Incommensurate magnetic phase and its dynamics in a hexagonal antiferromagnet without an inversion center

E. P. Stefanovskiĭ and A. L. Sukstanskiĭ
Donetsk Physicotechnical Institute, Ukrainian Academy of Sciences, 340114, Donetsk, the Ukraine

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A phenomenological theory is constructed for modulated triangular antiferromagnetic structures of exchange-relativistic origin in a hexagonal magnetic material lacking an inversion center. The magnetic structure of the compound $CsCuCl_3$ is explained. The spectrum of natural linear magnetic excitations is derived. The theoretical field dependence of the antiferromagnetic-resonance frequencies agrees well with experimental data.

Neutron diffraction experiments[1] have shown that a modulated triangular antiferromagnetic structure exists in the compound $CsCuCl_3$ below $T_N=10.7$ K. The space group of the paramagnetic phase of this compound is $P6_122$, and there are six magnetically active Cu^{2+} ions in b-sites. The wave vector of this modulated antiferromagnetic structure is oriented along the c axis of the hexagonal crystal. The modulation period is about 12 lattice constants. The magnetic moments of the Cu^{2+} ions lie in the basal plane of the crystal (in the absence of an external magnetic field). Previous theoretical work[2,3] has not drawn a comprehensive picture of the equilibrium state in this system, and the resonance properties have received essentially no attention.

Although the magnetic unit cell of $CsCuCl_3$ contains 18 Cu^{2+} ions, symmetry considerations (in particular, use of the method of expanded translational symmetry[3,4]) make it possible to describe the static and low-frequency dynamic magnetic properties of this material in terms of three magnetic sublattices. In the continuum approximation the thermodynamic potential of this system can be written

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

$$w=\sum_{n=1}^{3}\left\{\frac{\alpha}{2}(\mathbf{M}_n')^2+\alpha_1(M_{nx}M_{ny}'-M_{ny}M_{nx}')+\frac{\beta}{2}M_{nz}^2+\frac{\rho}{12}[(M_n^+)^6+(M_n^-)^6]\right\}$$
$$+\delta(\mathbf{M}_1\mathbf{M}_2+\mathbf{M}_1\mathbf{M}_3+\mathbf{M}_2\mathbf{M}_3), \qquad (1)$$

where $\mathbf{M}_n$ are the magnetizations of the sublattices ($n=1,2,3$; $|\mathbf{M}_n|=M_0$); $\alpha\sim Ic^2$, where c is the lattice constant; $I>0$ and $\delta>0$ are the constants of, respectively, the intra- and intersublattice exchange interactions; $\alpha_1\sim dc$ is the constant of the nonuniform exchange-relativistic interaction;[5] $\beta>0$ and $\rho>0$ (for definiteness) are the constants of the uniaxial and hexagonal anisotropies; $M_n^{\pm}=M_{nx}\pm iM_{ny}$; the prime means

differentiation with respect to the coordinate z, which runs along the c axis of the crystal [only the gradients along the hexagonal axis are considered in (1)]; and $\mathbf{H}=H\mathbf{e}_z$ is the external magnetic field.

We parametrize the vectors $\mathbf{M}_n$ by means of the angle variables θ_n and φ_n:

$$M_{nz}=M_0\cos\theta_n, \quad M_n^{\pm}=M_0\sin\theta_n\exp(\pm i\varphi_n).$$

It is not difficult to show that the equilibrium modulated antiferromagnetic structure corresponding to the minimum of potential (1) is described by the following expressions in a weak external field ($H\ll\delta M_0$):

$$\sin 3\varphi_1(z)=\mathbf{sn}(z/sz_0,s), \quad \varphi_{2,3}(z)=\varphi_1(z)\pm 2\pi/3, \quad z_0=(\alpha/18\rho)^{1/2},$$

$$\theta_n(z)=\pi/2-\theta_0(H)+\psi(z;H), \tag{2}$$

where $\theta_0(H)\sim H/\delta M_0\ll 1$. The function $\psi(z;H)$ describes a sort of nutation of the vectors $\mathbf{M}_n$, i.e., a modulation of their projections onto the hexagonal axis. Since this function is rather unwieldy, we will simply point out that it is periodic along z with a period of $2sz_0K(s)$, and its amplitude is small: $|\psi(z;H)|\ll\theta_0(H)\ll 1$. If $\rho=0$ or $H=0$, this nutation effect does not occur.

The modulus of the elliptic functions s, which determines the spatial period of the modulated antiferromagnetic structure, $Z_p=12sz_0K(s)$, is found by minimizing thermodynamic potential (1) with respect to this parameter. As a result, we find the equation

$$K^2(s)=\frac{\pi\alpha_1}{2\sqrt{2\alpha\rho}}\frac{sE(s)}{s'^2}, \tag{3}$$

where $s'^2=1-s^2$, and $K(s)$ and $E(s)$ are the complete elliptic integrals of the first and second kinds, respectively.

