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ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED SYSTEM

Dynamic Properties of Magnets with Spin $S = 3/2$ and Non-Heisenberg Isotropic Interaction

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Abstract—The dynamic properties of a magnet with magnetic-ion spin of 3/2 and an isotropic spin interaction of a general form have been investigated. Only four phase states can be realized in the system under consideration at various relationships between the material parameters: the ferro- and antiferromagnetic phases with saturated spin and the states with tensor order parameters, the nematic and antinematic ones. For these phases, the spontaneous symmetry breaking is determined by the octupole order parameter containing the mean values trilinear in spin operator components at a given site. The spectra of elementary excitations have been determined in all phases. Additional branches of excitations arise in all four phase states

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1. INTRODUCTION

The ordering in spin systems is usually associated with the standard magnetic order for which the mean spins $\langle \mathbf{S}_n \rangle$ at the sites are nonzero and form various magnetic structures (ferromagnets, antiferromagnets, etc.; see [1, 2]). The main property of magnetically ordered systems is symmetry breaking with respect to time reversal, $\langle \mathbf{S}_n \rangle \longrightarrow -\langle \mathbf{S}_n \rangle$ when $t \longrightarrow -t$. However, the possibility of the existence of a spin-nematic state for which the mean spins at the sites $\langle \mathbf{S}_n \rangle$ are zero, but the spontaneous symmetry breaking in the spin system is associated with the anisotropy of some higher spin projection correlators was pointed out fairly long ago [3]. The spin-nematic state can arise from the correlation of spins at various sites, such that the symmetry with respect to time reversal for the entire system is not broken [3]. Such states were probably detected for the low-dimensional $LiCuVO_4$ magnet [4, 5]. The possibility of the realization of nematic states through the existence of spin multipole order parameters including the products of the mean spin operator projections at the same site is no less interesting. Such order is attributable to nontrivial means of the form $\langle S_{\alpha_1} S_{\alpha_2} \ldots S_{\alpha_n} \rangle$; for spin S, it makes sense to consider $n \le 2S$. Here, $n = 1$ corresponds to the dipole order parameter, i.e., the mean spin $\langle \mathbf{S} \rangle$, $n = 2$ corresponds to the quadrupole one, $n = 3$ corresponds to the octupole one, etc. An example of a spin nematic with quadrupole order for a system with spin $S = 1$ was considered in [3]. The problem of such (single-site) spin nematics is closely related to the problem of quadrupole ordering and peculiar quadrupole dynamics that has long been discussed in the literature (see, e.g., [6–14]).

The nematic order associated with the nontrivial spin multipole order parameters at a single site at $\langle \mathbf{S} \rangle = 0$ is a purely quantum phenomenon. Its description is outside the scope of the so-called spin coherent states or coherent states of the Lie group $SO(3) \sim SU(2)$ (see [13, 14]). For these states, the absolute value of the mean spin operator is always nonzero, with $|\langle \mathbf{S} \rangle| = S$ for spin S. The spin coherent states give an accurate description of the system for a Hamiltonian containing only the terms linear in spin operators at a given site, in particular, for an ordinary bilinear exchange interaction of the form $-J(\mathbf{S}_l \cdot \mathbf{S}_{l'})$. For such a Hamiltonian in the case where the initial state is an $SU(2)$-coherent state, the spin dynamics is defined by the system of Landau–Lifshitz equations for the spins [13, 14]. Therefore, for the nematic states to be realized, apart from the bilinear interaction, the Hamiltonian of a system with spin $\mathbf{S} = 1$ must also include higher (non-Heisenberg) terms. A biquadratic exchange interaction of the form $-K(\mathbf{S}_l \cdot \mathbf{S}_{l'})^2$ is possible for an isotropic system with spin $S = 1$, where S_l and $S_{l'}$ are the spin operators at neighboring sites l and l', J and K are, respectively, the Heisenberg and biquadratic exchange constants, and it is the constant K that determines the existence of a nematic state [3].

The systems with magnetic-ion spin $S = 1$ have been studied most extensively, and we will discuss their properties to the extent to which this is necessary for the purposes of our paper—the analysis of nematic states for systems with higher spins. The spin-nematic state with $S = 1$ in which the dipole spin ordering

parameter is zero, $\langle \mathbf{S} \rangle = 0$, is characterized by spontaneous rotational symmetry breaking associated with the spin quadrupole parameters $S_{ik} = \langle S_i S_k + S_k S_i \rangle$, i, $k = x, y, z$ [10, 15]. This state is invariant with respect to time reversal, but spontaneous rotational symmetry breaking associated with the quadrupole means takes place for it.

The quadrupole ellipsoid, i.e., the ellipsoid with the directions of its principal axes $\mathbf{e}_1$, $\mathbf{e}_2$, and $\mathbf{e}_3$ chosen in such a way that $\langle S_i S_k + S_k S_i \rangle = 0$ at $i \neq k$, $i, k = 1, 2, 3$ and with its semiaxes being $\langle S_1^2 \rangle$, $\langle S_2^2 \rangle$, and $\langle S_3^2 \rangle$ can be chosen as a geometric image of these means. At zero temperature $T = 0$, the spin state at each site is determined by the pure quantum states of spin $S = 1$. In the mean-field approximation, the nematic phase of a magnet with the bilinear and biquadratic exchange interactions of the nearest neighbors is stable at $J < K$ and $J > 0$. Using the states $|0\rangle$ and $|\pm 1\rangle$ with a specified spin projection onto some quantization z axis, it can be shown that the state at the site $|\psi_{SN}\rangle = |0\rangle$ corresponds to the spin-nematic phase at $T = 0$ in this approximation [3]. This is probably the simplest case of nematic order: the quadrupole ellipsoid degenerates into a flat disk, $\langle S_x^2 \rangle = \langle S_y^2 \rangle = 1$ and $\langle S_z^2 \rangle = 0$. At a temperature that is nonzero but below the critical one, $T < T_C$, $0 < \langle S_z^2 \rangle < \langle S_{x,y}^2 \rangle$; the rotational symmetry $S_{\alpha\beta}$ is restored at $T > T_C$ [8, 16, 17]. Since the direction of the quantization z axis is arbitrary due to the isotropy of spin interactions, the spin-nematic state can be described by introducing a director vector $\mathbf{n}$ directed along the rotation axis of the quadrupole ellipsoid. Clearly, the states with $\mathbf{n}$ and $-\mathbf{n}$ are indistinguishable, and $S_{\alpha\beta}$ is a quantum analog of the de Gennes order parameter that was introduced for ordinary nematic liquid crystals [18]. The question about the nematic-phase stability outside the scope of the mean-field approximation in the two-dimensional case (or in the one-dimensional case at zero temperature) is an open one (see [15, 19–21]). However, in the three-dimensional case of interest to us, there is no reason to doubt that the nematic state is stable at a finite temperature, and the mean-field approximation is suitable for its description far from the critical region [15, 19]. In the case where the exchange integral $J < 0$, states with two magnetic sublattices arise for a crystalline magnet. (The lattice is assumed to admit a breakdown into two equivalent sublattices (a bipartite lattice).) If the Heisenberg exchange exceeds the biquadratic one, then an ordinary antiferromagnetic state is realized in the magnet. In the opposite case, the situation is more interesting, and the question about the ground state becomes nontrivial, because the states with $\mathbf{n}$ and $-\mathbf{n}$ are identical. It can be shown within the mean-field approximation that an orthogonal-nematic state for which the directions of $\mathbf{n}$ are orthogonal in the two sublattices is realized in the system [19, 22, 23]. Since there exist three such directions of the vector $\mathbf{n}$, this state is treated in the one-dimensional case as a semi-ordered one [22], although the stability of the two-and three-sublattice phases within the mean-field approximation has been proven for square [23] and triangular [24, 25] lattices, respectively (see Figs. 1 and 2 in [25]). For low-dimensional systems, the question is still an open one; numerical simulations of one-dimensional systems point to states with trimerization [26, 27]. The orthogonal nematic–ferromagnet and orthogonal nematic–antiferromagnet phase transitions occurring as the parameter J/K changes are degenerate first-order phase transitions [23].

Thus, studies of the simplest spin-nematic model suggest that such systems possess a number of unusual properties. Such states have been actively studied in crystalline magnets [9, 10], including low-dimensional system [15, 19–21], over the last twenty years. An additional interest in such states related to the study of ultracold atomic gases with integer spins [28–32], especially the Bose–Einstein condensation for such gases in optical traps or lattices [28], has appeared at present. It is also significant that such condensates are characterized by a strong non-Heisenberg spin interaction necessary for the existence of nematic states [30].

Thus, the nematic states for spin $S = 1$ have been studied fairly extensively: the interactions of elementary excitations and the relaxation processes were investigated for them [19, 33–36], and nonlinear excitations, solitons, were found [37–40]. Both one-dimensional solitons [37, 38] that resemble the Lieb states of a nonideal Bose gas [41] discovered many years ago and topological two-dimensional solitons [39, 40] were obtained. These solitons also resemble the corresponding excitations in antiferromagnets, see [42–44] (in both cases, an approximate description based on the sigma-model [15] can be used), but they have unique properties near the nematic–ferromagnet transition point [40] and at this point itself [40, 45]. A phenomenological theory of the relaxation dynamics [36] and nonequilibrium thermodynamics [46, 47] of spin nematics was constructed. The inelastic scattering of neutrons was studied theoretically, and it was shown how it could be used to analyze the elementary excitations in these systems [25].

The question about the existence of nematic states in systems with spins $S > 1$ is more complex, although it has been studied for a fairly long time [19, 48, 49]. The most interesting feature of such systems is the possibility of the appearance of nontrivial correlators from an odd number of spins (for example, three) that are not invariant with respect to time reversal even at $\langle \mathbf{S} \rangle = 0$. Although the states invariant with respect to the substitution $t \longrightarrow -t$ are usually called the spin nematics [3, 48], using the term "spin nematic" for such states [19, 50] in this paper will not lead to any misunderstanding. Great interest in analyzing the

nematic states in an ultracold Bose gas with spin $S = 2$ has arisen recently [51–55].

