

# Collective modes of incommensurate structures in systems with dipole interactions

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In hexagonal magnets, dipole interactions can lead to the appearance of incommensurate structures, whose existence is due to instability at conical points. The temperature behavior of the collective modes for such structures is studied: in triangular antiferromagnets, the spectrum of collective excitations is found for two intermediate phases, one of which has only longitudinal modulation waves while the other has a combination of longitudinal and transverse waves with different periods (i.e., incommensurate relative to one another). The dispersion curves for the first of these phases have a point of intersection if the momentum of the excitations coincides with the wave vector of the structure. Gaps in the spectral band arise only when the latter phase has a (doubly) sinusoidal structure; it also has two gapless branches. The frequencies of the collective modes of these states are investigated, taking into account the effects of mixing in of higher order harmonics, which leads to spatial variation both in the phase and amplitude of the nonlinear wave. The fluctuation spectrum is determined for a vortical incommensurate structure.

## 1. INTRODUCTION

In crystal without a center of inversion, an incommensurate magnetic structure can occur which is related to the degeneracy of certain dispersion curves at symmetry points in the Brillouin zone. As we move away from such points, the degeneracy is lifted at a rate proportional to the slope of the wave vector, which is caused by the presence in the free energy of terms linear in the derivatives of Lifshitz invariants. Some incommensurate structures which arise in crystals can have a spiral configuration; these were first studied by Dzyaloshinskii.<sup>1</sup> In addition to these examples of structural phase transitions, it was shown<sup>2</sup> that in some compounds (among them crystals with the space groups  $C_6^1$ ,  $C_{6v}^1$ ,  $D_{6h}^1$  –  $D_{6h}^4$  and others), the occurrence of the incommensurate state is related to an instability at the conical points  $K$ , at which the degeneracy in the dispersion relations is lifted as we move away from the symmetry point  $K$  in wave vector space in any direction in the plane.

It is well known that in hexagonal magnets antiferromagnetic interactions between the magnetic ions of various chains cannot give rise to antiparallel positioning of the spins on a triangular lattice: for the  $XY$ -like spins the minimum in the volume free energy is attained when a  $120^\circ$ -structure appears consisting of three magnetic sublattices. In experiments on thermal and magnetic variations,<sup>3</sup> and subsequently in neutron diffraction experiments,<sup>4</sup> it was observed that in several such materials (such as, e.g., compounds like  $RbFeCl_3$ , which have been actively investigated recently) the transition from the paramagnetic phase to the  $120^\circ$  structure occurs through two intermediate incommensurate phases, in which either one of the two spin components or both spin components simultaneously are ordered. In Refs. 5 and 6 it was shown that the observed results can be well understood from the point of view of instabilities at the conical point, whose existence is due to dipole interactions.<sup>1)</sup> As

the energy  $J_2$  of the antiferromagnetic interaction decreases relative to the dipole energy  $\gamma_d$ , the character of the modulation structure changes; in the limit  $J_2 \ll \gamma_d$  the intermediate phases have a vortex configuration due to the transverse polarization of the eigenvectors of the dipole tensor.<sup>8</sup>

In the present work, the problem is to investigate the collective modes of the system in which the long-period modulation is caused by the dipole interactions. We will investigate the frequency spectrum of the modes for two limiting cases: when the antiferromagnetic interaction in the triangular lattice is significantly larger than the dipole interaction, and conversely when the antiferromagnetic interaction is negligible compared to it. The question of the excitation spectrum of a magnetic structure in which the homogeneous state is modulated because of competition between the exchange interactions of different signs was investigated by Izyumov and Laptev<sup>9</sup>; we will base most of the work in the present paper on their results. The collective modes for a pure sinusoidal modulation of the original  $120^\circ$  structure are phase and amplitude modes (this is also found to be the case in investigations, e.g., of the excitation spectrum above a state with a charge density wave<sup>10–12</sup>). In the intermediate state, when only one spin component is ordered, the longitudinal modulation mode corresponding to it retains its sinusoidal form as the temperature is decreased. However, after the transition to the state in which both spins order, a transverse mode is present as well as a longitudinal one. The interaction of these two waves further lowers the free energy; as the induced harmonics grow in intensity, the resulting wave changes from a pure sinusoid to a train of solitons. The appearance of this doubly commensurate configuration will result in not only a spatial change of phase but, also, generally speaking, a spatial change of amplitude of the nonlinear waves. For such strongly deformed states, collective modes have also been (numerically) investigated. Finally, the question of the vortex fluctuation spectrum of an

incommensurate structure will be investigated; these fluctuations are generated as a result of modulation of the homogeneous state in the basal plane.

Below we will investigate compounds with a hexagonal lattice, which are described by a Hamiltonian

$$\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2, \quad (1)$$

where

$$\mathcal{H}_1 = -2J_1 \sum_i S_i S_{i+1}$$

is the ferromagnetic interaction between spins within a chain along a  $c$ -axis (the  $z$ -axis); the second term  $\mathcal{H}_2$  describes the antiferromagnetic interaction between nearest spin chains and the dipole-dipole interactions

$$\mathcal{H}_2 = 2J_2 \sum_n S_n S_{n+a} + \sum_{i>j} D_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta, \quad (2)$$

$$D_{ij}^{\alpha\beta} = (g\mu_B)^2 \left( \frac{\delta_{\alpha\beta}}{R_{ij}^3} - 3 \frac{R_{ij}^\alpha R_{ij}^\beta}{R_{ij}^5} \right),$$

where  $R_{ij}$  is the spacing between spins  $S_i$  and  $S_j$ . The ordered state which arises as a result of instability of the paramagnetic phase is determined by the smallest eigenvalue of the Fourier component  $A_{\alpha\beta}(\mathbf{Q})$  of the Hamiltonian (1):

$$A_{\alpha\beta}(\mathbf{Q}) = J(\mathbf{Q}) \delta_{\alpha\beta} + D_{\alpha\beta}(\mathbf{Q}). \quad (3)$$

