STRENGTHENING OF MAGNETOELASTIC AND MAGNETOELECTRIC INTERACTIONS IN FERROELECTROANTIFERROMAGNETICS WITH ORTHORHOMBIC SYMMETRY

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Interaction of spin waves with elastic and ferroelectric waves in ferroelectroantiferromagnetics with orthorhombic symmetry is considered. The possibility of exchange strengthening the parameters of magnetoelastic and magnetoelectric interactions for certain magnitude and orientation of an external magnetic field with respect to the crystallographic axes is demonstrated.

In recent years there has been a growing interest in the study of ferroelectroantiferromagnetics (crystals with the perovskite structure) in which the antiferromagnetic and the ferroelectric long-range orders can coexist [1-3]. The interest was primarily caused by the quite attractive prospects of using these materials in modern electronics [4].

In the present paper we will study the possibility of exchange strengthening the parameters of the magnetoelastic and magnetoelectric coupling in antiferromagnetic structures with orthorhombic symmetry (space group \( D_{2h} \)).

We describe the system by the Hamiltonian

\[
\mathcal{H} = \mathcal{H}_M + \mathcal{H}_{EI} + \mathcal{H}_F + \mathcal{H}_{M-EI} + \mathcal{H}_{MF} + \mathcal{H}_{F-EI},
\]

where the energies of the magnetic \( (M) \), elastic \( (E) \), and ferroelectric \( (F) \) parts of the system as well as their interaction energy are included.

We assume the magnetic subsystem to consist of two mirror magnetic sublattices, and in the presence of an external constant homogeneous magnetic field \( (H_0) \) we choose the magnetic energy to have the form

\[
\mathcal{H}_M = \int d\mathbf{r} \left\{ \frac{1}{2} \alpha_{ijmn}^\alpha (\nabla_i M_j^\alpha)(\nabla_m M_n^\alpha) + I_{ij}^{\alpha\beta} M_i^\alpha M_j^\beta - H_{ij}^\alpha M_j^\alpha \right\},
\]

where \( i, j, m, n = x, y, z; \alpha, \beta = 1, 2; \alpha_{ijmn}^\alpha \) is the tensor of the inhomogeneous exchange interaction; the tensor \( I_{ij}^{\alpha\beta} = \delta_{ij}^{\alpha\beta} + \beta_{ij}^{\alpha\beta} \) includes the tensors of the exchange \( \delta_{ij}^{\alpha\beta} \) and relativistic \( \beta_{ij}^{\alpha\beta} \) interactions,

\[
\nabla_i M_j^\alpha = \frac{\partial M_j^\alpha}{\partial r_i}.
\]

The energy of the elastic subsystem is written in the form

\[
\mathcal{H}_{EI} = \frac{1}{2} \int d\mathbf{r} \left\{ \rho u^2 + \Lambda_{ijmn} u_{ij} u_{mn} \right\},
\]

where \( \Lambda_{ijmn} \) is the tensor of the elasticity modules and \( u_{ij} \) is the strain tensor.
The ferroelectric subsystem is described by the Hamiltonian

$$H_F = \frac{1}{2} \int d\mathbf{r} \left\{ \frac{4\pi}{\lambda} \left[ p^2 + s_{ijmn}(\nabla_j p_i)(\nabla_n p_m) \right] + \kappa_{ij} p_i p_j \right\},$$

where $p$ is the deviation of the polarization vector from its equilibrium value, $\kappa_{ij}$ is the tensor of the inverse dielectric susceptibility, $\lambda$ is the square of the frequency of the longitudinal optical phonons at $k \to 0$.

To describe the interactions of the subsystems we consider:
- the magnetoelastic interaction energy
  $$H_{M-E1} = \int d\mathbf{r} \{ \lambda_{ijmn}^{\alpha\beta} M_i^\alpha M_j^\beta u_{mn} \},$$
  where $\lambda_{ijmn}^{\alpha\beta}$ is the magnetostriction tensor;
- the magnetoelectric interaction energy
  $$H_{MF} = \int d\mathbf{r} \{ a_{ijm}^{\alpha\beta} p_i M_j^\alpha M_m^\beta \},$$
  where $a_{ijm}^{\alpha\beta}$ is the tensor of the relativistic magnetoelectric interaction;
- the electroelastic interaction energy
  $$H_{E-E1} = \int d\mathbf{r} v_{ijmn}(\nabla_j p_i) u_{mn},$$
where $v_{ijmn}$ is the tensor responsible for the relation between the polarization and strain inhomogeneities.