The possibility of the existence of spin nematic states for systems with half-integer spins (the minimum half-integer spin that admits a non-Heisenberg interaction and nontrivial higher spin correlators is $S = 3/2$) is no less interesting. Such states have been studied less extensively, although this question is of great importance for investigating the physical properties of ultracold Fermi gases, with the ^{132}Cs, ^{9}Be, and ^{135}Ba gases with spin $S = 3/2$ in optical lattices in which there is one atom per cell being examples [56–59].

The nematic states of a magnet with spin $S = 3/2$ for which $\langle \mathbf{S} \rangle = 0$ and the means cubic in spin projection operators are nontrivial were obtained fairly long ago [19, 49]. For the general model of an isotropic magnet with spin 3/2 (see Eq. (1) below), a nematic state was shown [19] to exist in the immediate vicinity of the phase transition to the ferromagnetic state. Fridman et al. [50] constructed the phase diagram of an isotropic magnet with magnetic-ion spin 3/2 in the mean-field approximation and found antinematic states with two sublattices. The spin states in different sublattices differ, but they transform into one another in the case of time reversal. Thus, in addition to spontaneous rotational symmetry breaking, all nematic phases of a magnet with spin $S = 3/2$ also exhibit symmetry breaking with respect to time reversal.

The static properties of the nematic phases of a magnet with spin $S = 3/2$, in particular, the antinematic phases, give hope for the existence of nontrivial properties of collective excitations in such systems. However, the question about the dynamic properties of non-Heisenberg magnets, especially about the physical meaning of various branches in the spectrum, has been barely investigated (the spectra in [50] were discussed only to the extent to which they were important for studying the stability of phases). The goal of this paper is to investigate the spectrum of elementary excitations for an isotropic magnet with magnetic-ion spin 3/2 at various relationships between the exchange integrals and to analyze the question about the completeness of all those spin states that were found in [19, 48, 50].

2. THE MODEL, THE GROUND STATES, AND THE METHODS FOR ANALYZING THE SPECTRA

Consider an isotropic magnet with magnetic-ion spin $S = 3/2$. Generally, the Hamiltonian of a system with spin 3/2 and an isotropic exchange interaction between the nearest neighbors is

$$\mathcal{H} = -\sum_{l \neq l'} [J\mathbf{S}_l\mathbf{S}_{l'} + K(\mathbf{S}_l\mathbf{S}_{l'})^2 + L(\mathbf{S}_l\mathbf{S}_{l'})^3], \qquad (1)$$

where $\mathbf{S}_l$ is the spin operator at the lth site, J, K, and L are the exchange integrals between the nearest spins, the summation in Eq. (1) is over all pairs of nearest neighbors and each pair of nearest neighbors is taken into account once. We will restrict our analysis only to the lattices that admit a breakdown into two equivalent sublattices, for example, cubic or square. We will perform our subsequent calculations for the case of low temperatures ($T \longrightarrow 0$), because it is in this case that the quantum properties of the system are most conspicuous. The system described by Hamiltonian (1) is invariant with respect to the transformations of the rotation group $SO(3) \sim SU(2)$. It is convenient to represent Hamiltonian (1) in terms of Stevens operators, which are a set of irreducible combinations of the operators of spin components [60]. For spin 3/2, these operators are chosen in the form

$$O_2^0 = 3(S^z)^2 - S(S+1), \quad O_2^1 = [S^z, S^x]_+,$$

$$\tilde{O}_2^1 = [S^z, S^y]_+, \quad O_2^2 = \frac{1}{2}((S^+)^2 + (S^-)^2),$$

$$\tilde{O}_2^2 = \frac{1}{2i}((S^+)^2 - (S^-)^2),$$

$$O_3^0 = 5(S^z)^3 - 3S(S+1)S^z + S^z,$$

$$O_3^1 = \frac{1}{2}\left[\left(5(S^z)^2 - S(S+1) - \frac{1}{2}\right), S^x\right]_+,$$

$$\tilde{O}_3^1 = \frac{1}{2}\left[\left(5(S^z)^2 - S(S+1) - \frac{1}{2}\right), S^y\right]_+,$$

$$O_3^2 = \frac{1}{4}[S^z, ((S^+)^2 + (S^-)^2)]_+,$$

$$\tilde{O}_3^2 = \frac{1}{4i}[S^z, ((S^+)^2 - (S^-)^2)]_+,$$

$$O_3^3 = \frac{1}{2}((S^+)^3 + (S^-)^3), \quad \tilde{O}_3^3 = \frac{1}{2i}((S^+)^2 - (S^-)^3),$$

where $S^{\pm} = S_x \pm iS_y$, $[A, B]_+ = AB + BA$ denotes the anticommutator of the corresponding operators.

To within an additive constant, Hamiltonian (1) can be represented in terms of Stevens operators as a sum of bilinear combinations:

$$\mathcal{H} = -\frac{1}{2}\sum_{n \neq n'} \{\tilde{J}(\mathbf{S}_n\mathbf{S}_{n'}) + \tilde{K}O_{2n}O_{2n'} + LO_{3n}O_{3n'}\}, \quad (2)$$

where the following notation is used:

$$\tilde{K} = \frac{1}{2}K - L, \quad \tilde{J} = J - \frac{1}{2}K + \frac{587}{80}L,$$

$$O_{2n}O_{2n'} = \frac{1}{3}O_{2n}^0O_{2n'}^0 + O_{2n}^1O_{2n'}^1$$

$$+ \tilde{O}_{2n}^1\tilde{O}_{2n'}^1 + O_{2n}^2O_{2n'}^2 + \tilde{O}_{2n}^2\tilde{O}_{2n'}^2,$$

$$O_{3n}O_{3n'} = \frac{1}{10}O^0_{3n}O^0_{3n'} + \frac{3}{20}(O^1_{3n}O^1_{3n'} + \tilde{O}^1_{3n}\tilde{O}^1_{3n'})$$
$$+ \frac{3}{2}(O^2_{3n}O^2_{3n'} + \tilde{O}^2_{3n}\tilde{O}^2_{3n'}) + \frac{1}{4}(O^3_{3n}O^3_{3n'} + \tilde{O}^3_{3n}\tilde{O}^3_{3n'}).$$

This form allows us to use the representation of the coherent states for the group $SU(4)$ that were used in [50].

The system under consideration has no preferential direction, and the direction of the quantization (z) axis can be chosen arbitrarily. Separating out the mean fields associated with the mean magnetic moment (per site) $\langle S^i\rangle$ and the mean multipole fields $q^i_j = \langle O^i_j\rangle$ in Hamiltonian (2), we will obtain a single-site Hamiltonian in the form

$$\mathcal{H}_0 = -zJ\sum_i \langle S^i\rangle S^i - zK\sum_i q^i_2 O^i_2 - zL\sum_i q^i_3 O^i_3, \quad (3)$$

where z is the number of nearest neighbors.

To diagonalize Hamiltonian (3), we will use the method developed in [61] that is based on the algebra of Hubbard operators [62]. We will construct the Hubbard operators $X^{ij} \equiv |i\rangle\langle j|$, where the state vectors $|i\rangle$ comprise the standard complete set of eigenstates of the spin operator with a given z spin projection [62, 63], on the basis of eigenfunctions of the operator S^z. The relation between the spin and Hubbard operators is

$$S^+ = \sqrt{3}X^{\frac{3}{2}\frac{1}{2}} + 2X^{\frac{1}{2}-\frac{1}{2}} + \sqrt{3}X^{-\frac{1}{2}-\frac{3}{2}},$$
$$S^- = (S^+)^\dagger; \quad S^z = \frac{1}{2}(S^+S^- - S^-S^+). \quad (4)$$

Formally, the single-site Hamiltonian can be written in the representation of Hubbard operators as

$$\mathcal{H}_0 = \sum_i \varepsilon_i X^{ii} + \sum_{i\neq j} V_{ij}X^{ij}, \quad V_{ji} = (V_{ij})^*,$$

where ε_i and V_{ij} are the diagonal and off-diagonal amplitudes, respectively. As a result of the unitary transformation

$$U = U\left(\alpha_{\frac{1}{2}-\frac{1}{2}}, \alpha_{-\frac{1}{2}-\frac{3}{2}}, \alpha_{\frac{1}{2}-\frac{3}{2}}, \alpha_{\frac{3}{2}\frac{1}{2}}, \alpha_{\frac{3}{2}-\frac{1}{2}}, \alpha_{\frac{3}{2}-\frac{3}{2}}\right)$$

the single-site Hamiltonian can be brought to the diagonal form

$$\tilde{\mathcal{H}}_0 = \sum_i E_i X^{ii},$$

with the parameters of the unitary transformations being defined by the system of equations

$$\tilde{V}_{ij}\left(\alpha_{\frac{1}{2}-\frac{1}{2}}, \alpha_{-\frac{1}{2}-\frac{3}{2}}, \ldots\right) = 0.$$

The diagonalization problem is simplified considerably by the fact that all of the means, except $\langle S^z\rangle$, q^0_2, q^0_3, and q^2_3, are zero. This condition follows from the system's symmetry and can be tested directly. The single-site Hamiltonian then takes the form

$$\tilde{\mathcal{H}}_0 = -\overline{H}S^z - B^0_2O^0_2 - B^0_3O^0_3 - B^3_3O^3_3, \quad (5)$$

where the following notation is used:

$$\overline{H} = z\tilde{J}\langle S^z\rangle, \quad B^0_2 = z\frac{\tilde{K}}{3}q^0_2,$$
$$B^0_3 = z\frac{L}{10}q^0_3, \quad B^3_3 = z\frac{L}{4}q^3_3,$$
$$q^0_2 = \langle O^0_2\rangle = 3\langle (S^z)^2\rangle - \frac{15}{4}, \quad (6)$$
$$q^0_3 = \langle O^0_3\rangle = 5\langle (S^z)^3\rangle - \frac{41}{4}\langle S^z\rangle,$$
$$q^3_3 = \langle O^3_3\rangle = \frac{1}{2}(\langle (S^+)^3\rangle + \langle (S^-)^3\rangle).$$