Here,

$$J(\mathbf{Q}) = -2J_1 \cos(\mathbf{Q}c) + 2J_2 [\cos(\mathbf{Q}a) + \cos(\mathbf{Q}b) + \cos(\mathbf{Q}(a-b))],$$

$$D_{\alpha\beta}(\mathbf{Q}) = \sum_{i,j} D_{ij}^{\alpha\beta} \exp(i\mathbf{Q}\mathbf{R}_{ij}), \quad (4)$$

where

$$\mathbf{a} = a(1, 0, 0), \quad \mathbf{b} = a(1/2, \sqrt{3}/2, 0), \quad \mathbf{c} = c(0, 0, 1)$$

are elementary translation vectors of the hexagonal lattice.

The expression for the free energy can be written down in the form of a Landau expansion correct to terms of fourth order in the average value of the spin component  $\langle S_i^\alpha \rangle$ :

$$F = \frac{1}{2} \sum_{i,j;\alpha,\beta} a_{ij}^{\alpha\beta} \langle S_i^\alpha \rangle \langle S_j^\beta \rangle + b \sum_i \langle S_i^\alpha \rangle^4, \quad b > 0. \quad (5)$$

Here the Fourier components

$$a_{\alpha\beta}(\mathbf{Q}) = A_{\alpha\beta}(\mathbf{Q}) + \alpha T \delta_{\alpha\beta} \quad (\alpha > 0)$$

are components of the inverse susceptibility tensor. The smallest eigenvalue of the inverse susceptibility  $a_-(\mathbf{Q}) = \lambda_-(\mathbf{Q}) + \alpha T$  [where  $\lambda_-(\mathbf{Q})$  is analogously the smallest eigenvalue of the matrix  $A_{\alpha\beta}(\mathbf{Q})$ ] changes sign at the point  $T_1 = -\lambda_-(\mathbf{Q}_1)/\alpha$ , which corresponds to the minimum value of the function  $\lambda_-(\mathbf{Q})$  for  $\mathbf{Q} = \mathbf{Q}_1$ .

Before we proceed to an investigation of the collective mode spectrum, in the following section we will investigate the thermodynamic equilibrium solution for a triangular an-

tiferromagnet, taking into account the possibility of deformation of the sinusoidal structure.

## 2. DIPOLE ANISOTROPY. PHASE AND AMPLITUDE MODULATION

When the dipole force is significantly smaller than the interchain exchange (antiferromagnetic) force, i.e.,  $\gamma_d \ll J_2$ , where  $\gamma_d = (g\mu_B)^2/a^3$ , the wave vector of the ordered state is close to the point  $\mathbf{Q}_k = (4\pi/3a, 0, 0)$  for which the quantity  $J(\mathbf{Q})$  in (4) is a minimum. In this case it is convenient to express the average value of the spin components by using a slowly-varying complex spatial amplitude

$$\langle S_i^\alpha \rangle = \psi_\alpha(\mathbf{R}_i) \exp(i\mathbf{Q}_k \mathbf{R}_i) + \text{c.c.},$$

$$\langle S_i^\beta \rangle = \psi_\beta(\mathbf{r}_i) \exp(i\mathbf{Q}_k \mathbf{R}_i) + \text{c.c.} \quad (6)$$

After substituting (6) into (5), we can write the free energy in terms of the variables  $\psi_\alpha$  in the following fashion<sup>6</sup>:

$$F = \int d\mathbf{r} \left\{ \sum_{\alpha,\beta} \psi_\alpha^* a_{\alpha\beta}(\mathbf{Q}_k - i\nabla) \psi_\beta + b [4(|\psi_x|^2 + |\psi_y|^2)^2 + 2(\psi_x^2 + \psi_y^2)^* (\psi_x^2 + \psi_y^2)] \right\}, \quad (7)$$

where the expression for the operator  $a_{xx}(\mathbf{Q}_k - i\nabla) = A_{xx}(\mathbf{Q}_k - i\nabla) + \alpha T$  is in the form [ $\mathbf{p} = -i\nabla$ ,  $r = \alpha(T - T_1)$ ]

$$a_{xx}(\mathbf{Q}_k + \mathbf{p}) = r - \gamma_d \eta (p_x - q_1^0) + \frac{3}{4} J_2 [p_x^2 - (q_1^0)^2] - \frac{3^h}{8} J_2 [p_x^3 - (q_1^0)^3] + \frac{3}{4} J_2 p_y^2 + J_1 p_z^2 + \frac{3^h}{8} J_2 p_x p_y^2 \quad (8)$$

( $\eta$  is a numerical constant depending on the lattice constants  $a$  and  $c$ ), while  $a_{yy}$  differs from  $a_{xx}$  by changing the sign in front of the term in (8) linear in  $p_x$ , i.e.,

$$q_1^0 = \frac{2}{3} \frac{\gamma_d \eta}{J_2} + \frac{3^h}{9} \left( \frac{\gamma_d \eta}{J_2} \right)^2$$

and corresponds to the minimum of  $\lambda_-(\mathbf{Q}_k + \mathbf{q})$ . The non-zero off-diagonal component (7) is due only to the dipole interaction

$$a_{xy}(\mathbf{Q}_k + \mathbf{p}) = \gamma_d \eta p_y. \quad (9)$$

The stable phases correspond to two inhomogeneous structures which exist within different temperature ranges and can be described in terms of functions of the spatial coordinate  $x$ . In the temperature interval  $T_2 \leq T < T_1$  ( $T_2$  is the point separating the two intermediate phases), there is only a single longitudinal wave  $\psi_x^0(x)$  [Ref. 5]:

$$\psi_x^0(x) = |\varphi_0| \exp[i(q_1^0 x + \vartheta)],$$

$$|\varphi_0|^2 = -r/12b \quad (10)$$

(here  $\vartheta$  is an arbitrary initial phase), which remains purely sinusoidal over this entire interval. However, below the point  $T_2$ , along with the longitudinal modulation wave there is also a transverse wave  $\psi_y^0(x)$ : their mutual interaction induces higher harmonics which are due to the nonlinear coupling in the free energy [the cross-terms  $(\psi_x^0 \psi_y^0)^2$  and  $(\psi_x^0 \psi_y^0)^2$  in (7)]. The growth in amplitude of the multiple

