Modulated magnetic structure in hexagonal $CsCuCl_3$ type antiferromagnets (equilibrium state and spin dynamics)

E. P. Stefanovskiĭ

Donetsk State University, 340055 Donetsk, Ukraine

A. L. Sukstanskiĭ

Donetsk Physicotechnical Institute, Ukrainian Academy of Sciences, 340114 Donetsk, Ukraine

(Submitted 20 March 1993)
Zh. Eksp. Teor. Fiz. **104**, 3434–3449 (October 1993)

A theory is developed for triangular helicoidal antiferromagnetic structures due to the relativistic-exchange inhomogeneous Dzyaloshinskiĭ–Moriya interaction in hexagonal crystals. The magnetic structure observed in the $CsCuCl_3$ system is explained, and the spin-wave spectrum for this equilibrium state is found. The theoretical results are in agreement with experimental antiferromagnetic-resonance data for this compound.

Studies of physical systems whose magnetic properties (and not only magnetic properties) approach those of one-dimensional and two-dimensional systems have recently attracted continuing interest. Magnets with a highly anisotropic interaction are among them. Two factors are largely responsible for this. First, the behavior of such objects is significantly different from three-dimensional magnetic objects. Second, the chain-coupling exchange parameters are small in comparison with internal chain parameters of quasi-one-dimensional systems (or plane-coupling parameters in quasi-two-dimensional magnetically-ordered systems as compared with internal plane parameters). This means that such systems can be investigated with readily accessible external-parameter values.

The systems include the well-defined and extensive class of hexagonal magnetic compounds of the form ABX_3, where A and B are cations and X is a halogen. The compund $CsCuCl_3$ occupies a special place in this class and presents us with a relatively rare example of a modulated magnetic structure due to the relativistic-exchange interaction, i.e., a structure resulting from the competition between the Dzyaloshinskiĭ–Moriya and exchange interactions.[1)]

Although an extensitve literature is now available[5–10] on the experimental and theoretical studies of the magnetic properties of $CsCuCl_3$, there are a substantial number of unresolved problems relating to its magnetic structure and the dynamics of this structure.

In this paper, we examine some of the details of the magnetic structure of $CsCuCl_3$ that had previously escaped our attention, and investigate the acoustic branches of its intrinsic linear excitations.

1. GENERAL ASPECTS OF THE CRYSTAL AND MAGNETIC STRUCTURE OF $CsCuCl_3$ (EXPERIMENT)

Below $T_t=423$ K, in its paramagnetic state, the compound $CsCuCl_3$ has the $P6_122$ space group (without a center of symmetry). Six magnetoactive Cu^{2+} ions (spin $S=1/2$) occupy b-positions, i.e., they are shifted in the basal plane of the crystal relative to the hexagonal 6_1-axis by an amount $\varkappa$. The Cu^{2+} ions thus form spiral chains oriented along the 6_1 axis (the separation between neighboring ions in a chain is $\sim c_0/6$ for lattice parameters $c_0=18.1777$ Å and $a_0=7.2157$ Å). The important point is that, in this particular case, the shift $\varkappa$ (expressed in normalized units) is very small: $\varkappa\approx 0.06\ll 1$. The almost quasi-one-dimensional magnetic behavior of the system is dictated by the smallness of $\varkappa$ and the properties of the exchange, superexchange, and relativistic-exchange interactions[9] (in particular, the fact that interchain interactions are small in comparison with intrachain interactions).

Neutron diffraction data, obtained in the absence of an external magnetic field, suggest that, below $T_N=10.7$ K, the magnetically-ordered state of $CsCuCl_3$ displays a triangular antiferromagnetic (AFM) structure in the basal plane of the crystal and a long-period modulation of this structure along the hexagonal 6_1-axis (subsequently, the cartesian z-axis), i.e., the wave vector of the structure $\mathbf{q}$ is parallel to the $\mathbf{e}_z$ axis. The structure-period average of the angle between the magnetizations of the Cu^{2+} chain ions in neighboring basal planes is approximately 5.1°. Consequently, the modulation period is about 12 lattice constants.

2. THERMODYNAMIC POTENTIALS OF THE $CsCuCl_3$ SYSTEM

The triangular AFM structure in the basal plane requires the tripling of the magnetic unit cell as compared with the crystal unit cell in this plane. Since the triangular modulated AFM structure "grows out of" the simple (in Dzyaloshinskiĭ's sense[11]) triangular AFM structure with $\mathbf{q}=0$, the theoretical description of the magnetic properties of the $CsCuCl_3$ system requires, in general, eighteen magnetic sublattices.

However, if we exploit the results reported in Ref. 10, i.e., if we use the method of extended translational symmetry, we can go over to the three-sublattice description of the above object (each spin chain of Cu^{+2} ions is then described in terms of a single sublattice) and take the ab-

 1063-7761/93/100628-08$10.00

breviated Hamiltonian of the Cu^{+2} ion chain in the nearest-neighbor approximation:

$$\mathscr{H}_{ch}=\sum_{n}\{-J\mathbf{S}_n\mathbf{S}_{n+\Delta}+D(S_n^xS_{n+\Delta}^y-S_n^yS_{n+\Delta}^x)+KS_n^zS_{n+\Delta}^z-\mathbf{H}\mathbf{S}_n\},\quad(1)$$

where $\mathbf{S}_n$ is the nth spin in the chain, x and y are the cartesian coordinates in the basal plane of the system, $\mathbf{H}$ is the external magnetic field, and $J>0$, D, $K>0$ are the intrachain exchange, relativistic-exchange, and relativistic interaction constants, respectively. We note that (1) does not contain invariants corresponding to the single-ion magnetic anisotropy energy, since in our case $S=1/2$; in addition, we have omitted invariants that are small in comparison with the terms written out in the Hamiltonian, which is a quadratic function of the spins (this approximation is valid when $\varkappa$ is sufficiently small).