To diagonalize Hamiltonian (5), it will suffice to make one "unitary rotation" $\alpha_{3/2,-3/2}$ that has the meaning of Bogoliubov's generalized u–v transformation. Our analysis shows that the states $|\psi(1/2)\rangle$ and $|\psi(-1/2)\rangle$ are never the "extremal" ones; the states obtained by the unitary rotation from $|3/2\rangle$ and $|-3/2\rangle$ always correspond to the minimum and maximum eigenvalues. Let us choose the states $|\psi(3/2)\rangle$ and $|\psi(-3/2)\rangle$ in such a way that the smallest and largest eigenvalues denoted by $E_{\min}$ and $E_{\max}$ correspond to them and write the complete set of states as

$$|\psi(3/2)\rangle = \cos\alpha|3/2\rangle + \sin\alpha|-3/2\rangle,$$
$$|\psi(-3/2)\rangle = -\sin\alpha|3/2\rangle + \cos\alpha|-3/2\rangle, \quad (7)$$
$$|\psi(1/2)\rangle = |1/2\rangle, \quad |\psi(-1/2)\rangle = |-1/2\rangle,$$

where the energy levels are defined by the formulas

$$E_{\min} = -3B^0_2 - \frac{3}{2}(\overline{H} + B^0_3)\cos 2\alpha - 3B^3_3\sin 2\alpha,$$
$$E_{\max} = -3B^0_2 + \frac{3}{2}(\overline{H} + B^0_3)\cos 2\alpha + 3B^3_3\sin 2\alpha, \quad (8)$$
$$E_{1/2} = 3B^0_2 - \frac{1}{2}(\overline{H} - 9B^0_3),$$
$$E_{-1/2} = 3B^0_2 + \frac{1}{2}(\overline{H} - 9B^0_3).$$

The parameter α of the generalized u–v transformation is defined by the equation

$$(\overline{H} + B^0_3)\sin 2\alpha - 2B^0_3\cos 2\alpha = 0. \quad (9)$$

The order parameters of the system can be determined at arbitrary relationships between the exchange integrals from the relation between the spin operators and the Hubbard operators constructed on the basis of eigenfunctions (7):

$$\langle S^z\rangle = \frac{3}{2}\cos 2\alpha, \quad q^0_2 = 3,$$
$$q^0_3 = \frac{3}{2}\cos 2\alpha, \quad q^3_3 = 3\sin 2\alpha. \quad (10)$$

It is easy to see that, given equalities (6) and (10), Eq. (9) can be transformed to $\Lambda_1 \sin 2\alpha \cos 2\alpha = 0$, where we designate

$$\Lambda_1 = J - \frac{K}{2} + \frac{103}{16}L. \quad (11)$$

It follows from the derived equation that in the case where the combination of exchange integrals Λ_1 does not become zero, the Hamiltonian diagonalization parameter α can take on only the values that are multiples of $\pi/4$.

Since we consider the case of low temperatures, the system's free energy (per spin) in the mean-field approximation coincides with the lowest magnetic-ion energy level $E_{\min}$. Thus, for the free energy density we obtain

$$F = -3B_2^0 - \frac{3}{2}(\bar{H} + B_3^0)\cos 2\alpha - 3B_3^3 \sin 2\alpha,$$

which, given Eqs. (6) and (10), yields

$$F = -\frac{3z}{2}\left(\frac{K}{2} - L\right) - \frac{9z}{8}\left(J - \frac{K}{2} + \frac{119}{16}L\right)\cos^2 2\alpha - \frac{9z}{8}L\sin^2 2\alpha. \quad (12)$$

By minimizing the free energy density in α, we again arrive at the expression $\Lambda_1 \sin 2\alpha \cos 2\alpha = 0$ (see (11)), i.e., only $\alpha = 0$ and $\pi/4$ can correspond to the minimum of the free energy density.

If $\Lambda_1 > 0$, then the state with $\alpha = 0$ is favorable; if $\Lambda_1 < 0$, then the state with $\alpha = \pi/2$ is favorable. At $\alpha = 0$, $\langle S^z \rangle = 3/2$, $q_2^0 = 3$, $q_3^0 = 3/2$, and $q_3^3 = 0$, corresponding to the ferromagnetic phase with the maximum possible magnetic moment at the site. Similarly, at $\alpha = \pi/2$, we obtain the same saturated state but with the opposite spin direction, $\langle S^z \rangle = -3/2$. These states are equivalent, and below we discuss only the case of $\alpha = 0$.

For the state with $\alpha = \pi/4$, $\langle S^z \rangle = 0$ and $q_3^0 = 0$. The nonzero quadrupole mean has the same value as that for the ferromagnetic phase, $q_2^0 = 3$, but, in this case, the nontrivial means $\langle (S^+)^3 \rangle$ and $\langle (S^-)^3 \rangle$ arise and $q_3^3 = 3$ (see (6)). Thus, a nematic state with zero mean magnetic moment at the site is realized at $\alpha = \pi/4$. Note that the situation here is significantly different than that for a magnet with spin 1, for which the ferromagnetic and nematic phases differed not only by the mean spin but also by the quadrupole means. As will be shown below, this difference between the static properties leads to a fundamental difference between the spectra of collective modes in these two cases.

Thus, there exist phases with a considerable difference between the parameters when $\Lambda_1 \longrightarrow \pm 0$; in particular, $\langle S^z \rangle = 3/2$ when $\Lambda_1 \longrightarrow +0$ in the ferromagnetic phase, while $\langle S^z \rangle = 0$ when $\Lambda_1 \longrightarrow -0$ in the nematic phase (see (10)). At $\Lambda_1 = J - K/2 + 103L/16 = 0$, the energies of the phases coincide. These two properties are typical of a first-order phase transition. This result coincides with what was obtained previously by different methods [19, 50], in particular, by calculating the free energy based on the coherent states of the group $SU(4)$. However, in contrast to the standard situation, the parameter α at $\Lambda_1 = 0$ is indefinite, i.e., all states with $\alpha \neq 0$ and $\pi/4$ have the same energy at this point. These two phases are also known to lose their stability as the point Λ_1 is approached: the ferromagnetic one when $\Lambda_1 \longrightarrow +0$ and the nematic one when $\Lambda_1 \longrightarrow -0$ [50]. In the language of standard models of magnets, this means the existence of a degenerate first-order transition point. Such degeneracy is usually removed by generalizing the model (usually by taking into account the higher invariants); as a result, either a finite domain of existence of the phases or a "splitting of the point" into two second-order transitions through an intermediate phase with a low symmetry (in our case, $\alpha \neq 0$ and $\pi/2$) is obtained (see [64]). However, this way is impossible for the quantum model (1), because the higher terms like $(\mathbf{S}_l \mathbf{S}_{l'})^n$ with $n > 3$ are reduced to those that have already been written out in (1). Such a behavior is atypical of a first-order phase transition and is determined by an increase in model symmetry (from the rotational symmetry $SU(2)$ to at least $SU(3)$ at $\Lambda_1 = 0$), i.e., this value corresponds to a quantum critical point (see [50, 65]). The same situation also takes place for the transitions between other phases in this model (the antiferromagnetic and antinematic ones) and is also known for an isotropic magnet with spin $S = 1$ [22, 23, 49].

The method of Green's functions for the Hubbard operators [62, 66], which is most appropriate to the study of strongly correlated systems, can be used to analyze the elementary excitations in a magnet with $S = 3/2$. In our case, this approach requires cumbersome calculations, but it admits an analytical calculation of the spectra in the entire domain of parameters of the Hamiltonian and wave vectors when using some approximations. The energy spectra of excitations for strongly correlated systems are defined by the poles of the Green's function [66–68]. Just as in [68], the reciprocal interaction radius is used as a small parameter that allows the perturbation theory to be applied.

Let us define the Matsubara Green's functions as follows [49]:

$$G^{\lambda\lambda'}(n, \tau; n', \tau') = -\langle \hat{T} \tilde{X}_n^{\lambda}(\tau) \tilde{X}_{n'}^{\tilde{\lambda}}(\tau') \rangle,$$

where $\hat{T}$ is the Wick operator, $\tilde{X}_n^{\lambda}(\tau) = \exp(\mathcal{H}\tau) X_n^{\lambda} \exp(-J)$ is the Hubbard operator in the Heisenberg representation, $\lambda = \alpha_1, \alpha_2, \ldots, \alpha_{2S+1}, M_1, M_2, \ldots, M_{2S+1}$; $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{int}}$.