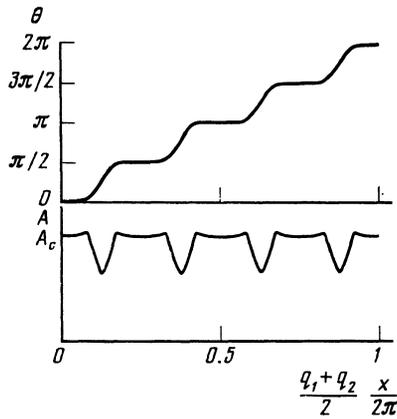


FIG. 1. Spatial dependence of the phase and amplitude of the longitudinal modulation wave  $\Phi_x^0(x) = A(x)\exp[i\theta(x)]$  for  $\delta_1 \equiv q_1/q_1^0 = 0.5$  ( $A_c$  is the amplitude of the corresponding commensurate  $120^\circ$  state).

harmonics leads in turn to a temperature variation in the wave vector. The higher harmonics of incommensurate magnetic structures generated by an external field or uniaxial anisotropy were studied earlier.<sup>13,14</sup> A more general solution to the equations which are obtained by variation of  $F$  has the following form in the temperature interval  $T_3 < T \leq T_2$  ( $T_3$  is the point of transition to the commensurate  $120^\circ$  state):

$$\psi_x^0(x) = e^{iq_1 x} \Phi_x^0(x), \quad \psi_y^0(x) = e^{-iq_2 x} \Phi_y^0(x), \quad (11)$$

$$\Phi_\alpha^0(x) = \sum_{m=-N}^N \varphi_\alpha^{(m)} \exp[-2mi(q_1 + q_2)x] \quad (\alpha = x, y),$$

where the harmonic amplitudes  $\varphi_\alpha^{(m)}$  and wave vectors  $q_1$  and  $q_2$  are determined by minimizing the free energy (7):

$$|\varphi_x^{(0)}|^2 = -\frac{3a_{xx}(Q_1) - 2a_{yy}(Q_2)}{20b},$$

$$\varphi_x^{(1)} = -\frac{4b\varphi_y^{(0)*}\varphi_x^{(0)*}}{a_{xx}(2Q_2 - Q_1)}, \dots, \varphi_x^{(m)} \sim \varphi_y^{(0)*}\varphi_x^{(m-1)*} \quad (12)$$

[here  $Q_1 = (Q_k + q_1, 0, 0)$ ,  $Q_2 = (Q_k - q_2, 0, 0)$ ]. Analogous expressions for  $\varphi_y^{(m)}$  are obtained after replacing the indices  $x, y$  and  $1, 2$  by  $y, x$  and  $2, 1$ , respectively:

$$q_1 = q_1^0 - (q_1^0 - q_2^0) \frac{2|\varphi_x^{(1)}|^2 + 4|\varphi_y^{(1)}|^2}{|\varphi_x^{(0)}|^2} - \dots, \quad (13)$$

$$q_2 = q_2^0 - (q_1^0 - q_2^0) \frac{2|\varphi_y^{(1)}|^2 + 4|\varphi_x^{(1)}|^2}{|\varphi_y^{(0)}|^2} - \dots,$$

$$q_2^0 = q_1^0 - \frac{2\sqrt{3}}{9} \left( \frac{\gamma_d \eta}{J_2} \right)^2.$$

The first term in the expansion (11) was obtained in Ref. 6.

If the dipole forces are increased, the values of the wave vectors  $q_1$  and  $q_2$  decrease more rapidly as the temperature is lowered. Mixing in the higher harmonics deforms the purely sinusoidal wave. Figure 1 shows the phase and amplitude of

the longitudinal wave  $\Phi_x^0(x)$  for  $\delta_1 = q_1/q_1^0 = 0.5$ . It is quite clear that for this value the nonlinear wave already has a domain-like form. The dependence of the phase on the spatial coordinate  $x$  is characterized by a step function, and is analogous to the dependence obtained earlier by Dzyaloshinskii in his investigation of phase transitions in spiral magnetic structures.<sup>1</sup> A similar step-function dependence also appears in other physical problems (see, e.g., Refs. 15–17). In our case, the domain walls (solitons) are due to both phase and amplitude variations. The latter decrease in the region of the domain wall after a small increase. In the region of the domains themselves, a practically commensurate structure is realized. The wave period which is determined by the spacing between domain walls equals  $\pi/(q_1 + q_2)$ . An analogous soliton form obtains for the phase and amplitude of the transverse wave  $\Phi_y^0(x)$ .

