Antiferromagnetic resonance in cubic crystals

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The effect of magnetoelastic interaction on the spin-wave spectrum is investigated theoretically for a cubic antiferromagnet in the spin-flop state. The analytical expressions obtained for the variation of the antiferromagnetic resonance frequency with the direction of the external field, in planes (100) and (110), are in satisfactory agreement with the results of experimental investigations of AFMR in the garnets CrGeG and FeGeG [V. I. Sokolov and O. I. Shevaleevskiĭ, Zh. Eksp. Teor. Fiz. 72, 2367 (1977) (this issue) Sov. Phys. JETP 45, 1245 (1977)]. It is found that in cubic antiferromagnets, in contrast to uniaxial, the magnetoelastic contribution to the spin-wave gap is large; it is comparable with the contribution from the magnetocrystalline anisotropy. The paper also discusses the question of the Dzyaloshinskiĭ interaction in cubic crystals.

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In cubic magnetic materials the role of magnetoelastic interaction is much more significant than in uniaxial. This is evident if only from the fact that the ration of the magnetostrictive energy to the anisotropy energy is much larger in the former than in the latter (see, for example, Ref. 1). Nevertheless, the question of the influence of magnetoelastic interaction on the spin-wave spectrum of cubic ferro- and antiferromagnets has not so far been investigated theoretically. In uniaxial magnets, the magnetoelastic contribution to the spectrum is perceptible only near phase-transition points, where the main gap, due to magnetocrystalline anisotropy, vanishes (in one of the branches of the spectrum).[2–4] In cubic magnetic crystals, however, the magnetoelastic part of the spectral gap can, it would seem, be detected even against the background of the "large" anisotropic part (due to the constant K_1) of the gap—that is, far from phase-transition points—in the low-frequency branch, and even in the high-frequency branch, of the spectrum of a cubic antiferromagnet.

The theoretical and experimental investigation of cubic antiferromagnets (without allowance for magnetoelastic interaction) has been the subject of a quite large number of papers,[5–8,1] in which crystals of the perovskite-type structure were studied. Eastman[9] investigated the dependence of the antiferromagnetic resonance (AFMR) frequency of a cubic $RbMnF_3$ crystal on uniaxial stresses, but he neglected the effect of the spontaneous magnetostrictive deformations in comparison with those produced by the external pressure (probably because he supposed that the effect of magnetostriction on the AFMR frequency reduces merely to renormalization of the anisotropy constant). In the work of Belov *et al.*,[10] in which AFMR was first observed in garnets, in order to interpret the experimental data in the high-frequency branch of the spectrum, there was introduced empirically into the formula for the AFMR frequency (obtained in Ref. 5) an isotropic energy gap that was, according to the authors' assumption, of magnetoelastic origin. This enabled them to give a qualitative explanation of the experiment.

Because of the situation described above, we have carried out a calculation of the spin-wave spectrum in cubic antiferromagnets with allowance for magnetoelastic interaction. Also discussed is the effect on the AFMR spectrum of the Dzyaloshinskiĭ interaction and of the second anisotropy constant. Antiferromagnets are considered in which, in the absence of a field, the easy directions for the antiferromagnetism vector are both the axes of [100] type and the axes of [111] type. The analytical dependences of the AFMR spectrum on the direction of the external field are obtained in the (100) and (110) planes. Our theoretical results are in satisfactory agreement with experimental investigations of AFMR in garnets with positive and with negative anisotropy constants, made by Sokolov and Shevaleevskiĭ.[11]

The phenomenological Hamiltonian of a cubic antiferromagnet has the following form;

$$\mathcal{H} = \mathcal{H}_m + \mathcal{H}_{me} + \mathcal{H}_e,$$

where

$$\mathcal{H}_m = Am^2 + K_1(l_x^2 l_y^2 + l_x^2 l_z^2 + l_y^2 l_z^2) + K_2 l_x^2 l_y^2 l_z^2 - \frac{D}{2}\sum_{i\neq j\neq k}[\mathbf{m}\times\mathbf{l}]_i l_j l_k - \mathbf{mh};$$

$$\mathcal{H}_{me} = b_1(l_x^2 u_{xx} + l_y^2 u_{yy} + l_z^2 u_{zz}) + b_2(l_x l_y u_{xy} + l_x l_z u_{xz} + l_y l_z u_{yz});$$

$$\mathcal{H}_e = {}^1/_2 c_{11}(u_{xx}^2 + u_{yy}^2 + u_{zz}^2) + c_{12}(u_{xx}u_{yy} + u_{xx}u_{zz} + u_{yy}u_{zz}) + {}^1/_2 c_{44}(u_{xy}^2 + u_{xz}^2 + u_{yz}^2).$$