In this approximation, and since nearest neighbors in neighboring chains form layers of magnetoactive Cu^{2+}, we obtain the following Hamiltonian for the interchain interaction if we confine our attention to the exchange interaction within a layer:

$$\mathscr{H}_{int}=\sum_{n,n'}I_{nn'}\mathbf{S}_n\mathbf{S}_{n'},\quad(2)$$

where $\mathbf{n}$ is an arbitrary Cu^{2+} ion, $\mathbf{n}'$ are its nearest neighbors in the basal plane, and $I_{nn'}=I$ is the exchange interaction constant in the interior of a layer (it is clear that we must have $I>0$ if a triangular AFM structure is to be formed).

The experimental values of J and I reported in Ref. 9 suggest that $I/J\sim0.1$, which means that the Cu^{+2} magnetic system is almost quasi-one-dimensional. Moreover, for temperatures in the range $I<T<J$, the triangular AFM structure breaks up and the magnetic material becomes quasi-one-dimensional in the full sense of the phrase.

It is readily shown that the transition to the continuous limit and inclusion, in its simplest form, of the energy of hexagonal magnetic anisotropy in the basal plane lead to the following expresion for the thermodynamic potential in the weak-gradient approximation (we confine our attention to long-period modulations):

$$W=\int d\mathbf{r}w(\mathbf{r}),$$

$$w(\mathbf{r})=w_{ch}(\mathbf{r})+w_{int}(\mathbf{r})=\sum_{n=1}^{3}\left\{\frac{\alpha}{2}\left(\frac{\partial\mathbf{m}_n}{\partial z}\right)^2+\left[\frac{\alpha_\perp}{2}\left(\frac{\partial\mathbf{m}_n}{\partial x}\right)^2+\left(\frac{\partial\mathbf{m}_n}{\partial y}\right)^2\right]+\frac{\beta}{2}m_{nz}^2+\alpha_1\left(m_{nx}\frac{\partial m_{ny}}{\partial z}-m_{ny}\frac{\partial m_{nx}}{\partial z}\right)+\frac{\rho}{12}[(m_n^+)^6+(m_n^-)^6]-\mathbf{h}\mathbf{m}_n\right\}+\delta(\mathbf{m}_1\mathbf{m}_2+\mathbf{m}_1\mathbf{m}_3+\mathbf{m}_2\mathbf{m}_3),\quad(3)$$

where $\mathbf{m}_n=\mathbf{M}_n/M_0$, $\mathbf{M}_n$ is the magnetization of the sublattices, $n=1, 2, 3$, $|\mathbf{M}_n|=M_0=\text{const}$, $m_n^\pm=m_{nx}\pm im_{ny}$, $\alpha>0$, $\alpha_\perp>0$ are the inhomogeneous exchange interaction constants along chains and in the basal plane, respectively, $\delta>0$ is the homogeneous interchain (interplane) exchange interaction constant ($\delta\sim I$), $\alpha_1\sim(v/c)\delta c_0^{-1}$ is the intrachain relativistic-exchange interaction constant[1] whose sign, as will be seen later, is unimportant for the formation of a modulated AFM structure, v is the Fermi velocity of electrons, c is the velocity of light, $\beta>0$ and ρ are the crystal magnetic anisotropy constants, $\mathbf{h}=\mathbf{H}/M_0$, and $\mathbf{H}$ is the external magnetic field (we shall suppose that $\mathbf{h}=h\mathbf{e}_z$). We note that $\alpha_\perp\ll\alpha$, $\delta\gg\beta$, $\delta\gg|\rho|$.

3. MAGNETIC STRUCTURE OF $CsCuCl_3$ (THEORY)

We now parametrize the magnetization vectors of the sublattices in terms of the polar and azimuthal angles θ, φ_n (the polar axis of the spherical coordinate system lies along the hexagonal axis), i.e., we put

$$m_{nz}=\cos\theta_n,\quad m_n^\pm=\sin\theta_n\exp(\pm i\varphi_n),\quad(4)$$

so that (3) gives the following expression for the thermodynamic potential density:

$$w=\sum_{n=1}^{3}\left\{\frac{\alpha}{2}(\theta_n'^2+\varphi_n'^2\sin^2\theta_n)+\alpha_1\varphi_n'\sin^2\theta_n+\frac{\beta}{2}\cos^2\theta_n+\frac{\rho}{6}\sin^6\theta_n\cos6\varphi_n-h\cos\theta_n\right\}+\delta[\sin\theta_1\sin\theta_2\cos(\varphi_1-\varphi_2)+\sin\theta_1\sin\theta_3\cos(\varphi_1-\varphi_3)+\sin\theta_2\sin\theta_3\cos(\varphi_2-\varphi_3)+\cos\theta_1\cos\theta_2+\cos\theta_1\cos\theta_3+\cos\theta_2\cos\theta_3],\quad(5)$$

where the primes indicate differentiation with respect to z [we have omitted from (5) the invariants that are spatially inhomogeneous in the basal plane, since the magnetic structure of our system in its ground state varies only in the z direction].

It is readily verified that the equations $\delta W/\delta\theta_n=0$, $\delta W/\delta\varphi_n=0$ have solutions of the form

$$\theta_1=\theta_2=\theta_3=\theta,\quad\varphi_1=\varphi,\quad\varphi_{2,3}=\varphi\pm2\pi/3,\quad(6)$$

where the angles θ, φ satisfy the equations

$$\sin\theta\{\alpha\varphi''\sin\theta+2(\alpha\varphi'+\alpha_1)\theta'\cos\theta+\rho\sin^5\theta\sin6\varphi\}=0,\quad\alpha\theta''+\sin\theta\{(3\delta+\beta-\alpha\varphi'^2-2\alpha_1\varphi'-\rho\sin^4\theta\cos6\varphi)\cos\theta-h\}=0.\quad(7)$$

We note that, whatever the external magnetic field strength, the solution of (7) with $\theta=\text{const}=0$ is obviously

of no interest because it describes the "easy axis" ferromagnetic state that cannot arise in the absence of the external magnetic field for $\delta > 0$.

Let us examine the set of equations given by (7) for certain special cases.