### 3. COLLECTIVE MODES OF A TRIANGULAR ANTIFERROMAGNET

Let us now investigate the temperature dependence of the collective modes for the various states which can occur in triangular antiferromagnets. We write the order parameter which describes fluctuations in the system in the form

$$\psi_\alpha(\mathbf{R}, t) = \psi_\alpha^0(\mathbf{R}) + \xi_\alpha(\mathbf{R}, t), \quad (14)$$

where  $\psi_\alpha^0(\mathbf{R})$  is the equilibrium-state order parameter and  $\xi_\alpha(\mathbf{R}, t)$  is a small deviation which depends on both the space and time coordinates. Substituting (14) into (7) and using the condition  $\delta F / \delta \psi_\alpha^*(\mathbf{R}) = 0$  for  $\psi_\alpha = \psi_\alpha^0$ , we can write  $\delta F = F - F_0$  to second order in  $\xi_\alpha$  (where  $F_0$  is the equilibrium-state free energy) and obtain

$$\delta F = \int d\mathbf{R} \left\{ \sum_{\alpha, \beta} \xi_\alpha^* a_{\alpha\beta} (\mathbf{Q}_k - i\nabla) \xi_\beta + 8b [3(|\psi_x^0|^2 |\xi_x|^2 + |\psi_y^0|^2 |\xi_y|^2) + |\psi_x^0|^2 |\xi_y|^2 + |\psi_y^0|^2 |\xi_x|^2] + 2b [(\psi_x^{02} + \psi_y^{02}) (\xi_x^2 + \xi_y^2) + 2(\psi_x^0 \xi_x^* + \psi_y^0 \xi_y^*)^2 + 4\psi_x^0 \psi_y^{0*} (\xi_x \xi_y^* + \xi_x^* \xi_y) + \text{c.c.}] \right\}. \quad (15)$$

The kinetic energy  $\mathcal{K}$  for the magnetic system in the general case consists of vibrational and precessional parts, which are described by quadratic and linear terms in the  $\xi_\alpha$ , respectively.<sup>18</sup> However, in the limit of strong anisotropy the precessional motion is suppressed.<sup>9</sup> Thus it is clear that in our case this motion will be suppressed because of the planar character of the spins ( $XY$  spins) in triangular antiferromagnets, so that the behavior of the collective modes will be determined by the vibrational motion. The kinetic energy of this motion takes the form

$$\mathcal{K} = \mu \int d\mathbf{R} (|\dot{\xi}_x|^2 + |\dot{\xi}_y|^2),$$

where  $\mu$  is the effective mass of an oscillating fluctuation. The dynamic behavior of the quantity  $\xi_\alpha$  is determined by the complex-conjugate equations:

$$\frac{d}{dt} \frac{\delta L}{\delta \dot{\xi}_\alpha^*} - \frac{\delta L}{\delta \xi_\alpha^*} = 0, \quad (16)$$

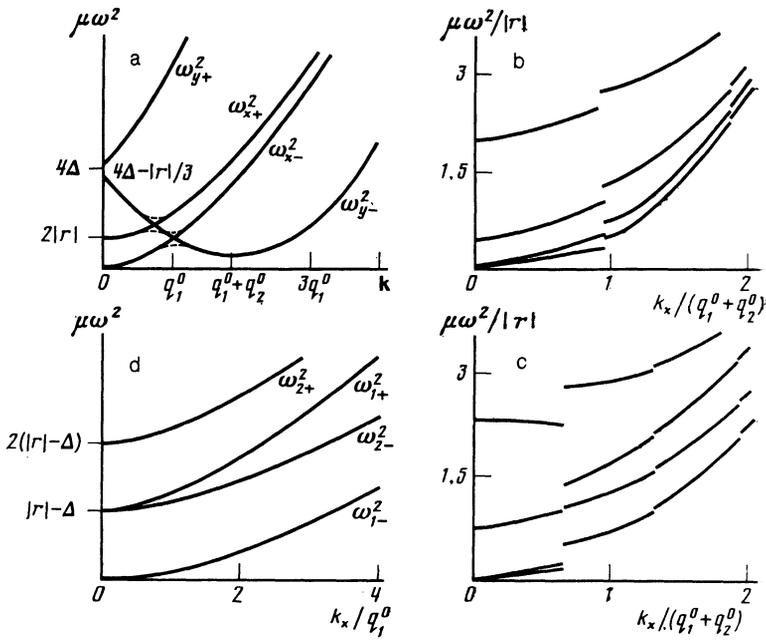


FIG. 2. Spectrum of collective modes: a—for a purely sinusoidal structure, when only one of the spin components is ordered (the continuous curves correspond to frequencies with different values of  $k_x$  for  $k_y = 0$ , the dotted curves are for  $k_y \neq 0$ ); b, c—for the doubly incommensurate structure with simultaneous ordering of both spin components for the values  $\delta_1 \approx \delta_2 = 0.97$  (b) and  $\delta_1 \approx \delta_2 = 0.65$  (c); d—for the commensurate  $120^\circ$  structure.  $\Delta \equiv \lambda_-(Q_k) - \lambda_-(Q_1) = (\gamma_d \eta)^2 / 3J_2$ —the depth of the potential well.

where  $L = \mathcal{H} - \delta F$  is the Lagrangian.

Let us turn to a study of the spectrum of excitations above the state which occurs at the instability point ( $T = T_1$ ) for the paramagnetic phase. In this case the incommensurate structure has only one component [ $\psi_x^0 = \varphi_0 \exp(iq_1^0 x)$ ] different from zero. Thus, we obtain from the Lagrange equation (16)

$$\begin{aligned} \mu \ddot{\xi}_x + \sum_{\alpha} a_{\alpha\alpha} (Q_k - i\nabla) \xi_{\alpha} + 12b\varphi_0^2 e^{2iq_1^0 x} \xi_x^* + 24b|\varphi_0|^2 \xi_x &= 0, \\ \mu \ddot{\xi}_y + \sum_{\alpha} a_{y\alpha} (Q_k - i\nabla) \xi_{\alpha} + 4b\varphi_0^2 e^{2iq_1^0 x} \xi_y^* + 8b|\varphi_0|^2 \xi_y &= 0. \end{aligned} \quad (17)$$