Since we will perform our subsequent calculations in the mean-field approximation, we will need only the "transverse" part of the exchange Hamiltonian $\mathcal{H}_{\text{int}}$ that can be represented as

$$\mathcal{H}_{\text{int}} = -\frac{1}{2}\sum_{n,n',\lambda,\lambda'}\{\mathbf{c}(\lambda), \hat{A}_{nn'}\mathbf{c}(\lambda')\}X_n^{\lambda}X_{n'}^{\lambda'},$$

where the components of the vector $\mathbf{c}(\lambda)$ are determined from the relation between the spin and Hubbard operators, and the matrix $\hat{A}_{nn'}$ can be represented as

$$\hat{A}_{nn'} = \frac{\tilde{J}_{nn'}}{2}(2\cdot\hat{1}\oplus\hat{I})\oplus\frac{\tilde{K}_{nn'}}{2}(6\cdot\hat{1}\oplus\hat{I}\oplus\hat{I})$$

$$\oplus\frac{\tilde{L}_{nn'}}{160}(16\cdot\hat{1}\oplus 3\hat{I}\oplus 30\hat{I}\oplus 20\hat{I}),\quad \hat{I}\equiv\begin{pmatrix}0&1\\1&0\end{pmatrix}.$$

The Fourier transform of the system of equations for the Green's functions can then be written as

$$G^{\lambda\lambda'}(\mathbf{k},\omega_n) = \Sigma^{\lambda\lambda'}(\mathbf{k},\omega_n) - \frac{1}{2}\Sigma^{\lambda\lambda^1}(\mathbf{k},\omega_n)$$

$$\times\{\mathbf{c}(-\lambda_1), \hat{A}(\mathbf{k})\mathbf{c}(\lambda_2)\}G^{\lambda_2\lambda'}(\mathbf{k},\omega_n),$$

and the Fourier components $G^{\lambda\lambda'}(\mathbf{k},\omega_n)$ are nonzero only for even frequencies $\omega_n = 2\pi nT$, $n = 0, \pm1, \pm2, \ldots$, while the Larkin-irreducible graphs can be of the following types:

$$\Sigma^{\alpha\beta}(\mathbf{k},\omega_n),\ \Sigma^{\alpha M}(\mathbf{k},\omega_n),\ \Sigma^{M\alpha}(\mathbf{k},\omega_n),\ \Sigma^{MM'}(\mathbf{k},\omega_n).$$

However, the system of equations for the Green's functions is simplified considerably in the zeroth approximation in reciprocal interaction radius, because in this approximation

$$\Sigma^{\alpha\beta}(\mathbf{k},\omega_n) = \delta_{\alpha\alpha'}b(\alpha)G_0^{\alpha}(\omega_n),$$

$$\Sigma^{\alpha M}(\mathbf{k},\omega_n) = \Sigma^{M\alpha}(\mathbf{k},\omega_n) = 0,$$

$$\Sigma^{MM'}(\mathbf{k},\omega_n) = -\frac{n_{MM'}}{T}\delta_{\omega_n,0},$$

where $G_0^{\alpha}(\omega_n) = [i\omega_n + \alpha\cdot\mathbf{E}]^{-1}$ is the zeroth Green function, $b(\alpha) = \langle\alpha\cdot\mathbf{X}\rangle_0$ is the end factor, α is the root vector whose components are determined by the algebra of Hubbard operators [62, 65]. Thus, in the zeroth approximation in reciprocal interaction radius, the dispersion equation is

$$\det\|\delta_{ij} + x_{ij}\| = 0;\quad i,j = 1, 2, \ldots, 15,$$

where

$$x_{ij} = G_0^{\alpha}(\omega_n)b(\alpha)c_{ij}(\alpha),\quad c_{ij}(\alpha,\beta) = a_{ik}(\alpha,\beta)A_{kj};$$

$$a_{ik}(\alpha,\beta) = c_i(\alpha)c_k(-\beta).$$

Since the technique of Hubbard operators allows the single-site correlators to be accurately taken into account, the dispersion equation is valid at arbitrary relationships between the exchange integrals, i.e., in various phase states. The results of our analysis of the spectra in various phases are presented below in Sections 3–6.

After discussing the general approaches and formulating the problem, we will turn to a specific analysis of the spin states and the spectra of elementary excitations in the various phases of a magnet described by Hamiltonian (1).

3. THE FERROMAGNETIC PHASE

Consider a system in the ferromagnetic phase. As has been pointed out above, the solution of Eq. (9) $\alpha = 0$ corresponds to this phase state. The spin means characterizing the ferromagnetic phase are

$$\langle S^z\rangle = \frac{3}{2},\quad q_2^0 = 3\langle(S^z)^2\rangle - S(S+1) = 3,$$

$$q_2^2 = \langle(S^x)^2\rangle - \langle(S^y)^2\rangle = 0,$$

$$q_3^0 = 5\langle(S^z)^3\rangle - [3S(S+1)-1]\langle S^z\rangle = \frac{3}{2},$$

$$q_3^3 = \frac{1}{2}(\langle(S^+)^3\rangle + \langle(S^-)^3\rangle) = 0.$$

$E_{\min}$ is the lowest energy level in the ferromagnetic state, while the ground-state wave function is the state with the maximum spin projection, $|\psi(3/2)\rangle = |3/2\rangle$. In addition, the quadrupole means in this state are defined by the expressions

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

Thus, the geometric image of the quadrupole means in the ferromagnetic phase (quadrupole ellipsoid) is an ellipsoid of revolution in spin space. The symmetry of this ellipsoid is determined by the direction of the magnetic moment that coincides with the direction of the principal axis of the ellipsoid. In contrast to a magnet with spin $S = 1$, the shape of the quadrupole ellipsoid is the same for the ferromagnetic and nematic phases. In addition, the nontrivial octupole mean q_3^3 is zero, while q_3^0 is a trivial constant and does not affect the system's dynamics. Consequently, in the ferromagnetic phase, just as for a system with spin $S = 1$, the symmetry of the quadrupole means is completely determined by the symmetry of the magnetic moment. The presence of "nondipolar" means does not manifest itself in the symmetry of the ground state of the ferromagnetic phase.

However, the possibility of the existence of such nondipolar means changes fundamentally the system's dynamics compared to the case of $S = 1$. Let us determine the spectra of excitations in the ferromagnetic phase using the general dispersion equation. In the low-temperature approximation, there will exist three mag-

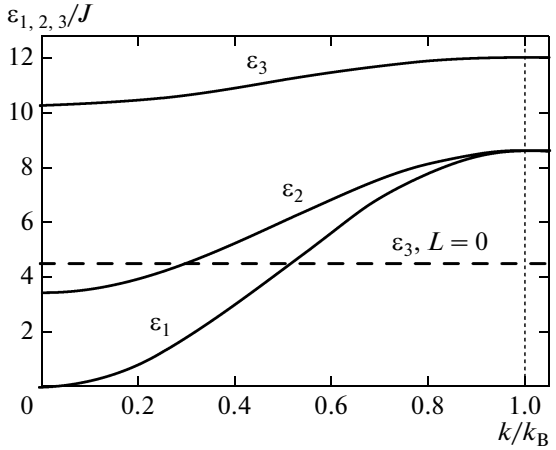

Fig. 1. Dispersion laws for elementary excitations in the ferromagnetic phase, the frequencies are normalized to J, k_B defines the boundary of the Brillouin zone. Here and below, the construction was made for the one-dimensional case (we chose $z = 2$) for simplicity, the generalization to the case of an arbitrary dimension for a symmetric direction $\mathbf{k}$ presents no difficulty. The solid lines indicate the spectra of elementary excitations at the center of the stability region for the ferromagnetic phase (we chose $K = J$ and $L = J/10$). The dashed line indicates the absence of any dispersion of the mode with $\varepsilon_3(k)$ at $L = 0$.

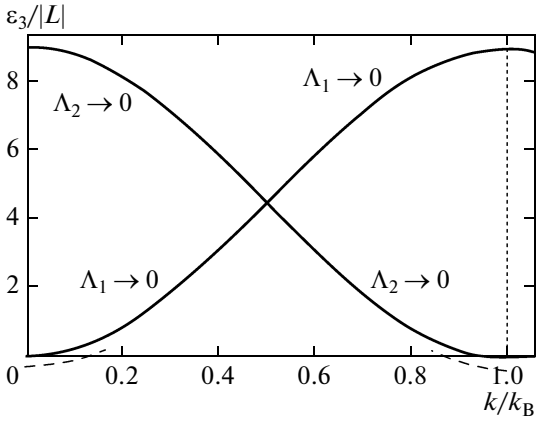

Fig. 2. Dispersion law for the "octupole" branch of elementary excitations $\varepsilon_3(k)$ in the ferromagnetic phase near the phase transitions to the nematic ($\Lambda_1 \longrightarrow 0$) and antinematic ($\Lambda_2 \longrightarrow 0$) phases. For the case under consideration, it is convenient to normalize the frequencies to $|L|$ (L is positive when $\Lambda_1 \longrightarrow 0$ and negative when $\Lambda_2 \longrightarrow 0$). The segments of the thin dashed lines at $k \sim 0$ and in the corner of the Brillouin zone $k \sim \pi/a$ schematically represent the dependence $\varepsilon_3^2(k)$ at $\Lambda_1 < 0$, $L > 0$ and $\Lambda_2 < 0$, $L < 0$, corresponding to the phase instability region.

non branches of excitations [6] in the system whose spectra in the nearest-neighbor approximation are

$$\varepsilon_1(k) = \frac{3z}{4}\left[J - J_k + \frac{3}{2}(K - K_k) + \frac{63}{16}(L - L_k)\right],$$

$$\varepsilon_2(k) = \frac{3z}{2}\left[K - K_k - \frac{5}{4}(L - L_k) + J - \frac{1}{2}K + \frac{103}{16}L\right], \quad (13)$$

$$\varepsilon_3(k) = \frac{9z}{4}\left[(L - L_k) + J - \frac{K}{2} + \frac{103}{16}L\right].$$

Here and below, $J_k = JC(\mathbf{k})$, $K_k = KC(\mathbf{k})$, and $L_k = LC(\mathbf{k})$ define the Fourier components of the corresponding exchange integrals,

$$zC(\mathbf{k}) = \sum_{\mathbf{a}} \cos(\mathbf{a} \cdot \mathbf{k}),$$

the summation is over all z vectors of the nearest neighbors $\mathbf{a}$, and $\mathbf{k}$ is a dimensionless wave (to be more precise, quasi-wave) vector. For our estimates of a hypercubic lattice, $C(\mathbf{k}) = 1$ when $\mathbf{k} \longrightarrow 0$ and $C(\mathbf{k}) = -1$ at the edge of the Brillouin zone (this value is $k = k_B = \pi$ in the one-dimensional model and the boundary of the zone in a direction of type (111) in the three-dimensional model), $C(\mathbf{k}) = 1/3$ and $-1/3$ at the boundary of the zone in the (100) and (110) directions, respectively. Note a common property of the spectra: the frequencies $\varepsilon_1(k)$ and $\varepsilon_2(k)$ at all parameters of the problem coincide at the edge of the Brillouin zone, where $C(\mathbf{k}) = -1$ (see Fig. 1).