A. Neglecting the hexagonal magnetic anisotropy in the basal plane of the crystal ($\rho = 0$), we can readily show that (7) assumes the form

$$\alpha\varphi'' \sin\theta + 2(\alpha\varphi' + \alpha_1)\theta' \cos\theta = 0,$$
$$\alpha\theta'' + \sin\theta[(3\delta + \beta - \alpha\varphi'^2 - 2\alpha_1\varphi')\cos\theta - h] = 0. \quad (8)$$

This set of equations has as its energetically most favorable solution the *FS*-structure (see Ref. 12 for an explanation of this notation) with the following parameters:

$$\varphi(z) = q_0 z, \quad q_0 = \alpha_1/\alpha,$$
$$\cos\theta = \text{const} = h/(3\delta + \beta - \alpha q_0^2 - 2\alpha_1 q_0), \quad (9)$$

where in view of the above estimates of the phenomenological constants in (3), we have

$$q_0 \sim (v/c)c_0^{-1} \ll c_0^{-1}.$$

Taken in conjunction with (4) and (6), this solution describes the equilibrium long-period *FS*-type modulation of the simple triangular AFM structure with the wave vector of the structure having the direction of the hexagonal axis and magnitude q_0. In other words, we are dealing here with three *FS*-structures of the same type, first described in Ref. 1, which are correlated into a triangular AFM structure in the basal plane of the system.

In the absence of an external magnetic field, the harmonic *FS*-structure transforms into the *SS*-structure ($\theta = \pi/2$), whereas in the presence of an external magnetic field $h = h_c = (3\delta + \beta - \alpha_1^2/\alpha)$, the modulated AFM structure collapses and there is a continuous phase transition to the ferromagnetic state.

B. If we drop the external magnetic field, but retain the energy of hexagonal magnetic anisotropy in the basal plane ($\rho \neq 0$), the second equation in (7) obviously has the solution

$$\theta = \text{const} = \pi/2, \quad m = 0\text{–}5, \quad (10)$$

whereas the first equation assumes the form

$$\alpha\varphi'' + \rho \sin 6\varphi = 0. \quad (11)$$

Unfortunately, we are not aware of any experimental data on the hexagonal magnetic anisotropy constant in the basal plane of $CsCuCl_3$. We shall therefore assume henceforth that $\rho > 0$, which will have no effect on our qualitative conclusions. If we were concerned in this case with spatially homogeneous (simple) magnetic states, the energetically favorable states would be those with

$$\varphi^{(m)} = \frac{\pi}{6}(2m+1), \quad m = 0\text{–}5$$

i.e., we would have a six-fold degenerate ground state.

The solution of (11) that describes the modulated AFM structure can be expressed in terms of the elliptic Jacobi functions[11,12]

$$\varphi = \frac{1}{3}\,\mathrm{am}\left(\frac{z-z'}{sz_0}, s\right), \quad \text{for } \sin 3\varphi = \mathrm{sn}\left(\frac{z-z'}{sz_0}, s\right), \quad (12)$$

where $z_0 = (\alpha/18\rho)^{1/2}$ is the characteristic length and z', s are constants of integration. The first of these constants is determined by the choice of the origin and can be set equal to zero without loss of generality. The second constant is the modulus of the elliptic functions and can be found by minimizing the total energy of the system (3) as a function of s. Substituting (10) and (12) in (3), and using (4)–(6), we obtain after some simple algebra the following expression for the equilibrium value of the parameter s:

$$\frac{E(s)}{s} = \frac{\pi\alpha_1}{2(2\alpha\rho)^{1/2}}, \quad (13)$$

where $E(s)$ is the complete elliptic integral of the second kind.

If we take (13) into account, we find that the equilibrium energy of the modulated AFM structure (per unit length) can be written in the form

$$W = -\rho M_0^2 \frac{2 - s_*^2}{2s_*^2}, \quad (14)$$

where s_* is the solution of (13).

The spatial period $\mathscr{L}_p$ of the magnetization distribution, i.e., the period of the modulated AFM structure, is given by

$$\mathscr{L}_p = 12sz_0K(s), \quad (15)$$

where $K(s)$ is the complete elliptic integral of the second kind. For small values of the hexagonal magnetic anisotropy constant $\rho \ll \alpha_1^2/\alpha$, the solution of (13) takes the form

$$s \approx \frac{1}{\alpha_1}(2\alpha\rho)^{1/2} \ll 1,$$

and the spatial period of the modulated AFM structure is

$$\mathscr{L}_p = \frac{2\pi}{q_0} + O(\rho^{1/2}). \quad (16)$$

It is readily seen that, for $\rho \to 0$, the spatial distribution of the magnetization of the AFM sublattices (12) takes the form of (9) for $h = 0$.

A different picture arises for $s \to 1$ (the case of high energy of magnetic anisotropy in the basal plane). In this case, the period of the spatial distribution of AFM-sublattice magnetization contains six segments with practically constant phases φ, namely,

$$\varphi = \varphi^{(m)} = \frac{\pi}{6}(2m+1),$$

and a sharp change in φ at the boundaries between these segments. The sublattice magnetizations in the modulated AFM structure form the triangular AFM structure and are 'held' in the energetically most favored directions $\varphi^{(m)}$

(this spatial distribution is sometimes referred to as a soliton lattice). The magnetic lattice of the crystal can then be thought of as being a periodic system of 'domains' $\varphi^{(m)}$, i.e., phases forming a coherent structure with a monotonic variation of φ.

The presence of a well-defined soliton lattice in the crystal is readily seen to explain the experimental results reported in Ref. 13 on the NMR spectra of Cu^{2+} nuclei in magnetic fields of different orientation in the basal plane of the system.

Obviously, if the energy of hexagonal magnetic anisotropy in the basal plane is high enough, long-period modulation of sublattice magnetization of the triangular AFM structure becomes impossible, and a spatially homogeneous triangular AFM distribution of sublattice magnetization is established (which does not, of course, exclude the presence in the crystal of true "domains" $\varphi^{(m)}$ due to the six-fold degeneracy of the ground state in the angle φ. We note that such domain systems are not thermodynamically stable states of a magnetic material, but are nevertheless observed experimentally.