Equation (17) contains  $\xi_{\alpha}^*$  multiplied by a periodic coefficient [this also holds for  $\xi_{\alpha}$  in the second pair of equations complex-conjugate to (17)], which, however, disappears if we use the transformation  $\xi_{\alpha} \rightarrow \xi_{\alpha} \exp(iq_1^0 x)$ . After going to Fourier components

$$\xi_{\alpha}(\mathbf{k}, \omega) = \int d\mathbf{R} dt \xi_{\alpha}(\mathbf{R}, t) \exp[-i(\mathbf{k}\mathbf{R} - \omega t)]$$

we can easily find an expression for the eigenfrequencies in equations (17) for  $k_y = 0$ , which also corresponds to the vanishing of the nondiagonal component  $a_{xy} = A_{xy}$ ; this is apparent from (9) upon substituting  $k_y$  for  $p_y$ . In this case, the equations for  $\xi_x$  and  $\xi_y$  decouple, and as a result we have for the frequencies ( $k_y = k_z = 0$ ):

$$\begin{aligned} \mu \omega_{x\pm}^2 &= 24b|\varphi_0|^2 + \frac{1}{2} [a_{xx}(Q_1 + k_x) + a_{xx}(Q_1 - k_x)] \\ &\pm \left\{ \frac{1}{4} [a_{xx}(Q_1 + k_x) - a_{xx}(Q_1 - k_x)]^2 + 144b^2 |\varphi_0|^4 \right\}^{1/2}, \\ \mu \omega_{y\pm}^2 &= 8b|\varphi_0|^2 + \frac{1}{2} [a_{yy}(Q_1 + k_x) + a_{yy}(Q_1 - k_x)] \\ &\pm \left\{ \frac{1}{4} [a_{yy}(Q_1 + k_x) - a_{yy}(Q_1 - k_x)]^2 + 16b^2 |\varphi_0|^4 \right\}^{1/2}, \end{aligned} \quad (18)$$

where  $|\varphi_0|^2$  is determined from (10) while  $a_{\alpha\alpha}(Q_1 \pm k_x)$  is given by expressions of the type (8), if we put  $p_y = p_z = 0$  in them and make the substitution  $p_x \rightarrow q_1^0 \pm k_x$ .

The collective mode spectrum is shown in Fig. 2a. As a consequence of invariance under arbitrary changes in the initial phase  $\alpha$  in (10), one of the excitation branches of the longitudinal components ( $\omega_{x-}^2$ ) in the vicinity of  $k_x = 0$  is a phason branch: at the point  $k_x = 0$  the frequency of the  $\omega_{x-}^2$  branch remains equal to zero (a Goldstone mode) for all temperature intervals in which the equilibrium state  $\psi_x^0(x)$  is present. The frequency of the other branch ( $\omega_{x+}^2$ ) increases as the temperature falls (it is an amplitude mode<sup>9,10</sup>). As  $k_x$  increases both of these branches will intersect the phase (soft) mode  $\omega_{y-}^2$  for the transverse components  $\xi_y$ ,  $\xi_y^*$ . At the point  $k_x = q_1^0 + q_2^0$  (i.e., for  $Q_x = Q_k - q_2^0$  in the original system of coordinates) the soft mode becomes unstable if  $|r| = \alpha(T_1 - T_2)$ , where<sup>5</sup>

$$\alpha(T_1 - T_2) = 3[a_{yy}(Q_2) - a_{xx}(Q_1)] = 2 \cdot 3^{-1/2} \gamma_d \eta (\gamma_d \eta / J_2)^2.$$

For this value of  $|r|$  ( $T = T_2$ ) a phase transition occurs to a state in which the  $y$ -spin component with a wave vector  $Q_2 = Q_k - q_2$  (which differs from the relation  $Q_1 = Q_k + q_1$  satisfied by the  $x$ -component) also condenses out, so that as a result the intermediate state will be characterized by a doubly incommensurate structure, having an additional transverse component  $\psi_y^0(x)$  along with the longitudinal modulation component  $\psi_x^0(x)$ . In Fig. 2, the continuous line represents the fluctuation spectrum for  $k_y \neq 0$ : due to the mixing of the various oscillating components, hybridization of the branches occurs; repulsion of these branches leads ultimately to the appearance of a gap between them.

Let us now study the frequencies of the collective modes for the case when both spin components are ordered, i.e., below  $T = T_2$ . It is not hard to convince oneself that the Lagrange equations for fluctuations above the doubly incommensurate structure described by expressions (11) contain two types of periodic coefficients. After the transforma-

tion

$$\xi_x \rightarrow \xi_x e^{iq_1 x}, \quad \xi_y \rightarrow \xi_y e^{-iq_2 x} \quad (19)$$

these equations, for  $k_y = 0$  ( $a_{xy} = 0$ ) and  $k_x = 0$ , have the following form

$$\begin{aligned} & \mu \ddot{\xi}_x + a_{xx}(Q_k + q_1 - i\partial/\partial x) \xi_x + 4b[2(3|\Phi_x^0|^2 + |\Phi_y^0|^2) \xi_x + 3\Phi_x^0 \xi_x^* + 2\Phi_x^0 (\Phi_y^0 \xi_y^* + \Phi_y^0 \xi_y)] \\ & + \exp[-2i(q_1 + q_2)x] (\Phi_x^0 \xi_x^* + 2\Phi_x^0 \Phi_y^0 \xi_y) = 0, \end{aligned} \quad (20)$$

$$\begin{aligned} & \mu \ddot{\xi}_y + a_{yy}(Q_k - q_2 - i\partial/\partial x) \xi_y + 4b[2(3|\Phi_y^0|^2 + |\Phi_x^0|^2) \xi_y + 3\Phi_y^0 \xi_y^* + 2\Phi_y^0 (\Phi_x^0 \xi_x^* + \Phi_x^0 \xi_x)] \\ & + \exp[2i(q_1 + q_2)x] (\Phi_x^0 \xi_x^* + 2\Phi_y^0 \Phi_x^0 \xi_x) = 0. \end{aligned}$$

Equation (20), along with its complex conjugate, now contains periodic coefficients which give rise to terms like

$$\Phi_x^0 = \sum_m \varphi_x^{(m)} \exp[-2mi(q_1 + q_2)x],$$

all having the form  $\exp[\pm 2ni(q_1 + q_2)x]$ , which are non-vanishing under subsequent transformations. As a result, we find that the spectral band of the collective modes has breaks at the wave vectors  $k_x = n(q_1 + q_2)$ .