We write the Hamiltonian in the representation of the approximate secondary quantization. To this end we express the magnetic moments of the sublattices $M^\alpha$ in terms of Golstein–Primakoff's operators $a_\alpha^\dagger$, $a_\alpha$, the vector of elastic displacements $u$ in terms of the phonon creation and annihilation operators $b_\omega^\dagger$, $b_\omega$ (see, e.g., [5]), and we represent the deviation of the polarization vector from its equilibrium value in the form

$$p_i = \left( \frac{\lambda}{8\pi V} \right)^{1/2} \sum_{k\delta} \frac{\varepsilon_{i\delta}^k}{\Omega_\delta^{1/2}} \left( d_{k\delta} e^{ikr} + d_{k\delta}^+ e^{-ikr} \right),$$

where $\varepsilon_{i\delta}^k$ is the unit polarization vector of segnetons, $\Omega_\delta \equiv \Omega_{k\delta}$ is the energy of segnetoelectric excitations, $\delta$ is the polarization index of transverse oscillations. Then, to diagonalize the Hamiltonian of the magnetic subsystem we use Bogolyubov's canonical transformation

$$a_{k\alpha} = u_{\alpha\gamma} c_{k\gamma} + v_{\alpha\gamma} c_{-k\gamma}^+, \quad a_{\alpha} = V^{-1/2} \sum_k a_{k\alpha} e^{ikr},$$

where the functions $u$, $v$ are calculated in [6].

Then Hamiltonian (1) can be written in the form

$$\hat{H} = \sum_{k\gamma} \varepsilon_{\gamma} c_{k\gamma}^+ c_{k\gamma} + \sum_{k\delta} \omega_\delta b_{k\delta}^+ b_{k\delta} + \sum_{k\delta} \Omega_\delta d_{k\delta}^+ d_{k\delta}$$

$$\quad + \left\{ \sum_{k\gamma\delta} \Gamma_{k\gamma\delta}^{M-E1} c_{k\gamma}(b_{-k\delta} - b_{k\delta}^+) + \sum_{k\gamma\delta} \Gamma_{k\gamma\delta}^{MF} c_{k\gamma}(d_{-k\delta} - d_{k\delta}^+) \right\}$$

$$\quad + \sum_{k\delta} \Gamma_{k\delta}^{E-E1} d_{k\delta}^+(b_{-k\delta} - b_{k\delta}^+) + \text{h.c.},$$

where $\Gamma_{k\gamma\delta}$ are the matrix elements of the relevant three-body interactions in the Brillouin zone.
where $\varepsilon_\gamma(\gamma = 1, 2), \omega_s(s = 1, 2, 3)$ are the energies of the corresponding branches of the spin and elastic waves. The parameters of magnetoelastic and magnetoelectric interactions are defined by the expressions

$$
\Gamma_{k\ell}^M = i \sum_{\alpha, \beta} \left( \frac{2}{\rho\omega_s} \right)^{1/2} \lambda_{ijmn} (C_{ij}^{\beta \gamma} u_{\beta\gamma} + C_{ij}^{\alpha \beta} v_{\beta\gamma}) e_{km}^\ell e_n
$$

(here $e_{km}^\ell$ is the unit vector of the phonon polarization), and

$$
C_{ij}^{\beta \gamma} = (\mu M_0^3)^{1/2} e_{ij}^\beta e_{ij}^\gamma
$$

where $e_{ij}^\beta, e_{ij}^\gamma$ are the coefficients of transformation of the operators $M_j^\beta$ to the proper representation, which can be chosen in the form

$$
e_{ij}^\beta = M_{ij}^\beta / M_0, \quad e_{ij}^\gamma = \frac{1}{\sqrt{2}} (e_{ij}^\gamma + ie_{ij}^\gamma),
$$

$$
e_{ij}^{\perp} (H_0, M_0^3), \quad e_{ij}^\gamma = [e_{ij}^\gamma, e_{ij}^\gamma],
$$