Comparison of the energies of the soliton lattice and the spatially homogeneous sublattice magnetization distribution, given by (14), readily shows that the soliton lattice is energetically more favorable for

$$\rho < \rho_c = \alpha_1^2/2\alpha. \tag{17}$$

If $\rho \to \rho_c$, then $s_* \to 1$ and $\mathscr{L}_p \to \infty$. Consequently, the period of the structure increases without limit with increasing anisotropy constant, and for $\rho \to \rho_c$ the system transforms into a system of 60° domain boundaries at an infinite (formally) distance from one another. In fact, we obtain a spatially homogeneous magnetization distribution.

C. We now return to the general set of equations (7) for $\rho \neq 0$ and $h \neq 0$. A general analytic solution of this system does not seem possible. However, when the external field is low enough ($h \ll h_c$), we may expect that the angle at which the magnetization vectors leave the basal plane is also small: $|\pi/2 - \theta| \ll 1$. It is readily verified that, as for $h=0$, the first equation in (7) reduces to (11) to first order in the external magnetic field, i.e., the azimuthal angle distribution $\varphi(z)$ is again given by (12). To first order in $h \ll h_c$ and $\tilde{\theta} = \pi/2 - \theta \ll 1$, the second equation in (7) is a Hill-type inhomogeneous differential equation with periodic coefficients:

$$-\alpha\tilde{\theta}'' + \tilde{\theta}[3\delta + \beta - \alpha\varphi'^2 - 2\alpha_1\varphi'] = h, \tag{18}$$

where $\varphi = \varphi(z)$ is given by (12).

Analysis of (18) becomes significantly simpler if we recall that, within the entire domain of existence of the modulated AFM structure, excluding a logarithmically narrow band near ρ_c, the system has only one characteristic length, namely, the structure period $\mathscr{L}_p$. If we also recall that $\rho \ll \delta$ and $\mathscr{L}_p \gg (\alpha/\delta)^{1/2}$, the solution of (18) can be approximately written in the form

$$\tilde{\theta}(z) \approx \frac{h}{3\delta}\left(1 - \frac{\beta}{3\delta}\right) + \frac{h\rho}{9\delta^2} N(z),$$

$$N(z) = \frac{2-s^2}{s^2} + 4\,\mathrm{sn}^2\left(\frac{z}{sz_0}, s\right) - \frac{2\alpha_1}{s}\left(\frac{2}{\alpha\rho}\right)^{1/2} \mathrm{dn}\left(\frac{z}{sz_0}, s\right). \tag{19}$$

It is clear from (19) that, when the external magnetic field and the hexagonal anisotropy in the basal plane are simultaneously taken into account, we obtain a 'nutation' effect in which the modulated AFM structure includes the modulation of not only the components of the magnetizations of the AFM sublattices in the basal plane, but also their projections along the wave vector of the structure. The 'nutation' amplitude is of the order of $h\rho/\delta^2$ and the corresponding spatial period [i.e., the period of the polar angle $\theta(z)$] is, as expected, smaller than the period of the main structure by a factor of six.

If a "soliton lattice" is established in the system, then there are two characteristic dimensions, namely, the structure period $\mathscr{L}_p$ and the size Δ_0 of the transition region between the "domains" $\varphi^{(m)}$ (see above), where $\Delta_0 \ll \mathscr{L}_p$. The solution of (18) can then no longer be written in the relatively simple form of (19), but the qualitative picture remains unaltered. Under these conditions, we have $\varphi' \approx 0$, $\tilde{\theta} \approx h/3\delta$ in the interior of each 'domain' $\varphi^{(m)}$. On the other hand, within the region of rapid variation of the phase φ in the "domain" boundaries, the polar angle θ changes abruptly by an amount $\sim h\rho/\delta^2$.

The linear dependence (19) of the "nutation" amplitude on the external magnetic field (for given ρ) is, of course, no longer valid when we leave the linear approximation ($h \ll h_c$). Moreover, the effect vanishes in the opposite limit ($h \to h_c$), since the projections of magnetization $\mathbf{m}_n$ on to the basal plane tend to zero. Consequently, an increase in the field is accompanied by a nonmonotonic dependence of the 'nutation' amplitude, with a maximum in the interval $(0, h_c)$. The amplitude vanishes as $h \to 0$ and $h \to h_c$.

4. SPIN DYNAMICS OF $CsCuCl_3$ (ACOUSTIC BRANCHES)

We now turn to linear magnetic excitations (spin waves) superposed on the modulated AFM structure discussed above.

As noted in Sec. 2, a triangular modulated AFM structure in the $CsCuCl_3$ system can be described by a model with three effective magnetic sublattices. The magnetic dynamics of the system can then be analyzed with the help of the usual equations of motion of the sublattice magnetization vectors $\mathbf{m}_n$ (Landau–Lifshitz equations). In our model, these equations reduce to a relatively cumbersome and inconvenient system of six differential equations for the angle variables θ_n and φ_n that parametrize the unit vectors $\mathbf{m}_n$ (cf. Ref. 4).

To investigate the modulated AFM structure in the $CsCuCl_3$ system, we therefore use the very productive method of effective Lagrangians developed in Refs. 14–16. According to this method, any magnetic structure can be described in the exchange approximation by not more than

three mutually perpendicular unit vectors $\mathbf{l}_\sigma(\mathbf{r},t)$, $\sigma=1,2,3$ that do not alter their mutual orientation in different excited states, i.e., they provide a rigid reference frame. In the method of phenomenological Lagrangians, the dynamics of long-wave excitations can be investigated by including the relativistic interactions, which fixes the orientation of the magnetic vectors $\mathbf{l}_\sigma$ relative to the crystal axes,[16] assuming that they are nevertheless much weaker than the exchange interactions.

Any excited state specified by the vectors $\mathbf{l}_\sigma(\mathbf{r},t)$ can be obtained from an initial homogeneous state $\mathbf{l}_\sigma^{(0)}$ by rotation through an angle $\Phi(\mathbf{r},t)$:

$$\mathbf{l}_\sigma(\mathbf{r},t)=\hat{D}(\Phi)\mathbf{l}_\sigma^{(0)}, \tag{20}$$

where $\hat{D}(\Phi)$ is a three-dimensional orthogonal matrix.