The picture of such a spectrum in the extended-zone scheme is given in Figs. 2b, 2c. The wave vector at the zone boundary is determined by the spacing between two solitons (Fig. 1). As a consequence of the breaking of translational symmetry there are two modes with eigenvectors

$$\begin{aligned} \Xi_1 &= (\xi_{1x}, \xi_{1x}^*, \xi_{1y}, \xi_{1y}^*) = 4^{-1/2}(1, -1, 1, -1), \\ \Xi_2 &= 4^{-1/2}(1, -1, -1, 1) \end{aligned}$$

which for  $k_x = 0$  are Goldstone modes (the invariance is with respect to a change in the initial phase of both components of the order parameter); the wave vectors corresponding to them are  $Q_x = Q_k + q_1$  and  $Q_x = Q_k - q_2$  [after a transformation inverse to (19)]. Numerical investigation of the spectrum of collective modes of Equation (2) based on finite-difference relations among the Bloch function coefficients<sup>9</sup> shows that when  $\delta_1 = q_1/q_1^0$  and  $\delta_2 = q_2/q_2^0$  decrease the gaps at the edges of the first Brillouin zone increase significantly. At the same time, a notably different behavior of the dispersion curves is observed at the limits of the first zone and outside of it. In the region  $k_x \gg q_1 + q_2$ , the frequency spectrum in Fig. 2c ( $\delta_1 \approx \delta_2 = 0.65$ ) has a form close to the spectrum of modes for the commensurate state (which corresponds to Fig. 2d, as will be shown below). Within the first zone, the frequencies of the two lower modes are close to zero for the whole interval  $0 < k_x < q_1 + q_2$ . The weakly dispersive dependences of these modes are caused by the increase in spacing between solitons (due to the decrease in  $\delta_1$  and  $\delta_2$ ) which, in turn, reduces the interaction between them: these modes correspond to oscillations of the solitons. The upper modes of the first zone correspond to oscillation of the thickness of the domain walls (solitons). Thus, as must be the case, the distortion in the sinusoidal structure leads to a separation of the modes in the frequency spectrum: the presence of some of these is related to the solitons themselves, while the others are related to the regions between

them, where the commensurate structure is realized. As  $\delta_1$  and  $\delta_2$  decrease, the number of modes in the spectrum corresponding to domains increases, and conversely the number of modes due to domain walls decreases. Such variations in the band spectrum take place up to the point where the system reaches the point  $T = T_3$ , at which it changes its spin configuration by a second-order phase transition from domain-like to the commensurate  $120^\circ$  structure.

In the new state, described by three magnetic sublattices, the components  $\psi_\alpha^0$  of the order parameter are coupled by the relation  $\psi_\alpha^0 = \pm i\psi_x^0$  [where  $|\psi_x^0|^2 \equiv |\psi_c|^2 = -a_{xx}(Q_k)/16b$ ], which minimizes the free energy for  $T < T_3$ . The eigenfrequencies are found from the equations of motion with constant coefficients obtained from (16), and for  $k_y = k_z = 0$  have the following form:

$$\begin{aligned} \mu\omega_{1\pm}^2 &= 1/2[a(Q_k + k_x) + a(Q_k - k_x)] + 24b|\psi_c|^2 \\ & \pm \{1/4[a(Q_k + k_x) - a(Q_k - k_x)]^2 + 64b^2|\psi_c|^4\}^{1/2}, \\ \mu\omega_{2\pm}^2 &= 1/2[a(Q_k + k_x) + a(Q_k - k_x)] + 40b|\psi_c|^2 \\ & \pm \{1/4[a(Q_k + k_x) - a(Q_k - k_x)]^2 + 64b^2|\psi_c|^4\}^{1/2}. \end{aligned} \quad (21)$$

These expressions are determined by neglecting terms of third order in  $k_x$  in the matrix elements  $a_{xx}$ ,  $a_{yy}$  in (8), so that

$$a_{yy}(Q_k \pm k_x) = a_{xx}(Q_k \mp k_x) \equiv a(Q_k \mp k_x).$$

The spectrum of excited modes above the commensurate state is shown in Fig. 2d, from which it is clear that one of the phase branches with eigenvector  $\Xi_1 = 4^{-1/2}(1, -1, 1, -1)$  in the vicinity of  $k_x = 0$  (i.e., for  $Q_x = Q_k$ ) is a Goldstone mode. The presence of Goldstone modes with wave vector  $Q_k = (4\pi/3a, 0, 0)$  is caused by breaking of the continuous symmetry of the ground state relative to rotations. The two other branches, corresponding to phase [ $\Xi_2 = 4^{-1/2}(1, -1, -1, 1)$ ] and amplitude [ $\Xi_3 = 4^{-1/2}(1, 1, -1, -1)$ ] modes, are degenerate at this value  $k_x = 0$ , which in turn is related to the intersection of two eigenvalues (the energy surfaces) of the matrix  $a_{\alpha\beta}(Q)$  at the conical point  $Q_k$ .

