, at $\Lambda_1 < 0$ the minimum corresponds to the spin nematic state with $\alpha = \pi/2$, $\mu = 0$, $\langle \vec{S} \rangle = 0$, and

$$
|\Psi_{\text{nematic}}\rangle = \frac{1}{\sqrt{2}} \left( \frac{3}{2} \left| \frac{3}{2} \rightangle + \frac{-3}{2} \right),
$$

$$
\langle (S^z)^2 \rangle = \frac{9}{4}, \langle (S^x)^2 \rangle = \langle (S^y)^2 \rangle = \frac{3}{4}.
$$

(10)

The image of this state is an elongated ellipsoid of evolution which can be described by unit vector-director $\vec{u}$, $\vec{u}$ is parallel to $z-$axis for $\langle 10 \rangle$. So, the quantum phase transition “ferromagnetic state – spin nematic state” occurs at $\Lambda_1 = 0$. In a natural parametrization of the exchange parameters through “angular variables” of the form

$$
J = \tilde{J} \cos \Theta, \quad K = \tilde{J} \sin \Theta \cos \Phi, \quad L = \tilde{J} \sin \Theta \sin \Phi,
$$

(11)

the set of system parameters can be presented as a point on a sphere $J^2 + K^2 + L^2 = \tilde{J}^2$. The condition $\Lambda_1 = 0$ is presented by a big circle on the sphere, passing through the direction $\vec{p}$ with $\tan \Theta = 2J/K$ and $\Phi = 0$, and the point with $L = 8K/103$ on the equator, see Fig. 1. On this line, all uniform states of the system are continuously degenerated on the parameters $\alpha$ and $\mu$. Thus there are lines with high symmetry in the parameter space of the $S = 3/2$ magnet. Note the equality $\Lambda_1 = 0$ corresponds to the condition of lifting of the symmetry of spin $S = 3/2$ Fermi gas in one band Habbard model, see Eq.(55) of Ref. 13.

The complete investigation of the stability of any phase with respect to arbitrary small perturbation should include an analysis of evolution of small deviations from the uniform state, $\delta \alpha_{i,i} = \alpha_{i,i} - \alpha_{i,0}$, where $\alpha_i$ denotes one of six parameters of the state $\langle 2 \rangle$, $\alpha_{i,0}$ corresponds to the ground state. Following the Bloch theorem, one can write $\delta \alpha_{i,i} = \Sigma \delta \alpha_{k,i} \cdot \exp(i\vec{k}\vec{r})$, $\vec{k}$ takes values within the first Brillouin zone. Information about stability can be extracted from the spectrum of elementary excitations $\omega = \omega_i(\vec{k})$ found
for the state of interest. Such excitations can be learned by use of different approximate quantum approaches, e.g. bosonization of spin operators with the $1/N$ expansion \cite{14}, or the Habbard operator technique, see, for example, \cite{15}. Alternatively, one can apply a semiclassical treatment based on the Lagrangian \cite{3}, with the usage of the concrete form of $W$, that is consistent with mean-field ground state calculations \cite{16}. We had used two last methods for a $d$--dimensional hypercubic lattice, and found (as for spin $S = 1$ magnets, see \cite{16}) the same results for the both approaches.

Variation of the Lagrangian \cite{3} leads to the Hamilton system for generalized coordinates $\delta \gamma_{\vec{k}}$, $\delta \beta_{\vec{k}}$ and $\delta \phi_{\vec{k}}$, its solution gives three branches of elementary excitations. Here we discuss briefly the properties of modes important for understanding of the stability of states.

For the spin nematic state, the properties of two spin wave modes are common to that for a spin $S = 1$ nematic, their spectra are degenerated with the gapless dispersion law, linear over $k = |\vec{k}|$ at $k \to 0$, $\omega_{1,2}(\vec{k}) \to c k$, where $\hbar c = (3a/8) \sqrt{-2z\Lambda_1 \cdot (4K - 5L)}$. The third mode also has a gapless spectrum with linear asimptotics at $k \to 0$, $\hbar \omega_3(\vec{k}) = \frac{9}{4} \sqrt{L_k (z\Lambda_1 + 16J_k - 8K_k + 119L_k)}$, \eqref{12}, \hfill \( \text{(12)} \)

where $z$ is a coordination number, $J_k$, $K_k$ and $L_k$ are Fourier-components of exchange constants of the form

$$ J_k = J \sum_{\delta} \left( 1 - e^{i\vec{k}\vec{\delta}} \right), \quad J_k \sim J(ak)^2 \text{ at } k \to 0. $$