Here $\mathbf{m} = (\mathbf{M}_1 + \mathbf{M}_2)/2M_0$ and $\mathbf{l} = (\mathbf{M}_0 - \mathbf{M}_2)/2M_0$ are the dimensionless ferromagnetism and antiferromagnetism vectors; $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations; $M_0 = M_1 = M_2$; A is the exchange-interaction parameter; K_1 and K_2 are the first and second anisotropy constants; D is the Dzyaloshinskii-interaction parameter; $\mathbf{h} = 2M_0\mathbf{H}$, where $\mathbf{H}$ is the external magnetic field; b_1 and b_2 are magnetoelastic-interaction constants; c_{11}, c_{12}, and c_{44} are the elastic moduli; u_{ij} is the strain tensor; and $i, j, k = x, y, z$. We note that in cubic antiferromagnets, weak ferromagnetism (not yet observed experimentally in such structures) is in principle possible only in crystals belonging to the crystallographic groups T_d, O, and O_h (but not to groups T and T_h).

The equilibrium values of the strain tensor have the form

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$$u_{ii}^0=b_1[c_{11}-\alpha_i^2(c_{11}+2c_{12})]/(c_{11}-c_{12})(c_{11}+2c_{12})$$

(here there is no summation over i),

$$u_{ij}^0=-b_2\alpha_i\alpha_j/c_{44}\quad(i\neq j),$$

where α_i are the direction cosines of the equilibrium value of the vector **l**.

In calculation of the AFMR spectrum, we shall set $u_{ij}=u_{ij}^0$, since we are considering uniform precession with wave vector zero. We shall suppose that the external field **H** is strong enough to flip the sublattices into the spin-flop plane ($h^2>4AK_1$).

The directions of the external field and of the sublattice magnetizations with respect to the crystallographic axes of the cubic crystal are conveniently described by the following angles (see Fig. 1); θ and φ, the azimuthal and polar angles of the external field; ξ, the angle determining the position of the antiferromagnetism vector in the spin-flop plane; and η, the angle of bending together of the sublattices ($\sin\eta=m$, not shown in Fig. 1).

On the assumption that $A\gg K_1$, K_2, we have

$$\sin\eta=\{h+D|p_1\alpha_1(\alpha_2^2-\alpha_3^2)+\text{c.p.}|\}/2A,\qquad(1)$$

where $p_1=\sin\theta\cos\varphi$, $p_2=\sin\theta\sin\varphi$, and $p_3=\cos\theta$ are the direction cosines of the external field, and where

$$\alpha_1=\sin\xi\sin\varphi-\cos\xi\cos\theta\cos\varphi,$$
$$\alpha_2=-\sin\xi\cos\varphi-\cos\xi\cos\theta\sin\varphi,\quad \alpha_3=\cos\xi\sin\theta;$$

c.p. denotes cyclic permutation of the indices. The absolute-value sign in (1) is determined by the fact that the symmetry operation C_2 and C_4 connects crystallographic sites of different magnetic sublattices (in these operations, in particular, **l** changes sign).

To find the AFMR spectrum, we shall apply the well-known method of solution of the Landau-Lifshitz equations for small oscillations of the magnetic moments of the sublattices. In the general case of an arbitrary direction of the external field, it is impossible to obtain analytic dependences of the frequencies on θ and φ. We shall therefore limit ourselves to consideration of the most interesting special cases, in which **H** lies in the planes (100) and (110). We shall also suppose that the constant

$$\tilde{K}_1=K_1+\frac{b_1^2}{c_{11}-c_{12}}-\frac{b_2^2}{2c_{44}}$$

is much larger than K_2. We shall present formulas for the AFMR frequencies in these cases for different signs of the constant $\tilde{K}_1$, keeping only terms that are amplified by the exchange or external field. The following notation will be introduced in the formulas: γ, the gyromagnetic ratio; $H_E=A/M_0$; $H_{A1}=2K_1/M_0$ for $\tilde{K}_1>0$ and $H_{A1}=-4K_1/3M_0$ for $\tilde{K}<0$; $H_{A2}=4K_2/M_0$; $H_1=4b_1^2/M_0(c_{11}-c_{12})$; $H_2=2b_2^2/M_0c_{44}$; $H_D=D/2M_0$. Because the anisotropy field H_{A1}, as accepted in the literature, is defined differently for positive and for negative values of $\tilde{K}_1$, a constant q is introduced: $q=2$ for $\tilde{K}_1>0$ and $q=-3$ for $\tilde{K}_1<0$.