#### 4. COLLECTIVE MODES WITH VORTEX STRUCTURE

Let us now turn to an investigation of systems in which the dipole interactions between spins in a hexagonal lattice are significantly stronger than the exchange interaction  $J_2$ . Then the equilibrium state, determined from the equation

$$\sum_{j\beta} a_{ij}^{\alpha\beta} \langle S_j^\beta \rangle + 4b \langle S_i^\alpha \rangle \langle S_i \rangle^2 = 0, \quad (22)$$

obtained by varying the free energy (5) with respect to the variable  $\langle S_i^\alpha \rangle$ , is described already by a function of two spatial coordinates  $x_i$  and  $y_i$  in the basal plane; the thermodynamically stable structures have a vortex configuration, whose states near the transition point from the paraphase are in the form<sup>8</sup>

$$\langle S_x^0 \rangle = S_0 \sin q_1^0 y, \quad \langle S_y^0 \rangle = -S_0 \sin q_1^0 x, \quad (23)$$

where the index  $i$  on the spin and space variables  $x, y$  is omitted

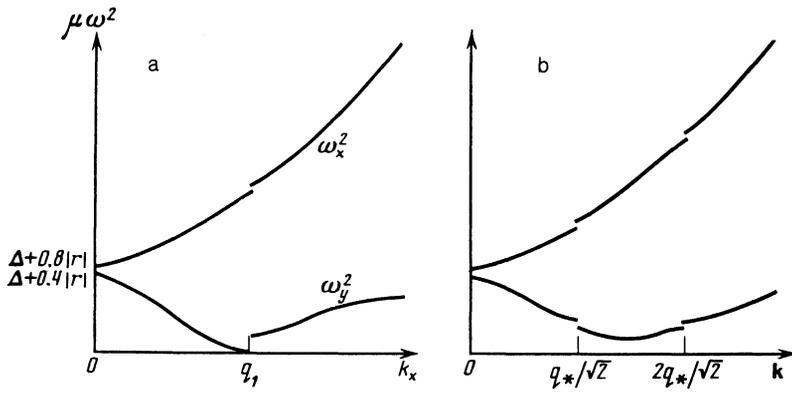


FIG. 3. Spectrum of collective modes of the vortex incommensurate structure for various orientations of the wave vector in the basal plane: a—frequency dependence on  $k_y$ , with  $k_x = 0$ , b—dependence of  $\mu\omega^2$  on  $\mathbf{k} = s\mathbf{q}_*$ , where  $\mathbf{q}_* = 2^{-1/2}(q_1, q_2)$ .  $\Delta$  is the potential well depth as before, but calculated for the case  $\gamma_d \gg J_2$  under investigation.

ted,

$$\langle S_\alpha \rangle \equiv \langle S_i^\alpha \rangle, \quad S_0 = [\alpha(T_1 - T)/5b]^{1/2},$$

where  $q_1^0$  corresponds to a minimum of the function  $a_-(Q)$  where  $Q = q_1^0$ . As the temperature is lowered, the fundamental wave (23) induces many new harmonics, so that the general solution to Equation (22) takes the following form

$$\langle S_x^{(0)} \rangle = \sum_{m,n} S_x^{(m,n)} \sin[2nq_1x + (2m+1)q_2y], \quad (24)$$

$$\langle S_y^{(0)} \rangle = \sum_{m,n} S_y^{(m,n)} \sin[(2m+1)q_1x + 2nq_2y],$$

where the amplitudes of the harmonics  $S_\alpha^{(m,n)}$  and projection of the wave vector  $q_1, q_2$  are determined from the condition that  $F$  in (5) be a minimum:

$$S_x^{(1,0)} = -b \frac{S_0^3}{a_-(3q_2)}, \quad S_x^{(0,1)} = -b \frac{S_0^3}{a_-(2q_1+q_2)}, \quad S_x^{(-1,1)} = -\frac{b}{4} \frac{S_0^3}{a_-(2q_1-q_2)}, \dots, \quad (25)$$

$$q_1 = q_1^0 \left\{ 1 - 6 \frac{(S_y^{(0,1)})^2}{S_0^2} - (\sqrt{5}-1) \frac{(S_y^{(1,0)})^2 + (S_y^{(-1,1)})^2 + 2[(S_x^{(0,1)})^2 + (S_x^{(-1,1)})^2]}{S_0^2} - \dots \right\}$$

$[(S_x^{(0,0)} = S_y^{(0,0)} \equiv S_0, \mathbf{q}_1 = (q_1, 0), \mathbf{q}_2 = (0, q_2)]$ . The quantities  $S_y^{(m,n)}$  and  $q_2$  are found from this by substituting  $y, x$  and  $2, 1$  for the subscripts  $x, y$  and  $1, 2$ , for which  $q_2^0 = q_1^0$ . As a result, we find that when higher harmonics are taken into account the amplitudes  $S_x^{(m,n)}$  and  $S_y^{(m,n)}$ , and also  $q_1$  and  $q_2$ , differ from one another due to the anisotropy of the smallest eigenvalue of the dipole tensor in  $Q$ -space [in expressions (25) this implies that  $a_-[\mathbf{m}\mathbf{q}_1 + \mathbf{n}\mathbf{q}_2] \neq a_-[\mathbf{n}\mathbf{q}_1 + \mathbf{m}\mathbf{q}_2]$ , and coincide only for the fundamental wave (23), when  $Q = q_1^0$ .