If the hexagonal anisotropy is sufficiently weak ($\rho\ll\alpha_1^2/\alpha$), we have $s\sim\rho^{1/2}$ and $Z_p=(2\pi\alpha/\alpha_1)+O(\rho^{1/2})$. If the hexagonal anisotropy is instead pronounced, the existence of a modulated antiferromagnetic structure becomes less favorable from the energy standpoint than a uniform distribution of the magnetization. The point at the phase transition from the modulated antiferromagnetic structure to a uniform triagonal antiferromagnetic structure is determined by Eq. (3) and the condition

$$K(s)=\pi\alpha_1 s/(2\alpha\rho)^{1/2}. \tag{4}$$

We have used the method of effective Lagrangians[6,7] to study the dynamic properties of this system, in particular, the acoustic branches of the spin-wave spectrum against the background of the modulated antiferromagnetic structure. The analysis by this method is much simpler than that which would be required if we took the standard approach and worked from the Landau–Lifshitz equations. In the effective-Lagrangian method, a magnetic subsystem of the crystal is described in terms of three mutually perpendicular unit vectors $\mathbf{l}_\sigma(\mathbf{r},t)$, $\sigma=1, 2, 3$, whose relative orientations remain the same in the various excited states. In other words, these vectors form a

rigid frame of reference. Any excited state $\mathbf{l}_\sigma(\mathbf{r},t)$ can be generated from some uniform state $\mathbf{l}_\sigma^{(0)}$ by rotation through an angle $\mathbf{\Phi}(\mathbf{r},t)$: $\mathbf{l}_\sigma=\hat{D}(\mathbf{\Phi})\mathbf{l}_\sigma^{(0)}$, where $\hat{D}(\mathbf{\Phi})$ is a three-dimensional orthogonal matrix.

The density of the effective Lagrangian L describing noncollinear antiferromagnets, such as $CsCuCl_3$ takes the following form[7] in a weak external magnetic field parallel to the z axis:

$$L=\frac{\chi_\perp}{2g^2}\Big[(\omega_1^2(\mathbf{\Phi},\dot{\mathbf{\Phi}})+\omega_2^2(\mathbf{\Phi},\dot{\mathbf{\Phi}})]+\frac{\chi_\parallel}{2g^2}[\omega_3(\mathbf{\Phi},\dot{\mathbf{\Phi}})+gH]^2-U\{\hat{D}(\mathbf{\Phi})\}. \tag{5}$$

Here g is the gyromagnetic ratio, $\chi_\perp$ and $\chi_\parallel$ are, respectively, the transverse and longitudinal magnetic susceptibilities ($\chi_\perp, \chi_\parallel \sim\delta^{-1}$), $\omega_k(\mathbf{\Phi},\dot{\mathbf{\Phi}})=\frac{1}{2}\epsilon_{klm}D_{lj}\dot{D}_{mj}$ are differential Cartan forms, the dot means differentiation with respect to the time, and the "potential energy" U of the magnetic material is expressed in terms of the components of the matrix $\hat{D}$. The particular structure of U is determined by the symmetry of the material. In particular, it can easily be found from the form of thermodynamic potential (1) through a determination of the specific relationship between the vectors $\mathbf{M}_n$ and $\mathbf{l}_\sigma$. In the case at hand it is convenient to choose the vectors $\mathbf{l}_\sigma$. In the case at hand it is convenient to choose the vectors $\mathbf{l}_\sigma$ as follows: $\mathbf{l}_1=(1/3M_0)\times(2\mathbf{M}_3-\mathbf{M}_1-\mathbf{M}_2)$, $\mathbf{l}_2=(1/\sqrt{3}M_0)(\mathbf{M}_1-\mathbf{M}_2)$, $\mathbf{l}_3=[\mathbf{l}_1,\mathbf{l}_2]$.

The following parametrization of the matrix $\hat{D}$ is the most convenient one for our problem:

$$D_{ik}=\delta_{ik}+2(\nu_i\nu_k-\boldsymbol{\nu}^2\delta_{ik})-2\nu_4\epsilon_{ikj}\nu_j. \tag{6}$$

Here $\nu_\mu=(\boldsymbol{\nu},\nu_4)$ are the components of a four-dimensional unit vector ($\boldsymbol{\nu}^2+\nu_4^2=1$) which is specified by three independent angle variables:

$$\nu_\mu=[\cos\xi,\ \sin\xi\cos\eta,\ \sin\xi\sin\eta\sin(\zeta/2),\ \sin\xi\sin\eta\cos(\zeta/2)]. \tag{7}$$

The Euler-Lagrange equations for Lagrangian (5) have the static solution $\xi_0=\eta_0=\pi/2$, $\sin 3\zeta_0=\mathrm{sn}(z/sz_0,s)$. It is not difficult to verify that this solution describes the same modulated structure as is described by expression (2).