It follows from expression (13) that the branch ε_1 is a gapless Goldstone mode with a parabolic dispersion law at small $\mathbf{k}$ typical for an isotropic ferromagnet. Our analysis shows that the "transverse" spin density oscillations in this mode are associated with the rotations of the direction of the principal axis of the quadrupole ellipsoid.

Let us now discuss the remaining modes with the frequencies ε_2 and ε_3. It is easy to see that in the case where the bicubic interaction constant is zero ($L = 0$), only two branches of excitations, ε_1 and ε_2, remain the well-determined modes, while the frequency ε_3 becomes a purely local state, $\varepsilon_3(k) \longrightarrow 9(2J - K)/8$ when $L \longrightarrow 0$. Note that for a magnet with $S = 1$ at $K = 0$, when the specificity of a non-Heisenberg magnet disappears (in particular, the nematic phase is absent), the situation is analogous: one of the modes loses its dispersion and becomes a purely local state. In the limiting case of $L = 0$, the form of the branches ε_1 and ε_2 is the same as that of the two collective modes in the spectrum for a magnet with $S = 1$ (cf. (13) and the formulas in [15, 22, 37]). All of this leads us to conclude that the physical meaning of the mode ε_2 is the same as that for a ferromagnet with spin $S = 1$, i.e., the mode with $\varepsilon = \varepsilon_2$ describes the "longitudinal" spin dynamics [37, 69]. This mode includes the longitudinal oscillations of the magnitude of the magnetization vector whose direction remains parallel to the principal axis

of the quadrupole moment ellipsoid (for more details, see [69] and Fig. 2 in it). On the other hand, the mode ε_3 is determined by the specificity of the dynamics of a magnet with spin $S = 3/2$ that is attributable to the nontrivial octupole means.

Let us now consider the stability of the ferromagnetic phase to arbitrary perturbations that correspond to spectra (13). As is easy to see from the form of the elementary excitation spectra in the ferromagnetic phase, the magnon branches $\varepsilon_{2,3}$ at $k = 0$ have an energy gap proportional to

$$\Lambda_1 = J - \frac{1}{2}K + \frac{103}{16}L.$$

Hence it follows that the condition for the stability to homogeneous perturbations is $\Lambda_1 > 0$, which coincides with the well-known result [19]. This result is also obtained by analyzing the free energy written out above with respect to a variation of the parameter α. Thus, the line $\Lambda_1 = 0$ is the line of the phase transition between the ferromagnetic and nematic phases.

Our analysis of the complete dispersion laws (13) shows that their extrema are reached at symmetric points of the Brillouin zone and the loss of stability (frequency sign reversal) is associated with the behavior of the spectrum either at $\mathbf{k} = 0$ or at the edge of the Brillouin zone, where $C(\mathbf{k}) = -1$. At this point, the frequency of the mode ε_3 is

$$\varepsilon_{3,\,\text{edge}} = J - \frac{1}{2}K + \frac{135}{16}L = \Lambda_2, \qquad (14)$$

$\varepsilon_{3,\,\text{edge}} \longrightarrow 0$ when $\Lambda_2 \longrightarrow 0$, with the other frequencies being positive. Thus, the stability condition includes the inequality $\Lambda_2 > 0$. Below, we will make sure that the line $\Lambda_2 = 0$ is the line of the phase transition from the ferromagnetic phase to the antinematic one.

Thus, the stability region for the ferromagnetic state is defined by the inequalities

$$\Lambda_1 > 0, \quad \Lambda_2 > 0. \qquad (15)$$

In Fig. 1, the collective excitation energies ε_1, ε_2, and ε_3 for the system being investigated are plotted against the wave vector in the ferromagnetic phase far from the boundaries of the stability region for the ferromagnetic phase. It can be seen that the spectrum ε_1 determines the Goldstone mode, while the branches ε_2 and ε_3 are the activation ones.

The stability of the system far from the phase transition lines is described by the "octupole" mode ε_3. The latter softens near the lines of the loss of ferromagnetic phase stability, namely the lines of the phase transition to the nematic phase $\Lambda_1 = 0$ and to the antinematic phase $\Lambda_2 = 0$, corresponding to a softening of the spectrum at the center of the Brillouin zone $k \longrightarrow 0$ and in the corner of the Brillouin zone (at $C(\mathbf{k}) = -1$) (see Fig. 2).

4. THE NEMATIC PHASE

Let us now investigate the dynamic properties of the system in the nematic phase. As has already been pointed out above, the $\alpha = \pi/4$ in this case. As follows from Eq. (10), the order parameters in this state are

$$\langle S^z \rangle = 0, \quad q_2^0 = 3, \quad q_3^0 = 0, \quad q_3^3 = 3.$$

$E_{\min}$ is also the lowest energy level in the nematic state, and the ground-state wave function is

$$|\psi\left(\frac{3}{2}\right)\rangle = \frac{1}{\sqrt{2}}\left(|\frac{3}{2}\rangle + |-\frac{3}{2}\rangle\right).$$

A simple calculation shows that the quadrupole means define some anisotropy of the system,

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

and an ellipsoid of revolution is the geometric image of the quadrupole means for the nematic state in spin space. The shape of this ellipsoid is the same as that in the ferromagnetic phase. Within the framework of the general phenomenological approach [2], one might expect the existence of two degenerate non-activation modes for the collective oscillations of the system of such ellipsoids. In fact, however, the symmetry of the nematic state does not contain the C_∞ axis; it is lower than that determined by the quadrupole ellipsoid. Indeed, in contrast to the ferromagnetic state, the cubic means of the form

$$\langle (S^x \cos\chi + S^y \sin\chi)^3 \rangle = \frac{3}{4}\cos 3\chi,$$

are nonzero in the nematic phase, while the mean $\langle S_z^3 \rangle = 0$. The angle χ actually defines the rotation of the spin system around the z axis; therefore, the nonzero cubic means define the third-order axis (and a combined transformation, i.e., rotation through $\pi/3$ in combination with time reversal). In this case, one might expect the appearance of yet another mode associated with the rotation of the "octupole triangle." Note that the same symmetry elements are present for an antiferromagnet with three magnetic sublattices located in a plane and making angles of $2\pi/3$, and the behavior of these significantly different systems must be similar from a macroscopic point of view [48, 49].

Let us determine the spectra of elementary excitations in the nematic phase. Note that two excited energy levels degenerate in this state, $E_{\frac{1}{2}} = E_{-\frac{1}{2}}$. As a result, three branches of excitations with the following frequencies are realized in the system:

$$\varepsilon_{1,2}(k) = \frac{3z}{2\sqrt{2}}\sqrt{\left[K - K_k - \frac{5}{4}(L - L_k)\right]\left[K - \frac{5}{4}L - \frac{1}{2}\left(J_k + \frac{3K_k}{2} + \frac{63}{16}L_k\right)\right]},$$
$$\varepsilon_3(k) = \frac{9z}{4}\sqrt{(L - L_k)\left(L - J_k + \frac{K_k}{2} - \frac{119}{16}L_k\right)}. \quad (16)$$

The dispersion laws for the first two modes coincide; their frequencies remain finite at $L = 0$. It follows from expression (16) that the frequency of the "octupole" branch $\varepsilon_3 = 0$ at $L = 0$, while for the modes with $\varepsilon = \varepsilon_{1,2}(k)$ in the limit $L \longrightarrow 0$ we obtain

$$\varepsilon_{1,2}(k) = (3z/2)\sqrt{(K - K_k)\{2K - (J_k + 3K_k/2)\}},$$

i.e., their spectrum resembles the spectrum of elementary excitations in the nematic phase of a magnet with $S = 1$ [22, 23]. Consequently, the branches $\varepsilon_{1,2}$ describe the oscillations of the quadrupole ellipsoid, which determines the double degeneracy of these modes. Thus, the main specificity of the system with $S = 3/2$ is determined by the branch ε_3 related to the exchange integral L and attributable to the dynamics of the octupole order parameters q_3^3. It is associated with the rotation of the "octupole triangle" around the z axis (see above).