The phase speed $c_3$ of this mode vanishes at $L \to 0$, $\hbar c_3 = (9a/4) \sqrt{-zL\Lambda_1}$. For $\Lambda_1 < 0$ these modes describe a long-wave instability at the line \cite{8}. Additionally, at $\Lambda_2 < 0$, where

$$ \Lambda_2 = 16J - 8K + 135L, \quad \text{(13)} $$

the values of $\omega_3$ are imaginary for the quasimomentum $\vec{k}$ at the edge of Brillouin zone, where $\vec{J}_k = 2zJ$, ect. This means the instability, which leads to the transition to two-sublattice phase (antiferromagnetic, see below) on the line \cite{13}. Thus, the spin nematic state is stable if and only under the conditions $\Lambda_1 < 0$ and $\Lambda_2 > 0$. Note the line $\Lambda_2 = 0$ describes a big circle, passing through the same direction $\vec{p}$ as for $\Lambda_1 = 0$, see Fig. 1. Thus, the nematic state possesses three Goldstone modes (not two as for $S = 1$ system). We will discuss this fact below.

The situation for the ferromagnetic state is simpler. One mode corresponds with the spin oscillations with $|\langle \vec{S} \rangle| = 3/2$; it have the gapless dispersion law, parabolic at $k \to 0$,\hfill \( \text{\cite{18}} \)
\[16\hbar \omega_1(k) = (ak)^2(48J + 72K + 189L),\] and standard for Heisenberg ferromagnets. Two other modes possess oscillations of \(|\langle \vec{S} \rangle|\). They have gaps, proportional to \(\Lambda_1\) and describe the long-wave instability at \(\Lambda_1 < 0\). As well, these modes at \(\Lambda_2 < 0\) show the instability with respect to the transition to a two–sublattice state (an antinematic phase, see below). Thus, ferromagnetic state is stable if and only \(\Lambda_1 > 0\) and \(\Lambda_2 > 0\).

Let us return to spin nematic state. An appearance of the third Goldstone mode \((12)\) for this state can not be explained within a standard imaging of a nematic state as a ellipsoid of rotation, connected with bilinear spin expectation values \((10)\). But for spin \(S = 3/2\) magnet, an additional break of symmetry is caused by non-trivial expectation values cubic over spin components. For the state \((1)\), such values are \(\sigma_{(+)} = (1/3)\langle (S_x + iS_y)^3 \rangle\) and \(\sigma_{(-)} = (1/3)\langle (S_x - iS_y)^3 \rangle\), \(\sigma_{(\pm)} = \cos^2 \mu \sin \alpha \exp (\pm i\gamma)\). The transformation properties of the quantities \(\sigma_x = (\sigma_{(+)} + \sigma_{(-)})/2\) and \(\sigma_y = i(\sigma_{(+)}) - (\sigma_{(+)})/2\) are the same as for the components of the planar (two-dimensional) vector \(\vec{\sigma}\), \(\vec{\sigma} \propto \vec{e}_x \cos \gamma + \vec{e}_y \sin \gamma\) under the rotation around \(z\)-axis, \(|\vec{\sigma}| = 1\) for the nematic state. Having in mind that \(\vec{\sigma}\) changes its sign at time reversal \(t \to -t\), we can say that the properties of this unit vector are common to that for spin expectation value and the mode \((12)\) is associated with the oscillations of this vector.

The presence of such characteristic as \(\vec{\sigma}\) is a feature of principal importance. As we mentioned above, for a spin \(S = 1\) nematic, the symmetry breaking is associated with the quadrupolar ellipsoid only. But the spin \(S = 3/2\) nematic state is characterized additionally by unit “pseudospin” vector \(\vec{\sigma}\), which changes the sign at time reversal. The additional mode with \(\omega_3(\vec{k})\) has the same properties as for an easy plane Heisenberg ferromagnet, with the vector \(\vec{\sigma}\) playing the role of the in-plane magnetization. Thus, we arrive to the following non-trivial picture of the spin-3/2 nematic state: it is independently SO(3) degenerated over the orientation of the unit vector-director \(\vec{u}\), parallel to the long axis of the ellipsoid, and SO(2)-degenerated over the direction of unit pseudospin vector \(\vec{\sigma}\), perpendicular to \(\vec{u}\). The presence of this complicated spontaneous symmetry breaking leads to the appearance of three aforementioned Goldstone modes for this phase.