Linear excitations (spin waves) against the background of the modulated antiferromagnetic structure are described by equations of motion which are linearized around the ground state. If we ignore the hexagonal anisotropy ($\rho=0$), the spin–wave spectrum consists of three branches. One corresponds to oscillations of the angle variable $\zeta(\mathbf{r},t)$; this branch is a Goldstone mode with the linear dispersion relation

$$\Omega_1(\mathbf{k})=\frac{gM_0}{\chi_\parallel^{1/2}}(\alpha k_z^2+\alpha_\perp\,\mathbf{k}_\perp^2\,)^{1/2}, \tag{8}$$

where $\mathbf{k}=(\mathbf{k}_\perp,k_z)$ is the wave vector of the wave, $\alpha_\perp\sim\delta a^2$ is the nonuniform-exchange constant in the basal plane of the crystal ($\alpha_\perp\ll\alpha$), and a is the lattice constant in this plane. This Goldstone mode is characteristic of easy-plane magnetic materials.

The two other branches of the spectrum correspond to oscillations of the variables $\xi(\mathbf{r},t)$ and $\eta(\mathbf{r},t)$ at frequencies

$$\Omega_{2,3}(\mathbf{k},H)=\left[\Omega_{2,3}^2(\mathbf{k},0)+\left(\frac{\chi_\parallel gH}{2\chi_\perp}\right)^2\right]^{1/2}\pm\frac{\chi_\parallel gH}{2\chi_\perp},$$

$$\Omega_{2,3}(\mathbf{k},0)=\frac{gM_0}{(2\chi_\parallel)^{1/2}}\left[\beta+\frac{\alpha_1^2}{\alpha}+\alpha_\perp\mathbf{k}_\perp^2+\alpha\left(k_z\pm\frac{\alpha_1}{\alpha}\right)^2\right]^{1/2}. \tag{9}$$

In the simpler model of a two-sublattice easy-plane antiferromagnet with a modulated structure as ground state, the Goldstone mode is accompanied by only one activation branch of the spectrum, like (9). For the latter, there is a nonreciprocity $[\Omega(\mathbf{k}_z)\neq\Omega(-\mathbf{k}_z)]$ due to the presence of the modulated structure. In our model the modulated structure also causes a nonreciprocity of each of the activation branches $[\Omega_{2,3}(\mathbf{k}_z)\neq\Omega_{2,3}(-\mathbf{k}_z)]$, but the presence of two such branches restores the symmetry: $\Omega_2(\mathbf{k}_z)=\Omega_3(-\mathbf{k}_z)$.

In two-sublattice antiferromagnets a symmetric spectrum like (9) holds for easy-axis antiferromagnets with a nonuniform exchange-relativistic interaction, in which the ground state is uniform. The field dependence of the antiferromagnetic-resonance frequencies in such materials is also similar to that of the antiferromagnetic-resonance frequencies [i.e., the quantities $\Omega_{2,3}(0,H)$] in our model. In weak fields ($gH\ll\Omega$) this dependence is linear, $\Omega_{2,3}(H)=\Omega(0)\pm(\chi_\parallel/2\chi_\perp)gH$, in agreement with experimental data.[8]

When the hexagonal anisotropy is taken into account, the linearized equations of motion become equations with variable coefficients, and the spin–wave spectrum becomes a band spectrum. A gap appears in the spectrum of the lower (Goldstone) branch. Two branches arise in the scheme of reduced bands, with dispersion relations (for $H=0$)

$$\Omega_1^{(1)}(v)=\frac{gM_0}{\chi_\parallel^{1/2}}\left\{\alpha_\perp\mathbf{k}_\perp^2+\frac{18\rho s'^2}{s^2}cd^2(v;s')\right\}^{1/2},$$

$$\Omega_1^{(2)}(v)=\frac{gM_0}{\chi_\parallel^{1/2}}\left\{\alpha_\perp\mathbf{k}_\perp^2+\frac{18\rho}{s^2}dc^2(v;s')\right\}^{1/2}, \tag{10}$$

where $cd(v;s')=dc^{-1}(v;s')=cn(v;s')dn^{-1}(v;s')$ and v is a dimensionless parameter satisfying $0\leqslant v\leqslant K(s')$. From (10) we find that the width of the band gap is proportional to $\rho^{1/2}$. For the system of equations for the variables $\xi(\mathbf{r},t)$ and $\eta(\mathbf{r},t)$, on the other hand, a potential of the type $\sin 5\zeta_0(z)$ appears. This potential makes it impossible to solve these equations analytically, so numerical calculations are required. Such calculations go beyond the scope of the present study.

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