Let us investigate the behavior of collective modes for this case. The structure of the excitation spectrum generated by several wave-vector "stars"  $\{q\}$  (i.e., multiple- $q$  structures) was investigated earlier in Ref. 19, where it was shown that this case is characterized by the appearance of additional gaps. In the linear approximation the equations for the small deviation  $m$  from the equilibrium states (24) have the following form:

$$\mu \ddot{m}_x + \sum_{\alpha} a_{x\alpha} (-i\nabla) m_{\alpha} + 4b [ (3\langle S_x^0 \rangle^2 + \langle S_y^0 \rangle^2) m_x + 2\langle S_x^0 \rangle \langle S_y^0 \rangle m_y ] = 0, \quad (26)$$

$$\mu \ddot{m}_y + \sum_{\alpha} a_{y\alpha} (-i\nabla) m_{\alpha} + 4b [ (3\langle S_y^0 \rangle^2 + \langle S_x^0 \rangle^2) m_y + 2\langle S_x^0 \rangle \langle S_y^0 \rangle m_x ] = 0.$$

Equations (26) contain two kinds of periodic coefficients  $\langle S_\alpha^0 \rangle^2$  ( $\alpha = x, y$ ) and  $\langle S_x^0 \rangle \langle S_y^0 \rangle$ : in contrast to the case investigated above, neither can be removed. Gaps in the spectral band will arise along lines in the  $k_x k_y$  plane perpendicular to the vectors  $\mathbf{k} = m\mathbf{q}_1 + n\mathbf{q}_2$  (from the terms  $\langle S_\alpha^0 \rangle^2$ , where  $\mathbf{i}, \mathbf{j}$  are unit vectors along the  $x$  and  $y$  axes respectively) and  $\mathbf{k} = (m+1/2)\mathbf{q}_1 + (n+1/2)\mathbf{q}_2$  (from the terms  $\langle S_x^0 \rangle \langle S_y^0 \rangle$ ). The solution to equations (26) can be cast in the form of Bloch waves  $m_\alpha(\mathbf{r}) = e^{i\mathbf{k}\mathbf{r}} U_\alpha(x, y)$ , where  $U_\alpha(x, y)$  is a periodic function of the two spatial variables  $x, y$ . Fig. 3 gives a picture of the spectral band for two orientations of the wave vector, namely when  $\mathbf{k}$  is oriented along the  $x$ -axis (Fig. 3a) and when  $\mathbf{k}$  takes on values equal to  $s(q_1\mathbf{i} + q_2\mathbf{j})$ , where  $s$  is a continuous variable (Fig. 3b); in both cases  $q_1 \approx q_2 \approx q_1^0$ . The excitation spectrum of the vortex incommensurate structure contains two Goldstone modes with mutually orthogonal momenta  $\mathbf{k} = q_1\mathbf{i}$  (Fig. 3a) and  $\mathbf{k} = q_2\mathbf{j}$ , caused by breaking of the translational symmetry along the  $x$  and  $y$  axes in the basal plane. For orientations  $\mathbf{k}$  along the projections  $q_1$  and  $q_2$  of the wave vector of the magnetic structure, the width of the breaks at the edges of the first zone is proportional to  $|r| = \alpha(T_1 - T)$ ; for values of  $\mathbf{k} = n(q_1\mathbf{i} + q_2\mathbf{j})/2$  (Fig. 3b) the width of the breaks depends not only on the magnitude of the departure of the temperature from  $T_1$  but also on the non-diagonal components of the tensor  $a_{\alpha\beta}(Q)$ , and for  $n = 1$  is

proportional to the quantity  $|r|a_{xy}$ . As the temperature falls, there appears in the lower branch of the spectrum (Fig. 3b) a gap at  $k = q_*$   $\equiv [(q_1^2 + q_2^2)/2]^{1/2}$ , whose magnitude grows linearly with the increase in  $|r|$ .

## 5. CONCLUSION

In this work the spectrum of collective modes has been studied for states in hexagonal magnets caused by dipole interactions. In triangular antiferromagnets, when the condition  $\gamma_d \ll J_2$  is fulfilled, the dispersion curves have been obtained for excitation of the two incommensurate phases. For pure sinusoidal states with longitudinal wave modulation, the excitation spectrum, consisting of two phase and two amplitude modes, contains no breaks. If an excitation wave vector coincides with a wave vector of the structure, the equations for the longitudinal and transverse components of oscillating fluctuations become independent, and the dispersion curves in this case can intersect. As the temperature is decreased, the phase branch for transverse oscillations becomes unstable at the temperature point for which the transverse wave modulation also condenses. In the new state with two incommensurate structures, the mode spectrum is band-like, containing breaks as the momentum varies along spatial directions which depend on the magnetic structure; within the first zone of such a spectrum are two gapless (phason) branches.

The interaction of the longitudinal and transverse wave modulations leads to a distortion of the purely sinusoidal structure, so that the nonlinear wave generated as a result of this is characterized by both a spatially dependent phase and a spatially dependent amplitude. When the nonlinearity in the system is large, the amplitude preserves its constant value, and only in regions of strong phase variations does it decrease (after a small increase). The amplitude modes of the first Brillouin zone in the excitation spectrum above such a soliton state correspond to oscillations in the thickness of domain walls.

In the commensurate  $120^\circ$  state there remains only one gapless (Goldstone) mode, whose existence is due to breaking of rotational symmetry. In addition, there is a degeneracy at the symmetry point  $Q_k$ : the frequencies of one of the phase and one of the amplitude modes coincide in the presence of a

conical point in the potential surface of the eigenvalues.

In the other limiting case  $\gamma_d \gg J_2$ , the effect of mixing in higher harmonics leads to differentiation of the amplitudes of the transverse modes of the vortex structure and their periods  $2\pi/q_1$  and  $2\pi/q_2$  (which correspond in the oscillation spectrum to two Goldstone modes with mutually orthogonal wave vectors  $\mathbf{k} = q_1\mathbf{i}$  and  $\mathbf{k} = q_2\mathbf{j}$ ). In contrast to the case  $\gamma_d \ll J_2$ , the band spectrum has breaks as the wave vector varies in the basal plane; additional breaks arise due to the anisotropy of the dipole forces.

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<sup>1</sup>By the terminology "instability at the conical point" we mean also to include the magnetic properties of the triangular antiferromagnet CsCuCl<sub>3</sub> which are observed in a magnetic field.<sup>7</sup>

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