Figure 3 shows the spectra of elementary excitations in the nematic phase $\varepsilon_{1,2}$ and ε_3 far from the boundaries of the phase stability region as well as the spectrum of "quadrupole" modes near the transition to the ferromagnetic phase (when $\Lambda_1 \longrightarrow 0$). It can be seen that the modes $\varepsilon_{1,2}(k)$ soften under the condition $\Lambda_1 \longrightarrow 0$ and determine the nematic phase instability at $\Lambda_1 > 0$. However, the complete conditions for the loss of nematic phase stability can be obtained only by investigating the "octupole" branch $\varepsilon_3(k)$. Indeed, the dispersion laws for all modes at the center of the Brillouin zone are linear, $\varepsilon_{1,2,3} \longrightarrow c_{1,2,3}k$ at $k = 0$. The speed of the octupole mode c_3 is proportional to $\sqrt{-\Lambda_1}$ (see Fig. 4) (the speed $c_1 = c_2$ also possesses the same property; see Fig. 3). Hence follows the necessary condition for the nematic phase stability $\Lambda_1 < 0$. In addition, it is easy to show that at the corner point of the Brillouin zone (at $C(\mathbf{k}) = -1$),

$$\varepsilon_{3,\,\mathrm{edge}} = \frac{9z}{4}\sqrt{L\left(J - \frac{K}{2} + \frac{136}{16}K\right)}, \quad (17)$$

i.e., the branch ε_3 is stable at $L\Lambda_2 > 0$ and softens near the line of the transition to the antiferromagnetic phase, $\Lambda_2 = 0$ (see Fig. 4). Taking into account the

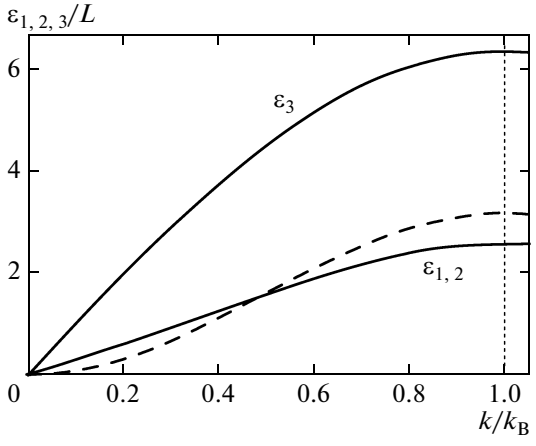

Fig. 3. Dispersion law for elementary excitations in the nematic phase (in units of L; recall that this phase is stable only at $L > 0$). The spectra of elementary excitations at the center of the stability region for the nematic phase under the condition $\Lambda_1 + \Lambda_2 = 0$, i.e., $\Lambda_1 = -L$ and $\Lambda_2 = L$, are indicated by the solid lines. For definiteness, we chose $K = 2L$ when constructing the functions $\varepsilon_{1,2}(k)$. The dashed line indicates the spectra of the modes $\varepsilon_{1,2}(k)$ near the nematic–ferromagnet phase transition (when $\Lambda_1 \longrightarrow 0$).

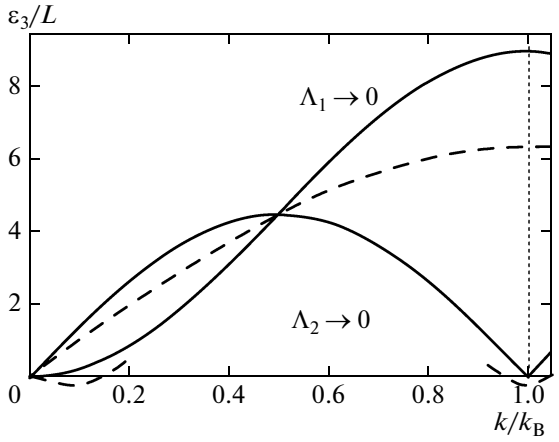

Fig. 4. Dispersion law for the "octupole" branch of elementary excitations $\varepsilon_3(k)$ in the nematic phase near the phase transitions to the ferromagnetic ($\Lambda_1 \longrightarrow 0$) and antiferromagnetic ($\Lambda_2 \longrightarrow 0$) phases in units of L (in the stability region $L > 0$). The segments of the thin dashed lines at $k \sim 0$ and in the corner of the Brillouin zone $k \sim \pi/a$ schematically represent the dependence $\varepsilon_3^2(k)$ at $\Lambda_1 > 0$ and $\Lambda_2 < 0$; the negative ε_3^2 correspond to the nematic phase instability. The dashed line is shown for comparison at the same parameters as those in Fig. 3 and defines the spectrum far from the critical lines.

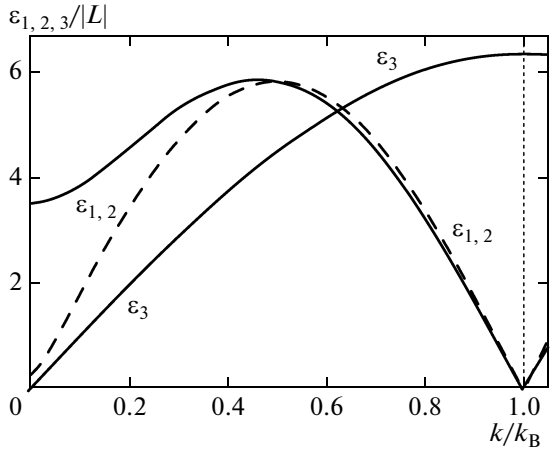

Fig. 5. Dispersion laws for elementary excitations in the antinematic phase (in units of $|L|$, the phase is stable only at $L < 0$) constructed in the extended zone scheme at the center of the region of existence of the antinematic phase, i.e., under the condition $\Lambda_1 + \Lambda_2 = 0$, $\Lambda_1 = -L = |L|$ and $\Lambda_2 = L = -|L|$ (solid lines). For definiteness, we chose $K = 0$ when constructing the functions $\varepsilon_{1,2}(k)$. The dashed line represents the dispersion law for the modes with $\varepsilon_{1,2}(k)$ near the critical line $\Lambda_1 = 0$.

condition written out above, we find that the nematic phase stability region is defined by the inequalities

$$\Lambda_1 < 0, \quad \Lambda_2 > 0, \tag{18}$$

which can hold only at $L > 0$.

5. THE ANTINEMATIC AND ANTIFERROMAGNETIC PHASES

The question arises as to what ordering takes place outside the stability regions for the two phases described above. In a Heisenberg magnet with only one bilinear exchange interaction at a negative exchange integral $J < 0$, there exists antiferromagnetic order associated with the appearance of two sublattices. The antiferromagnetic state also takes place for non-Heisenberg magnets with spins $S = 1$ and $3/2$. The possibility of the existence of an antinematic spin state that is also associated with the formation of two different sublattices was also pointed out for a system with spin $S = 3/2$. Thus, one might expect all states for the general model (1) to be exhausted by either simple one-sublattice or two-sublattice structures in the distribution of local spin states. The realization of both antiferromagnetic and antinematic orderings [50] is possible in the system we investigate.

The order parameters in the antinematic phase are

$$\langle S_1^z \rangle = 0, \quad q^0_{2(1)} = 3, \quad q^0_{3(1)} = 0, \quad q^3_{3(1)} = 3,$$

$$\langle S_2^z \rangle = 0, \quad q^0_{2(2)} = 3, \quad q^0_{3(2)} = 0, \quad q^3_{3(2)} = -3,$$

while the parameters of the generalized u–v transformation are $\alpha_1 = \pi/4$ and $\alpha_2 = -\pi/4$ for the first and second sublattices, respectively. The ground-state wave functions for the first and second sublattices in the antinematic phase are, respectively,

$$|\psi_1\rangle = \frac{1}{\sqrt{2}}(|3/2\rangle + |-3/2\rangle),$$
$$|\psi_2\rangle = \frac{1}{\sqrt{2}}(|3/2\rangle - |-3/2\rangle). \tag{19}$$

Naturally, the form of the quadrupole means for each sublattice is the same as that in the nematic phase. However, the cubic correlators for the first and second sublattices differ by the sign:

$$\langle (S_i^x \cos\chi + S_i^y \sin\chi)^3 \rangle = \pm\frac{3}{4}\cos 3\chi, \quad \langle (S_i^z)^2 \rangle = 0,$$

where $i = 1, 2$ are the sublattice numbers. The angle χ defines the rotation around the z axis in spin space. Thus, the third-order axes for the spin states of the sublattices coincide, while the cubic correlators defining a planar structure transform into one another after rotation through the angle $\chi = \pi/3$, which is equivalent to time reversal.

Let us investigate the spectra of elementary excitations in the antinematic phase. To simplify our mathematical calculations when calculating the spectrum for one of the sublattices (to be specific, for the second one), we will make a rotation around the z axis,

$$U(\varphi) = \prod_n \exp(i\varphi S_n^z) \tag{20}$$

through the angle $\varphi = \pi$ in spin space. This corresponds to an operator transformation of the form

$$S^x_{n_2} \longrightarrow \tilde{S}^x_{n_2} = -S^x_{n_2}, \quad S^y_{n_2} \longrightarrow \tilde{S}^y_{n_2} = -S^y_{n_2},$$
$$S^z_{n_2} \longrightarrow \tilde{S}^z_{n_2} = S^z_{n_2},$$

corresponding to the spin structure of the antinematic sublattices.

The single-site Hamiltonian in terms of the new operators $\tilde{S}^i_{n_2}$ has a form similar to expression (5) with the substitution $B^3_3 \longrightarrow -B^3_3$. Our analysis shows that for the calculations of the elementary excitation spectra, the Hamiltonian can be considered in terms of both old, $\tilde{S}^i_{n_1}$, and new, $\tilde{S}^i_{n_2}$, operators using the same quasi-momentum for both operators. In fact, this means using the extended zone scheme. In the case under consideration, it turns out that this representation is fairly convenient and clear, because the sublattices are equivalent. For this reason, no singularities arise at the boundary of the "magnetic" Brillouin zone, and the "magnetic" Brillouin zone does not manifest itself at all for the spectra of all modes in the antinematic phase (see Fig. 6 below). For the problems of lattice dynamics, such a situation arises if the crystal consists of different particles that, however, have the same mass and interaction constants.

Just as in the nematic phase, two excited energy levels degenerate in this state, $E_{\frac{1}{2}} = E_{-\frac{1}{2}}$, and, as a result, the two branches of excitations coincide. As a result, the spectra are defined by the expressions

$$\varepsilon_{1,2}(k) = \frac{3z}{2\sqrt{2}}\sqrt{\left[K + K_k - \frac{11}{4}(L + L_k)\right]\left[K - \frac{11}{4}L + \frac{1}{2}\left(J_k + \frac{5}{2}K_k + \frac{191}{16}L_k\right)\right]},$$
$$\varepsilon_3(k) = \frac{9z}{4}\sqrt{(L - L_k)\left(L + J_k - \frac{1}{2}K_k - \frac{119}{16}L_k\right)}. \quad (21)$$

Note that the energy of the branch $\varepsilon = \varepsilon_3(k)$, which is related to the exchange integral L and is determined by the dynamics of the octupole parameters q_3^3, is obtained from the formula for the nematic phase by changing the sign of the second parenthesis under the root. Therefore, a peculiar behavior of this branch is easy to see from the graphs for $\varepsilon = \varepsilon_3(k)$ for the nematic phase presented in Fig. 4 if the indices $\Lambda_1 \longrightarrow 0$ and $\Lambda_2 \longrightarrow 0$ are interchanged on them. Clearly, just as in all the previous examples, the system's stability for the antinematic phase is determined precisely by this branch. Additionally, the antinematic phase instability near the critical line $\Lambda_1 = 0$ is also seen from the dependence $\varepsilon_{1,2}(k)$ (see Fig. 5).