This features of uniform nematic state gives rise for even more non-trivial properties of multi-sublattice state with \(\langle \vec{S} \rangle = 0\). For the spin \(S = 1\), either a two-sublattice “orthogonal nematic” with \((\vec{u}_1 \cdot \vec{u}_2) = 0\) \([14]\), or a “threemerized” states \([17]\), have been discussed. As we have shown, for the spin-3/2 system with bipartite lattice, two-sublattice “antinematic”
state, with parallel $\vec{u}_1 = \vec{u}_2$, but antiparallel $\vec{\sigma}_1 = -\vec{\sigma}_2$, is stable in the region

$$\Lambda_2 < 0, \Lambda_1 > 0.$$ \hspace{1cm} (14)

For the rest of the parameter region, $\Lambda_2 < 0, \Lambda_1 < 0$, we have shown the presence of the two-sublattice antiferromagnetic state with $\langle \vec{S}_1 \rangle = -\langle \vec{S}_2 \rangle$, $|\langle \vec{S}_{1,2} \rangle| = 3/2$.

The above analysis has been done in the mean-field approximation only. A rich variety of “spin-liquid” states with properties governed by non-small quantum fluctuations, are known for one-dimensional (1d) spin-one systems (spin chains), where the mean-field approximation is not valid. But one can expect for the spin-3/2 system the role of such fluctuations should be different from those for a spin-1. In line with Haldane conjecture, for a spin $S = 3/2$ chain one can expect a transformation of the antiferromagnetic state to a “critical” state with gapless elementary excitations and antiferromagnetic correlations decaying with power law (compare with gapped Haldane state [1] for integer spins $S = 1, 2$ cases). The gapless Luttinger liquid state for one-dimensional spin $S = 3/2$ Fermi gas has been found by Wu [12].

For a spin $S = 1$ chain, nematic order is probably destroyed by non-perturbative quantum fluctuations [5]. As has been shown above for the spin $S = 3/2$ nematic and antinematic states the ordering of vector-director $\vec{u}$ is accompanied by “pseudospin” $\vec{\sigma}$ ordering. It is clear that the stability conditions for such states should differ from that for $S = 1$, a time-reversal “pure nematic” state. One can expect quite non-trivial behavior of the nematic and antinematic states for 1d systems, but a detail investigation of these features is going far from the scope of this Letter.

Thus, an isotropic magnet with spin $S = 3/2$ at $L \neq 0$ shows (at least, in mean-field approximation) the presence of spin nematic and spin antinematic states with unique dynamic and static properties. The adequate description of an isotropic magnet with $S = 3/2$ needs consideration of all three possible spin invariants in the exchange Hamiltonian [11]. Unfortunately, we can not provide a concrete example of a crystalline magnet with spin $S = 3/2$ and high enough biquadratic exchange. Moreover, to our knowledge, the values of bicubic exchange constant $L$, important for the presence of nematic states, has never been discussed for any real compounds. But the technique of ultracold gases of neutral atoms loaded into optical lattices gives the possibility of unprecedented control over the model parameters (exchange integrals, in our consideration). Alkali atoms $^{132}$Cs, as well as alkaline-earth atoms $^9$Be, $^{135}$Ba, and $^{137}$Ba, having hyperfine 3/2 spin, could be used for the study of quantum
phase transitions described above.

To conclude, as for well-studied spin $S = 1$ models, spin $S = 3/2$ system within the mean-field approximation have two magnetically ordered phases, the ferromagnetic state and the antiferromagnetic state with maximal possible magnitude of spin on a site. As well, there are two different nematic states, in which the average spin equals zero. The domains of the stability for the nematic states separate the stability regions of the ferromagnetic and the antiferromagnetic phases, see Fig. 1. On the phase transition lines, $\Lambda_1 = 0$ or $\Lambda_2 = 0$ the symmetry is higher than the SO(3)$\sim$SU(2) rotational symmetry of the Hamiltonian. The qualitative differences between the nematic state for the $S = 3/2$ and $S = 1$ magnets are as follows. The spin $S = 3/2$ nematic phases display specific symmetry break with respect to the time reversal. It is exhibited by the properties of the quantum averages cubic over the spin components, which can be organized in “pseudospin” vector $\vec{\sigma}$. By virtue of this fact the spectrum of spin oscillations in the spin nematic state includes the third Goldstone (quasiferromagnetic) mode. The antinematic phase has the antiparallel orientations of $\vec{\sigma}$ at different sublattices and possesses the following element of symmetry: the time reflection combined with the spatial translation on the nearest neighbor vector $\vec{\delta}$.

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* Electronic address: bivanov@i.com.ua

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