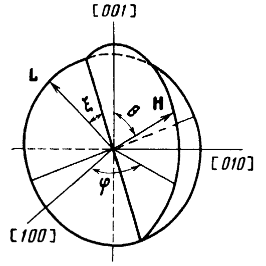

FIG. 1. Orientation of the antiferromagnetism vector $L=2M_0\mathbf{l}$ in the spin-flop state, for arbitrary direction of the external magnetic field **H**.

I. Constant $\tilde{K}_1>0$

A. The field **H** lies in the plane (001)($\theta=90°$). a) In the case $\sin\xi=0$, a stable state;

$$(\omega_1/\gamma)^2=H^2+qH_EH_{A1}+H_EH_1,\qquad(2)$$

$$(\omega_2/\gamma)^2=qH_EH_{A1}+H_EH_1;\qquad(3)$$

b) in the case $\cos\xi=0$, a metastable state;

$$(\omega_1/\gamma)^2=H^2+qH_EH_{A1}\cos4\varphi+{}^1/_2H_E(H_1+H_2)+{}^1/_2H_E(H_1-H_2)\cos4\varphi+{}^5/_2HH_D|\sin2\varphi|,\qquad(4)$$

$$(\omega_2/\gamma)^2={}^1/_4qH_EH_{A1}(3+\cos4\varphi)+{}^1/_8H_EH_{A2}(1-\cos4\varphi)+{}^1/_2H_E(H_1+H_2)+{}^1/_4H_E(H_1-H_2)(1+\cos4\varphi)+{}^7/_2HH_D|\sin2\varphi|.\qquad(5)$$

B. The field **H** lies in the plane (110) ($\varphi=45°$).[1)]

1) In the case $0°\leqslant\theta\leqslant54.7°$ ($\theta=54.7°$ corresponds to the [111] axis), $\sin^2\xi=(1+\sin^2\theta)/(2+\sin^2\theta)$, and we have

$$(\omega_1/\gamma)^2=H^2+qH_EH_{A1}\cos^2\xi(1-3\sin^2\theta)(2-\sin^2\theta)+{}^1/_2H_E(H_1+H_2)+{}^1/_2H_E(H_1-H_2)\cos^2\xi(2-7\sin^2\theta+2\sin^4\theta)+HH_D|\sin\xi|\cos^2\xi\sin\theta[14(1-\sin^2\theta)-5\sin^4\theta],\qquad(6)$$

$$(\omega_2/\gamma)^2=qH_EH_{A1}\cos^2\xi(2-3\sin^2\theta)(1+\sin^2\theta)+{}^1/_2H_E(H_1+H_2)+{}^1/_2H_E(H_1-H_2)(1-2\sin^2\theta)+HH_D|\sin\xi|\cos^2\xi\sin\theta(14-8\sin^2\theta-3\sin^4\theta).\qquad(7)$$

2) When $54.7°\leqslant\theta\leqslant90°$, in the case $\sin\xi=0$, a stable state:

$$(\omega_1/\gamma)^2=H^2+{}^1/_2qH_EH_{A1}(1-11\sin^2\theta+12\sin^4\theta)+{}^1/_2H_E(H_1+H_2)+{}^1/_2H_E(H_1-H_2)\sin^2\theta(6\sin^2\theta-5),\qquad(8)$$

$$(\omega_2/\gamma)^2={}^1/_2qH_EH_{A1}(3\sin^2\theta-2)(1+\sin^2\theta)+{}^1/_2H_E(H_1+H_2)+{}^1/_2H_E(H_1-H_2)(-7+11\sin^2\theta-3\sin^4\theta);\qquad(9)$$

in the case $\cos\xi=0$, a metastable state;

$$(\omega_1/\gamma)^2=H^2+{}^1/_2qH_EH_{A1}(1-\sin^2\theta)+{}^1/_2H_E(H_1+H_2)-{}^1/_2H_E(H_1-H_2)\sin^2\theta+{}^1/_2HH_D\sin\theta(1-6\sin^2\theta),\qquad(10)$$

$$(\omega_2/\gamma)^2={}^1/_2qH_EH_{A1}(3\sin^2\theta-2)+{}^1/_2H_E(H_1+H_2)+{}^1/_2H_E(H_1-H_2)(\sin^2\theta-1)+{}^1/_2HH_D\sin\theta(6\sin^2\theta-13).\qquad(11)$$