Analysis of the behavior of the spectra allows the stability region for the antinematic phase to be determined in the form

$$\Lambda_1 > 0, \quad \Lambda_2 < 0, \quad (22)$$

which can hold only at $L < 0$ (recall that the nematic phase can exist only at $L > 0$). In the model with $L = 0$, both nematic phases are absent (to be more precise, they exist only on the line $\Lambda_1 = \Lambda_2 = 0$). If this higher invariant is disregarded, then $\Lambda_1 = \Lambda_2$ and no phases with tensor order parameters are realized.

Note that the use of the extended zone scheme manifests itself in the fact that the modes $\varepsilon_{1,2}(k)$ and $\varepsilon_3(k)$ have a gapless behavior at different points of the Brillouin zone (see Fig. 5). Previously, such a behavior was established for the spectra of elementary excitations in the orthogonal-nematic phase of a magnet with spin $S = 1$ [23]. Clearly, when passing to the reduced zone scheme natural for a two-sublattice system, the standard form of the spectrum is restored (see Fig. 6).

Passing to the reduced "magnetic" Brillouin zone constructed for one sublattice is a simple geometric problem; as a result, the number of branches doubles. As a consequence, the Goldstone behavior is restored for both types of collective modes, and analogs of "acoustic" and "optical" collective modes appear for both curves. Here, the following important question arises: should we pass to the reduced magnetic Brillouin zone in this case? In other words, does it make sense to consider the oscillations in the reduced Brillouin zone and to talk about the existence of acoustic and optical branches and resonances associated with the latter at small wave vectors?

In the language of ordinary lattice dynamics, this question arises for a crystal consisting of particles that have the same mass and interaction constants but differ physically by the value of some parameter. The answer to this question is as follows. Clearly, if the particles in the crystal differ by a particular physical property, then there exists some field that acts differently on these particles of two types (if no such field exists in principle, then it makes no sense to talk about the difference between the particles). The action of a spatially uniform variable field (waves with zero wave vector and a finite frequency) on this system will leads to a different action on the particles from different sub-

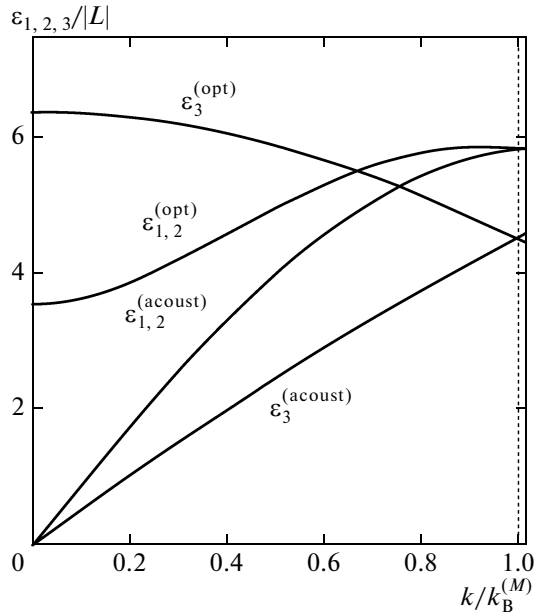

Fig. 6. The same dispersion laws at the center of the region of existence of the antinematic phase as those in Fig. 5 but constructed according to the reduced zone scheme, $k_B^{(M)}$ is the edge of the magnetic Brillouin zone, $k_B^{(M)} = \pi/2a$ in the one-dimensional case. The designations $\varepsilon_{1,2}^{(acoust)}$ and $\varepsilon_{1,2}^{(opt)}$, $\varepsilon_3^{(acoust)}$ and $\varepsilon_3^{(opt)}$ are used for clarity.

lattices, and, as a result, a linear resonant excitation of the normal mode in which these particles of two types oscillate in antiphase is possible.

Such a behavior is well known for optical phonons and some types of collective modes in magnetic superlattices [70]; it also takes place in the case of an antinematic. The differences between the magnetic states of the sublattices in an antinematic are not too significant to give rise to a gap at the boundary of the Brillouin zone of the main lattice. However, the optical methods for analyzing the nematic phases proposed in [50] lead to different effects for different sublattices. Antiphase oscillations corresponding to a finite frequency at $k = 0$ in Fig. 6 will then be excited in the case of a spatially uniform action on the system.

Let us now turn to analyzing the antiferromagnetic state for which the spins in the two sublattices are antiparallel and collinear to the z axis. Using expressions (10), it is easy to find that the order parameters of the sublattices in the antiferromagnetic phase are

$$\langle S_1^z\rangle = \frac{3}{2}, \quad q^0_{2(1)} = 3, \quad q^0_{3(1)} = \frac{3}{2}, \quad q^3_{3(1)} = 0,$$

$$\langle S_2^z\rangle = -\frac{3}{2}, \quad q^0_{2(2)} = 3, \quad q^0_{3(2)} = -\frac{3}{2}, \quad q^3_{3(2)} = 0,$$

while the parameters of the generalized u–v transformation for the first and second sublattices are $\alpha_1 = 0$ and $\alpha_2 = \pi/2$, respectively. The ground-state wave functions for the sublattices in this state are, respectively,

$$\langle \psi_1\rangle = |\frac{3}{2}\rangle \quad \text{and} \quad \langle \psi_2\rangle = |-\frac{3}{2}\rangle.$$

Just as for the antinematic state, it is convenient to make a rotation $U(\varphi) = \prod_n \exp(i\varphi S_n^x)$ through the angle $\varphi = \pi$ in spin space for one of the sublattices (to be specific, the second one). Then,

$$S^x_{n_2} \longrightarrow \tilde{S}^x_{n_2} = S^x_{n_2}, \quad S^y_{n_2} \longrightarrow \tilde{S}^y_{n_2} = -S^y_{n_2},$$

$$S^z_{n_2} \longrightarrow \tilde{S}^z_{n_2} = -S^z_{n_2},$$

and a "uniform" distribution of the mean values of the operators $\tilde{S}^i_{n_2}$ corresponds to the ground state. The single-site Hamiltonian is derived from Eq. (5) by the substitution

$$\overline{H} \longrightarrow -\overline{H}, \quad B^0_3 \longrightarrow -B^0_3$$

for the spins of the second sublattice. However, the modification of the Hamiltonian for the problem in terms of the new operators $\tilde{S}^i_{n_2}$ is more significant than that for the case of an antinematic considered above. Since the signs of the effective field $\overline{H}$ for the two sublattices differ, their dynamics is not equivalent (this property takes place for "standard" antiferromagnets), and the reduced zone representation arises naturally. A simple but cumbersome calculation of the spectra leads to the expressions

$$\varepsilon_1(k) = \frac{3z}{4}\sqrt{\left(\frac{5}{2}K - J - \frac{191}{16}L\right)^2 - \left(\frac{5}{2}K_k - J_k - \frac{191}{16}L_k\right)^2},$$

$$\varepsilon_2(k) = \frac{3z}{2}\sqrt{\left(\frac{3}{2}K - J - \frac{147}{16}L\right)^2 - \left(K_k - \frac{11}{4}L_k\right)^2}, \tag{23}$$

$$\varepsilon_3(k) = \frac{9z}{4}\sqrt{\left(\frac{K}{2} - J - \frac{119}{16}L\right)^2 - (L_k)^2}.$$

Note that only the squares of the Fourier transforms of the exchange constants enter into all these expressions, which formally determines a shorter period in the dependences $\varepsilon_{1,2,3} = \varepsilon_{1,2,3}(k)$. The dispersion laws for elementary excitations in the antiferromagnetic phase are presented in Fig. 7. The solid lines $\varepsilon_{1,2,3}$ describe the spectra at the "center" of the phase stability region: the dashed line describes the softening of the mode $\varepsilon = \varepsilon_2(k)$ near the phase transition to the nematic phase ($\Lambda_1 \longrightarrow 0$).

In the applied reduced zone scheme, the apparent number of curves defining the branches in the spectrum is three, just as for a single sublattice. This is because each curve corresponds to two degenerate branches of spin excitations. On the whole, this picture is typical for magnons in a standard Heisenberg antiferromagnet without an external field. Note that the standard magnon branch with the Goldstone behavior when $k \longrightarrow 0$ shows no instabilities, reflecting the fact that the antiferromagnetic phase, just as the ferromagnetic one, loses its stability according to the "multipole" scenario. As regards the branch $\varepsilon_2(k)$, as in the case of a ferromagnet, it is associated with the oscillations of the quadrupole ellipsoid. It is this branch that softens near the phase transition to the antinematic phase.