We emphasize that the use of the effective Lagrangian method must be confined to long-wave excitations for which the characteristic spatial inhomogeneity scale is much greater than the crystal lattice constant, and the frequencies are much smaller than the exchange frequencies. We shall now focus our attention on these particular (hydrodynamic) excitations in which the dynamic bending of the crystal sublattices is small. In particular, we shall confine our attention to the acoustic branches of the spin-wave spectrum. Exchange branches of intrinsic linear magnetic excitations of the system, for which the concept of the 'rigid reference frame' is not valid, lies outside the scope of this paper.

A. In the absence of the external magnetic field $\mathbf{h}$ (see Sec. 3), the sublattice magnetization vectors $\mathbf{m}_n$ lie in the basal XY plane and form a triangular AFM structure. The mutually perpendicular unit vectors $\mathbf{l}_\sigma$ can therefore be taken in the form

$$\mathbf{l}_1^{(0)}=\frac{1}{3}(2\mathbf{m}_3^{(0)}-\mathbf{m}_1^{(0)}-\mathbf{m}_2^{(0)}),$$
$$\mathbf{l}_2^{(0)}=\frac{1}{\sqrt{3}}(\mathbf{m}_1^{(0)}-\mathbf{m}_3^{(0)}),\quad \mathbf{l}_3^{(0)}=[\mathbf{l}_1^{(0)}\mathbf{l}_2^{(0)}]. \tag{21}$$

For $\rho=0$ the orientation of the vectors $\mathbf{l}_1^{(0)}$ and $\mathbf{l}_2^{(0)}$ relative to the Cartesian x,y axes is not fixed, so that without loss of generality we can put $\mathbf{l}_1^{(0)}=\mathbf{e}_x$, $\mathbf{l}_2^{(0)}=\mathbf{e}_y$, where $\mathbf{e}_{x,y}$ are the corresponding unit vectors. We then have

$$l_{1i}(\mathbf{r},t)=D_{xi}(\mathbf{r},t),\quad l_{2i}(\mathbf{r},t)=D_{yi}(\mathbf{r},t). \tag{22}$$

The effective Lagrangian $\mathscr{L}$ describing the noncollinear antiferromagnetic medium takes the form[16]

$$\mathscr{L}=\int d\mathbf{r}\left\{\frac{\chi_\perp}{2g^2}[\omega_1^2(\Phi,\dot{\Phi})+\omega_2^2(\Phi,\dot{\Phi})]+\frac{\chi_\parallel}{2g^2}\omega_3^2(\Phi,\dot{\Phi})-U(\hat{D})\right\}, \tag{23}$$

where $\omega_i(\Phi,\dot{\Phi})$ are differential Cartan forms related to the rotation matrix $\tilde{D}(\Phi)$ by

$$\omega_i(\Phi,\dot{\Phi})=\frac{1}{2}\varepsilon_{ikl}D_{kj}\dot{D}_{lj}, \tag{24}$$

g is the gyromagnetic ratio, dots indicate differentiation with respect to time, $\chi_\perp$ and $\chi_\parallel$ are the transverse and parallel (relative to the vector $\mathbf{l}_3^{(0)}=[\mathbf{l}_1^{(0)}\mathbf{l}_2^{(0)}]$) susceptibilities of the AFM medium, $\chi_\perp$, $\chi_\parallel\sim\delta^{-1}$), ε_{ikl} is the fully antisymmetric tensor of rank three, and U is the "potential" energy of the magnetic medium, whose form can readily be obtained from the expression for the density of the thermodynamic potential (3), taking into account (21) and (22).

In the parametrization

$$D_{ik}=\delta_{ik}+2(\nu_i\nu_k-\boldsymbol{\nu}^2\delta_{1k})-2\nu_4\varepsilon_{ikj}\nu_j \tag{25}$$

of the rotation matrix, the differential forms ω_i take the form

$$\omega_1=2(\nu_4\dot{\nu}_1-\nu_1\dot{\nu}_4+\nu_2\dot{\nu}_3-\nu_3\dot{\nu}_2),$$
$$\omega_2=2(\nu_4\dot{\nu}_2-\nu_2\dot{\nu}_4+\nu_3\dot{\nu}_1-\nu_1\dot{\nu}_3),$$
$$\omega_3=2(\nu_4\dot{\nu}_3-\nu_3\dot{\nu}_4+\nu_1\dot{\nu}_2-\nu_2\dot{\nu}_1), \tag{26}$$

where $\nu_\mu=(\boldsymbol{\nu},\nu_4)$ are the components of the unit 4-vector, $\boldsymbol{\nu}^2+\nu_4^2=1$, and $\boldsymbol{\omega}^2=4\dot{\nu}_\mu^2$.

The next step is to parametrize the unit vector ν_μ with the help of the three angle variables, namely,

$$\nu_1=\cos\xi,\quad \nu_2=\sin\xi\cos\eta,\quad \nu_3=\sin\xi\sin\eta\sin\zeta/2,$$
$$\nu_4=\sin\xi\sin\eta\cos\zeta/2. \tag{27}$$

We note that it is precisely this parametrization that is particularly convenient in the analysis of spin waves in modulated AFM structures. A more standard and widely used parametrization is

$$\boldsymbol{\Phi}=n\tan\psi/2,\quad \mathbf{n}^2=1,$$

which has a simple physical meaning (rotation of the reference frame by an angle ψ around the axis defined by the unit vector $\mathbf{n}$), but is found to be inadequate because the vector $\mathbf{n}$ becomes meaningless on a denumerable set of values of the angle ψ.