II. Constant $\tilde{K}_1<0$

A. The field **H** lies in the plane (001) ($\theta=90°$). Here $\sin^2\xi=4/(7+\cos4\varphi)$, and we have

$$(\omega_1/\gamma)^2=H^2+qH_EH_{A1}\sin^2\xi\cos4\varphi+{}^1/_8H_EH_{A2}\sin^6\xi(11+5\cos4\varphi)+{}^1/_2H_E(H_1+H_2)+{}^3/_2H_E(H_1-H_2)\sin^2\xi(\cos4\varphi-1)+{}^1/_2HH_D\sin^{3/2}\xi|\sin2\varphi|(\cos4\varphi-2),\qquad(12)$$

$$(\omega_2/\gamma)^2=-\frac{q}{2}H_EH_{A1}\sin^2\xi(3+2\cos4\varphi)-{}^1/_{128}H_EH_{A2}\sin^6\xi(1-\cos4\varphi)(43+16\cos4\varphi+5\cos^24\varphi)+H_EH_2+{}^1/_2HH_D\sin^{3/2}\xi|\sin2\varphi|(13+4\cos4\varphi).\qquad(13)$$

B. The field **H** lies in the plane (110) ($\varphi = 45°$).

1) The range $0° \leq \theta \leq 54.7°$. The frequencies for the stable and for the metastable states are determined, respectively, by formulas (8)–(9) and by formulas (10)–(11), with $q = -3$.

2) The range $54.7° \leq \theta \leq 90°$. The frequencies are determined by formulas (6)–(7), with $q = -3$.

From the results presented, it is evident that the magnetoelastic contribution to the AFMR frequency consists of an isotropic and an anisotropic part; the latter vanishes on passage to an isotropic magnet (together with terms due to magnetocrystalline anisotropy).

As was mentioned above, Sokolov and Shevaleevskiĭ[11] have carried out an experimental investigation of the high-frequency branch (ω_1) of AFMR in the garnets CrGeG ($\tilde{K}_1 > 0$) and FeGeG ($\tilde{K}_1 < 0$). Measurements were made for all cases of **H** and **l** orientation discussed above. Within the limits of experimental accuracy, good agreement of theory with experiment is obtained with the following values of the effective exchange, anisotropy, and magnetostrictive fields. For CrGeG ($H_E = 250$ kOe); $H_{A1} = 38.1$ Oe, $H_1 = 15$ Oe, $H_2 = 11.2$ Oe; for FeGeG ($H_E = 404$ kOe): $H_{A1} = 15.3$ Oe, $H_1 = 3.6$ Oe, $H_2 = 1.3$ Oe. In contrast to uniaxial antiferromagnets, for example hematite, where the magnetostrictive field $H_{ms} \approx 0.7$ Oe whereas $H_A = 10^3$ Oe, in garnets the anisotropy and magnetostrictive fields are almost commensurate in value.

The contribution to the AFMR spectrum from the Dzyaloshinskiĭ interaction has a unique angular dependence, which offers a possibility in principle of experimental determination of the value of this interaction. But the experimental accuracy in Ref. 11 was insufficient for drawing a definite conclusion regarding the presence of weak ferromagnetism in the cubic crystals investigated. We note that the question of the contribution of the weakly-ferromagnetic invariant to the AFMR frequency of a cubic anitferromagnet was considered by Guseinov[12]; his results, however, do not agree with ours.

The low-frequency branch of the AFMR (ω_2) is independent of the value of H (when $H > \sqrt{|q| H_E H_{A1}}$) but depends on its direction. We note that if **H** is directed along an axis of the [111] type, then for either sign of $\tilde{K}_1$ (with neglect of K_2), without allowance for magnetostriction, $\omega_2 = 0$. The presence of magnetoelastic interaction in this case may lead to a change of the dispersion law for elastic oscillations and to a sharp decrease of the velocity of sound in the vicinity of this state. This effect, for noncubic magnets (uniaxial and biaxial), was considered in Refs. 4.

In our work no account has been taken of hyperfine interaction, which, as is well known,[5] gives an isotropic addition $2H_E H_N$ to both frequency branches. Apparently the hyperfine gap in the AFMR spectrum becomes important either in antiferromagnets with Mn^{2+} ions[6,13] or at $T \lesssim 1$ K. In this connection we remark that in experiments on crystals with perovskite structure[5–8,1] and containing Mn^{2+} ions (for example $RbMnF_3$, $TlMnF_3$), the isotropic part of the spectral gap was considered to be caused solely by hyperfine interaction, whereas it might be the sum of nuclear and magnetoelastic contributions.

[1)]In this case we completely neglected the second anisotropy constant K_2, because when H ∥ [111] the expansion in powers of $(K_2/\tilde{K}_1)$ that we used in the previous case (H in the plane (001)) is not valid. Allowance for K_2 gives corrections of order $H_E H_{A2}$ in $(w_{1,2}/\gamma)^2$; the exact formulas are too unwieldy to be presented here.

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