The branch of excitations with $\varepsilon_3(k)$ describes the coupled oscillations of the octupole parameters q^3_3 for the sublattice spins whose equilibrium value in the antiferromagnetic phase is zero, just as in the ferromagnetic one. Its dispersion is determined only by the quantity L, just as for other phases. Note that its dispersion is very weak at sufficiently small $L \ll J$, K and

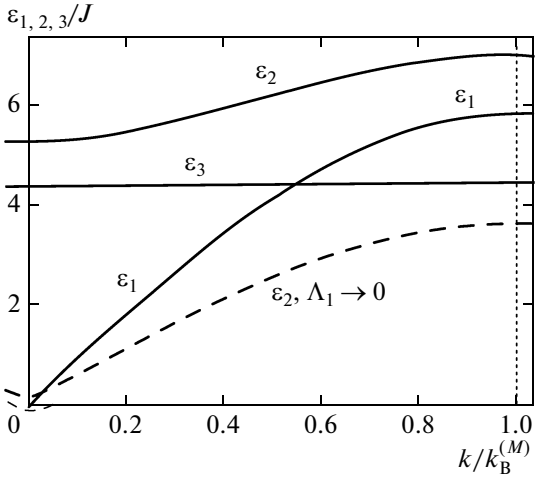

Fig. 7. Dispersion laws for elementary excitations in the antiferromagnetic phase in the reduced zone scheme, $k_B^{(M)}$. The solid lines indicate the spectra of elementary excitations in the stability region for this phase ($K = J > 0$ and $L = -J/5$ were chosen). The dashed line indicates the spectrum of the quadrupole branch $\varepsilon_2(k)$ near the antiferromagnet–antinematic phase transition (for $\Lambda_1 \longrightarrow 0$ and $\Lambda_1 < 0$). Here and in the next figure, the segment of the thin dashed line at the center of the Brillouin zone schematically represents this dependence in the phase instability region (for $\Lambda_1 \longrightarrow 0$ and $\Lambda_1 > 0$).

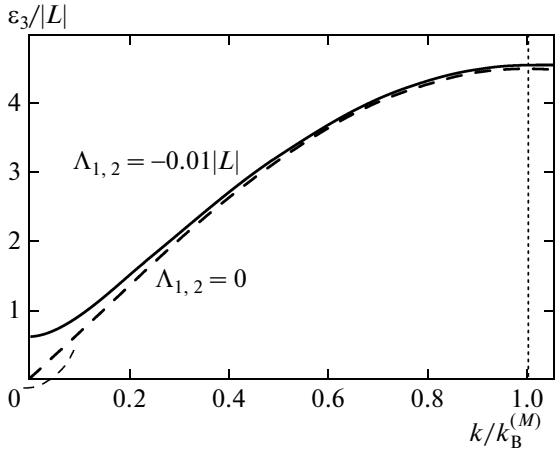

Fig. 8. Dispersion law for the branch $\varepsilon_3(k)$ (in units of $|L|$) in the antiferromagnetic phase near the phase transitions (solid lines) and on the lines themselves at $\Lambda_1 = 0$ or $\Lambda_2 = 0$ (dashed lines).

far from the lines of stability loss, because it is quadratic in small parameters L/J and L/K. However, the linear dependence on L is restored near the lines of phase instability, with the curves $\varepsilon_3(k)$ coinciding (in the reduced zone scheme) at identical $|\Lambda_1|$ and $|\Lambda_2|$ (see Fig. 8). This branch determines the antiferromagnetic phase instability at both $\Lambda_1 > 0$ and $\Lambda_2 > 0$.

Thus, our analysis of the spectra confirms that the loss of antiferromagnetic phase stability is associated with the center of the reduced (magnetic) Brillouin zone $k = 0$. The stability condition for the antiferromagnetic phase can be written as two inequalities:

$$\Lambda_1 < 0, \quad \Lambda_2 < 0. \tag{24}$$

6. DISCUSSION OF RESULTS

We analyzed the free energy density and the spectra of elementary excitations for a non-Heisenberg isotropic magnet with $S = 3/2$ and a dimension $d \geq 2$, which allows the mean-field approximation to be applied, and a lattice that admits a breakdown into two equivalent sublattices. The realization of only four phase states is possible in the system under consideration at various relationships between the material parameters. Apart from the states with a dominant Heisenberg exchange characterized by a vector order parameter (the ferro- and antiferromagnetic phases), the realization of states with tensor order parameters is possible in a magnet with $S = 3/2$ and predominant higher exchange integrals (the nematic and antinematic phases). The means cubic in spin operator components, primarily the octupole order parameter q_3^3 containing means like $\langle (S_x \pm iS_y)^3 \rangle$, play a major role in the properties of these phases. The octupole order formation is associated with allowance for the bicubic exchange interaction.

Using the results obtained, the phase diagram of a non-Heisenberg magnet with $S = 3/2$ can be constructed for various relationships between the exchange integrals. Since the general energy scale in the case of zero temperature considered is unimportant, in fact, only the relationships between the exchange integrals are important. Two independent parameters can be introduced for a clear presentation of the results; it is convenient to choose $\kappa = K/J$ and $\lambda = L/J$ as these parameters. The phase diagram in these variables for $J > 0$ is presented in Fig. 9.

The transitions between all phases occur on different rays of the line $\Lambda_1 = 0$ or the line $\Lambda_2 = 0$. The ferromagnetic phase is stable in the region $\Lambda_1 > 0$ and $\Lambda_2 > 0$, while the antiferromagnetic phase is stable at $\Lambda_1 < 0$ and $\Lambda_2 < 0$. In Fig. 9, these are the regions lying above or below both lines $\Lambda_1 = 0$ or $\Lambda_2 = 0$. The nematic phase exist in the range of parameters $\Lambda_1 < 0$ and $\Lambda_2 > 0$, while the antinematic phase is stable in the region $\Lambda_1 > 0$ and $\Lambda_2 < 0$, i.e., between the rays of these lines located, respectively, at $L > 0$ or $L < 0$. All phase transitions in material parameters are degenerate first-order transitions, i.e., the lines of mode stability loss coincide with the phase transition lines. The same property also takes place for a non-Heisenberg magnet with spin $S = 1$. Just as in the case of $S = 1$, the transition lines for the model with $S = 3/2$ are characterized by a higher symmetry. In our case, however, the situation is more interesting, because the regions of existence of all four phases touch at the point $\Lambda_1 = 0$, $\Lambda_2 =$

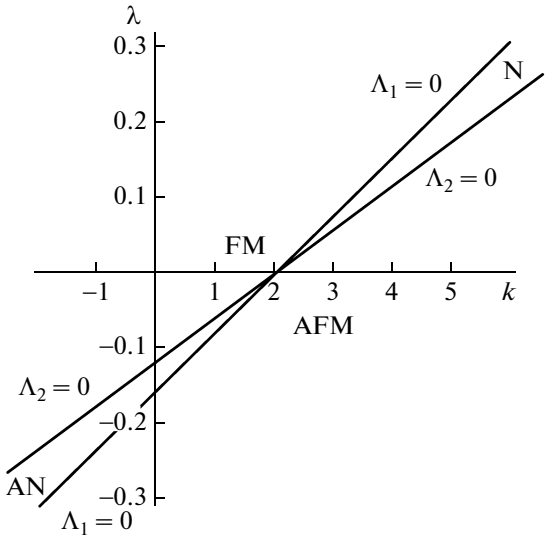

Fig. 9. Phase diagram for an isotropic non-Heisenberg magnet with magnetic-ion spin $S = 3/2$ in variables $\kappa = K/J$ and $\lambda = L/J$ at $J > 0$. The regions of existence of the ferromagnetic, antiferromagnetic, nematic, and antinematic phases are designated as FM, AFM, N, and AN, respectively.

0 or $2J = K$, $L = 0$ ($\kappa = 2$, $\lambda = 0$ in Fig. 9). All four phases have the same energy at this point and $\varepsilon_3(\mathbf{k}) = 0$, while the remaining perturbation spectra show a strong degeneracy.

The phase states of the system under consideration are characterized by nonzero quadrupole and octupole means. In contrast to the system with spin $S = 1$, the quadrupole means are not crucial for describing the phase symmetry, while the octupole order parameter in vector phases is zero. However, these variables turn out to have a significant influence on the dynamic (spectral) properties of a magnet with $S = 3/2$ in all phases, magnetic and nematic. As a result, three branches of elementary excitations are present in all phases. One of these branches, the octupole branch whose energy is designated above as $\varepsilon_3(\mathbf{k})$, is associated primarily with the oscillations of the octupole order parameter. This branch describes the positions of all phase instability lines, although the quadrupole branch with $\varepsilon = \varepsilon_2(\mathbf{k})$ also loses its stability on some of them. Interestingly, the energy at the point of coexistence of all four phases $2J = K$ and $L = 0$ is $\varepsilon_3(\mathbf{k}) = 0$, but the remaining modes have a finite hardness. The perturbation spectra exhibit a strong degeneracy for all phases, $\varepsilon_1(\mathbf{k}) = \varepsilon_2(\mathbf{k})$, and there is a universal dependence on the quasi-momentum: $\varepsilon_{1,2}(k) \propto (J - J_k) = J[1 - C(\mathbf{k})]$ in the nematic and ferromagnetic phases, while $\varepsilon_{1,2}(k) \propto \sqrt{J^2 - J_k^2} = J\sqrt{1 - C^2(\mathbf{k})}$ in the antinematic and antiferromagnetic phases.

The experimental realization of the effects considered here seems more difficult than that for materials with spin $S = 1$ (see the reviews [9, 10]). We know no standard crystalline magnetic materials for which a fairly large bicubic exchange has been established; moreover, even the works where this parameter was measured are not known. On the other hand, more and more materials with the so-called "hidden" order in which there is no magnetic order but whose properties also defy explanation in terms of simple spin-nematic (SN) phases (URu_2Si_2 whose properties have remained unexplained for almost ten years is cited as an example) have been found in recent years [71, 72]. However, model (1) is applicable for describing the purely spin states of ultracold Fermi gases with spin $S = 3/2$ (^{132}Cs, ^{9}Be, ^{135}Ba) in optical lattices with one atom per cell [56–59]. Here, it is important to note that considerable higher exchange integrals are a standard situation for ultracold gases. In particular, when calculating the exchange integrals for a gas with spin $S = 3/2$, the simplest approximation leads to a relationship close to the condition $\Lambda_1 = 0$ [56]. Thus, although the relative fraction of the regions on the phase diagram occupied by the nematic phases is, on the whole, small compared to the case of a system with spin $S = 1$, these states can be significant for analyzing ultracold gases of atoms with spin $S = 3/2$.

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