If we use the specific form (3) of the thermodynamic potential of the system $CsCuCl_3$ together with (25)–(27), we can take the effective Lagrangian for the problem in the form

$$\mathscr{L}=M_0^2\int d\mathbf{r}\left[\frac{2\chi_\perp}{(2M_0)^2}(\dot{\xi}^2+\dot{\eta}^2\sin^2\xi+\tfrac{1}{4}\dot{\zeta}^2\sin^2\xi\sin^2\eta)+\frac{2(\chi_\parallel-\chi_\perp)}{(gM_0)^2}\right.$$
$$\times\left(\frac{1}{2}\zeta\sin^2\xi\sin^2\eta+\xi\cos\eta-\frac{1}{2}\dot{\eta}\sin 2\xi\sin\eta\right)^2$$
$$-\alpha\left[(\tilde{\nabla}\xi)^2+(\tilde{\nabla}\eta)^2\sin^2\xi+\frac{1}{4}(\tilde{\nabla}\zeta)^2\sin^2\xi\sin^2\eta\right.$$
$$+\frac{1}{2}\left((\tilde{\nabla}\zeta)\sin^2\xi\sin^2\eta+(\tilde{\nabla}\xi)\cos\eta\right.$$
$$\left.\left.\left.-\frac{1}{2}(\tilde{\nabla}\eta)\sin 2\xi\sin^2\eta\right)^2\right]-\beta\sin^2\xi\sin^2\eta(\cos^2\xi$$

$$+\sin^2\xi\cos^2\eta)-\alpha_1[(\cos^2\xi+\sin^2\xi\cos^2\eta)$$
$$\times(2\xi'\cos\eta-\eta'\sin 2\xi\sin\eta)+\zeta'\sin^4\xi\sin^4\eta]\Big\}, \tag{28}$$

where $\tilde{\nabla}=\mathbf{e}_z(\partial/\partial z+\alpha_\perp/\alpha)[\mathbf{e}_x(\partial/\partial x)+\mathbf{e}_y(\partial/\partial y)]$.

We have omitted from this expression the terms connected with the external magnetic field and the energy of hexagonal magnetic anisotropy in the basal plane, whose influence on the form of the spin-wave spectrum superposed on the modulated AFM structure will be discussed below.

The equations of motion corresponding to the Lagrangian (28) are rather cumbersome in their general form, so that we shall not write them out in full here.

It is readily verified that these equations have the following static solution:

$$\xi_0=\eta_0=\pi/2, \quad \zeta_0=qz, \tag{29}$$

and the corresponding rotation matrix is

$$\hat{D}=\begin{pmatrix}\cos\zeta_0 & -\sin\zeta_0 & 0\\ \sin\zeta_0 & \cos\zeta_0 & 0\\ 0 & 0 & 1\end{pmatrix}. \tag{30}$$

This shows that the static solution (28) corresponds to a rotation of the basic reference frame through an angle ζ_0 around the z-axis (hexagonal axis of the crystal), i.e., to the above modulated AFM structure.

If we use the Lagrangian given by (28) to construct the energy functional, we can readily show that its minimum is reached for $q=q_0=-\alpha_1/\alpha$. In other words, the distribution given by (29) describes the same modulated AFM structure that was discussed above in terms of the standard sublattice approach [Sec. 3, Eq. (9) with $h=0$].

To analyze linear excitations superposed on the modulated AFM structure, we put

$$\zeta=\zeta_0+\tilde{\zeta}(\mathbf{r},t), \quad \xi=\pi/2+\tilde{\xi}(\mathbf{r},t), \quad \eta=\pi/2+\tilde{\eta}(\mathbf{r},t), \tag{31}$$

where $\tilde{\zeta},\tilde{\xi},\tilde{\eta}$ are small deviations from the equilibrium distribution of (29), i.e., $|\tilde{\zeta}|,|\tilde{\xi}|,|\tilde{\eta}|\ll 1$, and we then linearize the equations of motion in these deviations. The result is the following set of second-order linear differential equations with constant (this is important) coefficients:

$$\alpha\tilde{\Delta}\tilde{\zeta}-[\chi_\parallel/(gM_0)^2]\,\ddot{\tilde{\zeta}}=0, \tag{32}$$

$$\alpha\tilde{\Delta}\tilde{\xi}-\frac{2\chi_\perp}{(gM_0)^2}\,\ddot{\tilde{\xi}}+\alpha q_0\tilde{\eta}'+\left(\frac{5}{4}\alpha q_0-\beta\right)\tilde{\xi}=0, \tag{33}$$

$$\alpha\tilde{\Delta}\tilde{\eta}-\frac{2\chi_\perp}{(gM_0)^2}\,\ddot{\tilde{\eta}}-\alpha q_0\tilde{\xi}'+\left(\frac{5}{4}\alpha q_0-\beta\right)\tilde{\eta}=0, \tag{34}$$

where $\tilde{\Delta}=\partial^2/\partial z^2+(\alpha_\perp/\alpha)(\partial^2/\partial x^2+\partial^2/\partial z^2)$.

Equation (32) is not coupled to the other two equations in the system and describes the Goldstone mode of the spin-wave spectrum with the gapless dispersion relation

$$\Omega_1(\mathbf{k})=gM_0\left(\frac{\alpha k_z^2+\alpha_\perp\mathbf{k}_\perp^2}{\chi_\parallel}\right)^{1/2}, \tag{35}$$

where Ω is the spin-wave frequency and k_z, $\mathbf{k}_\perp$ are the components of the wave vector along the hexagonal axis and in the basal plane, respectively. Oscillations of the reference vectors $\mathbf{l}_1$, $\mathbf{l}_2$ (and also of the sublattice magnetization vectors $\mathbf{m}_n$) in the basal XY plane correspond to this branch, which is typical for easy-plane magnetic media.

On the other hand, equations (33) and (34) describe the two branches of the spin-wave spectrum with the dispersion relation (and a gap)

$$\Omega_{2,3}(\mathbf{k})=\frac{gM_0}{(2\chi_\perp)^{1/2}}\left[\beta+\frac{\alpha_1^2}{\alpha}+\alpha_\perp\mathbf{k}_\perp^2+\alpha\left(k_z\pm\frac{q_0}{2}\right)^2\right]^{1/2}. \tag{36}$$

These branches have corresponding oscillations of the reference frame in which the vectors $\mathbf{l}_1$, $\mathbf{l}_2$ leave the xy plane (the oscillations take place around mutually perpendicular directions in the basal plane).

It is interesting to note that a spin-wave spectrum analogous to (36) is generated by the simpler two-sublattice AFM model whose thermodynamic potential density contains an invariant of the form $w_d=\alpha_1(L_xL_y'-L_yL_x')$ and has an easy-axis symmetry (rather than the easy-plane symmetry of our case). The equilibrium state of this type of AFM medium is, of course, spatially homogeneous, and the invariant w_d manifests itself only in the excitation spectrum.

On the other hand, in an easy-plane two-sublattice AFM medium with an invariant w_d and a modulated magnetic structure as the equilibrium state, there is, apart from the Goldstone branch of the spin-wave spectrum, only one branch similar to (36). This branch typically exhibits the nonreciprocity property $\Omega(-k_z)\neq\Omega(k_z)$ due to the modulated magnetic structure that originates from the invariants in the thermodynamic potenial that are linear in the first space derivatives.

In our case of modulated AFM structure, on the other hand, we again have nonreciprocity of each of the two branches of the spectrum with the gap, but the presence of two such branches restores the symmetry: $\Omega_2(-k_z)=\Omega_3(k_z)$.

B. We now turn to the effect of an external magnetic field $\mathbf{h}\|\mathbf{e}_z$ on the spin-wave spectrum, particularly the antiferromagnetic resonance frequencies.

According to Ref. 16, if the external magnetic field strength is weak enough in comparison with the field associated with the exchange interaction between the sublattices ($h\ll\delta$), then it can be taken into account by the effective Lagrangian method when we analyze the dynamics of the magnetic medium. The new effective Lagrangian can be obtained from (23) by introducing the replacement $\omega_i\to\omega_i+gM_0h_i$. It is then readily verified that, in our case, this signifies the appearance in the effective Lagrangian density (28) of the additional term

$$\frac{\chi_\parallel hM_0}{g}\omega_3=\frac{\chi_\parallel hM_0}{g}(2\dot{\xi}\cos\eta-\dot{\zeta}\sin^2\xi\sin^2\eta$$
$$-\dot{\eta}\sin 2\xi\sin\eta). \quad (37)$$

Equation (32), which describes the gapless Goldstone mode of the spin-wave spectrum, then remains valid as before, and equations (33) and (34) have to be modified as follows:

$$\alpha\tilde{\Delta}\tilde{\xi}-\frac{2\chi_\perp}{(gM)^2}\ddot{\tilde{\xi}}+\alpha q_0\tilde{\eta}'+\left(\frac{5}{4}\alpha_1 q_0-\beta\right)\tilde{\xi}+\frac{2\chi_\parallel h}{gM_0}\dot{\tilde{\eta}}=0, \quad (38)$$

$$\alpha\tilde{\Delta}\tilde{\eta}-\frac{2\chi_\perp}{(gM_0)^2}\ddot{\tilde{\eta}}-\alpha q_0\tilde{\xi}'+\left(\frac{5}{4}\alpha_1 q_0-\beta\right)\tilde{\eta}-\frac{2\chi_\parallel h}{gM_0}\dot{\tilde{\xi}}=0. \quad (39)$$

These equations describe spin waves with dispersion relations of the form

$$\Omega_{2,3}(\mathbf{k};h)=\left\{\Omega_{2,3}^2(\mathbf{k};0)+\left(\frac{\mu gM_0h}{2}\right)^2\right\}^{1/2}\pm\frac{\mu}{2}gM_0h, \quad (40)$$

where $\mu=\chi_\parallel/\chi_\perp$ and $\Omega_{2,3}(\mathbf{k},0)$ is given by (36).

It is clear from (40) that an external magnetic field parallel to the wave vector of the modulated AFM structure lifts the degeneracy of the AFM frequencies $\Omega_{2,3}$ that is present for $h=0$:

$$\Omega_{2,3}^{(h)}=\Omega_{2,3}(0;h)=\left\{\Omega_0^2+\left(\frac{\mu gM_0h}{2}\right)^2\right\}^{1/2}\pm\frac{\mu gM_0h}{2}, \quad (41)$$

where

$$\Omega_0=\frac{gM_0}{(2\chi_\perp)^{1/2}}\left(\beta+\frac{5\alpha_1^2}{4\alpha}\right)^{1/2}$$

is the AFM resonant frequency for $h=0$. One of the AFM resonant frequencies then increases with increasing external magnetic field and the other decreases.[2)] For small values of the external magnetic field ($gM_0h\ll\Omega_0$), the corresponding functions are linear, in agreement with experiment.[18]

C. When the hexagonal anisotropy in the basal plane of the system is taken into account, the magnetization distribution of the modulated AFM structure in the absence of the magnetic field is described by (12). The corresponding term in the effective Lagrangian $\mathscr{L}$, which we shall denote by $\mathscr{L}_\rho$, is exceedingly cumbersome when it is written in its general form in terms of the angle variables ξ, η, ζ. We therefore reproduce only the quadratic terms in the expansion of $\mathscr{L}_\rho$ in terms of the small deviations $\tilde{\xi}$, $\tilde{\eta}$, $\tilde{\zeta}$ that describe the spin-wave spectrum:

$$\mathscr{L}_\rho=M_0^2\int d\mathbf{r}\{9\rho\tilde{\xi}^2\cos 6\zeta_0+3\rho[(\tilde{\eta}^2-\tilde{\xi}^2)\cos 5\zeta_0$$
$$-2\tilde{\xi}\tilde{\eta}\sin 5\zeta_0]\}, \quad (42)$$

where $\zeta_0=\zeta_0(z)$ is given by (29).

It is clear from the structure of (42) that, as before, the equation of motion for $\tilde{\zeta}(\mathbf{r},t)$ splits off from the other two equations in the set:

$$\alpha\tilde{\Delta}\tilde{\zeta}-\frac{\chi_\parallel}{(gM_0)^2}\ddot{\tilde{\zeta}}+18\rho\tilde{\zeta}\cos 6\zeta_0=0. \quad (43)$$

Assuming that

$$\tilde{\zeta}(\mathbf{r},t)=\tilde{\zeta}(z)\exp(i\mathbf{k}_\perp\mathbf{r}_\perp-i\Omega t)$$

we readily find that (43) takes the form of the well-known Lamé equation[3)]

$$z_0^2\tilde{\zeta}''(z)+s^2\tilde{\zeta}(z)\left[1+\frac{1}{18\rho}\left(\frac{\Omega^2\chi_\parallel}{(gM_0)^2}-\alpha_\perp\mathbf{k}_\perp^2\right)\right.$$
$$\left.-2\,\mathrm{sn}^2\left(\frac{z}{z_0},s\right)\right]=0, \quad (44)$$

where the characteristic size z_0 and the modulus of the elliptic function s are given by (12) and (13).

The solution of (44) is well-known (see, for example, Ref. 20). The corresponding spectrum consists of two branches. The wave function corresponding to the first of them is

$$\tilde{\zeta}_v^{(1)}(z)=\frac{H(z/z_0+iv+K;s)}{\Theta(z/z_0;s)}\exp[iq_1(v)z],$$
$$q_1(v)=\frac{1}{z_0}\left\{\frac{\pi v}{2KK'}+Z(v,s')-s'^2\mathrm{cd}(v,s')\mathrm{sn}(v,s')\right\}, \quad (45)$$

where Θ, θ, and Z are, respectively, the Jacobi functions, $K'=K(s')$, $\mathrm{cd}(v,s')=\mathrm{cn}(v,s')/\mathrm{dn}(v,s')$, v is a dimensionless parameter, and $0\leqslant v\leqslant K'$. On the other hand, the dispersion relation for this branch is

$$\Omega_{11}^2(v)=\frac{(gM_0)^2}{\chi_\parallel}\left\{\alpha_\perp k_\perp^2+18\rho\left(\frac{s'}{s}\right)^2\mathrm{cd}^2(v,s')\right\}. \quad (46)$$

The minimum value of $\Omega_{11}^2(v)$ is reached for $v=K'$, and is zero. We then have $q_1(v=K')=\pi/2Kz_0$. On the other hand, the maximum value $\Omega_{11}^2(v)$ (for $\mathbf{k}_\perp$) is reached for $v=0$ and is given by

$$\Omega_{11\,\max}^2=\Omega_{11}^2(v=0)=\frac{18\rho(gM_0)^2s'^2}{\chi_\parallel s^2}. \quad (47)$$

We then have $q_1(v=0)=0$.

The second solution (second branch) of (44) is

$$\tilde{\zeta}_v^{(2)}(z)=\frac{H(z/z_0+iv;s)}{\Theta(z/z_0,s)}\exp[iq_2(v)z],$$
$$q_2(v)=\frac{1}{z_0}\left\{\mathrm{dc}(v,s')\mathrm{sn}(v,s')-\frac{\pi v}{2KK'}-Z(v,s')\right\}. \quad (48)$$

The corresponding dispersion relation is

$$\Omega_{12}^2(v)=\frac{(gM_0)^2}{\chi_\parallel}\left\{\alpha_\perp\mathbf{k}_\perp^2+18\rho\frac{\mathrm{dc}(v,s')}{s^2}\right\}, \quad (49)$$

where $\mathrm{dc}(v,s')=\mathrm{dn}(v,s')/\mathrm{cn}(v,s')$.

The minimum value of $\Omega_{12}^2(v)$ is reached for $v=0$ and is given by (for $\mathbf{k}_\perp=0$)

$$\Omega_{12\,\min}^2=\Omega_{12}^2(v=0)=\frac{18\rho(gM_0)^2}{\chi_\parallel s^2}. \quad (50)$$

If, on the other hand, $v\to K'$, we have $\Omega_{12}\to\infty$, $q_2\to\infty$.

Comparison of (47) and (50) shows that there is a gap between the above branches of the spin-wave spectrum. The gap is given by

$$\Omega_{12\,\min}^2-\Omega_{11\,\max}^2=\frac{18\rho(gM_0)^2}{\chi_\parallel}. \quad (51)$$

We thus find that, when the hexagonal magnetic anisotropy in the basal plane of the system is taken into account, the spectrum of the Goldstone mode acquires a forbidden band of magnon energy values, and the size of this gap is proportional to $\rho^{1/2}$. As $\rho\to 0$, both the forbidden-gap width and the width Ω_{11}^2 of the lower branch are found to vanish. In this situation, we again obtain the plane-wave solution with the acoustic dispersion relation (35).

If we consider the effect of the hexagonal magnetic anisotropy in the basal plane of the system on the two other spin-wave modes described by the angle variables $\tilde{\xi}$ and $\tilde{\eta}$, we find that the presence of the variable functions $\cos 5\zeta_0$ and $\sin 5\zeta_0$ in front of these variables in the expression for $\mathscr{L}_\rho$ ensures that an analytic solution of the corresponding set of equations is practically impossible. The determination of the spectrum of the corresponding modes would require relatively cumbersome numerical calculations, which are outside the scope of the present paper. Here we merely mention that the spectrum is again of the band type.[12]

The authors are indebted to T. K. Soboleva and V. I. Belykh for useful discussions.

This research was supported by a grant from the Soros Foundation, provided by the American Physical Society, and by the Ukrainian Ministry of Education.

[1)]As far as we know, apart from $CsCuCl_3$, this type of modulated magnetic structure has been reliably established only in MnSi and FeGe (Refs. 1 and 2) and in $TbAsO_4$ (Refs. 3 and 4). In most magnetically-ordered objects, the presence of a modulated magnetic structure is due to the competition of exchange interactions.

[2)]We note that the above analogy between the spectrum of spin waves in the two-sublattice easy-axis antiferromagnetic medium,[17] on the one hand, and (36), on the other, is also found to occur for the dependence of the spin-wave frequencies on the external magnetic field.

[3)]An analogous equation arose in the study of the spin-wave spectrum superposed on modulated magnetic structures in easy-plane two-sublattice antiferromagnetic medium with rhombic magnetic anisotropy, or when the external magnetic field was taken into account in the basal plane (cf. Ref. 19).

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Translated by S. Chomet