$$
\Gamma_{k\ell}^M = - \left( \frac{\mu M_0^3}{4\pi \omega_s} \right)^{1/2} \sum_{\alpha, \beta} \left[ i a_{ij}^{\alpha \beta} (u_{\beta\gamma} - v_{\beta\gamma}) + a_{ij}^{\alpha \beta} (u_{\beta\gamma} + v_{\beta\gamma}) \right] e_{kJ}^\ell,
$$

$$
a_{ij}^{\alpha \beta} = \sum_{lm} a_{ijlm} e_{ij}^\alpha e_{lm}^\beta, \quad l, q = 1, 2, 3.
$$

The parameter of quasielastic coupling has a simple form:

$$
\Gamma_{k\ell}^M = - \left( \frac{\lambda}{16\pi \rho\omega_s} \right)^{1/2} \sum_{ijklm} e_{klm}^\ell e_{kmn}^\ell e_{nkl}^\ell.
$$

Diagonalizing Hamiltonian (2) by means of the canonical transformation

$$
c_{k\gamma} = U_{\gamma f} \alpha_{k f} + V_{\gamma f} \alpha_{k f}^\dagger,
$$

$$
b_{k\gamma} = U_{f\gamma} \alpha_{k f} + V_{f\gamma} \alpha_{k f}^\dagger, \quad f = (\gamma, s),
$$

we obtain the following dispersion equation, which determines the spectrum of the coupled magnetoferrohydroelastic waves $E = E_{k\ell}^f$:

$$
\prod_{\delta \gamma} \left( E^2 - \varepsilon_{\gamma s}^2 \right) \prod_{\Omega_s} \left( E^2 - \Omega_s^2 \right) - 4 \prod_{\gamma, \delta} \left| \Gamma_{k\ell}^M \right|^2 e_{\gamma s}^2 \prod_{\ell \neq s} \left( E^2 - \Omega_\delta^2 \right) \times \left( E^2 - \varepsilon_{\gamma s}^2 \right) \prod_{\gamma, \delta} \left( E^2 - \varepsilon_{\gamma s}^2 \right) \prod_{\Omega_s} \left( E^2 - \Omega_s^2 \right) - 4 \prod_{\delta \s} \left| \Gamma_{k\ell}^M \right|^2 e_{\delta s}^2 \prod_{\gamma, \delta} \left( E^2 - \varepsilon_{\gamma s}^2 \right) \prod_{\Omega_s} \left( E^2 - \varepsilon_{\gamma s}^2 \right) (E^2 - \Omega_{\delta s}) = 0.
$$

The picture of a spectrum of coupled magnetoferrohydroelastic waves in a particular case is shown in Fig. 1.

The interaction of the corresponding branches of elementary excitations outside the resonance region is weak, and this is taken into consideration in calculating transformation coefficients (5). Then an approximate calculation of the functions $U, V$ with consideration for the resonance interaction regions can be carried out. Thus, in the region of resonance interaction of the spin and elastic waves $|\varepsilon_{\gamma s} - \omega_s| \lesssim |\Gamma_{k\ell}^M|$, with the energies $\varepsilon_{\gamma s}$ and $\omega_s$ respectively we find

$$
U_{\gamma s} = (\delta_{gs} - \delta_{gs}) P_g \Gamma_{k\ell}^M \omega_s (E_g + \varepsilon_{\gamma s}),
$$

$$
U_{s g} = -(\delta_{gs} - \delta_{gs}) P_g (E_g + \omega_s) (E_g^2 - \varepsilon_{gs}^2)/2,
$$

$$
V_{\gamma s} = (E_g - \varepsilon_{gs})(E_g + \varepsilon_{gs})^{-1} U_{\gamma s}, \quad V_{s g} = (E_g - \omega_s)(E_g + \omega_s)^{-1} U_{s g},
$$

$$
g = 1, 2,
$$

$$
P_g^{-1} = \{ E_g \omega_s [4|\Gamma_{k\ell}^M |^2 |\varepsilon_{\gamma s} + (E_g^2 - \varepsilon_{gs}^2)^2] |1/2, \quad E_{\gamma, s} = 1/2 \{ \varepsilon_{\gamma s} + \omega_s \pm \sqrt{(|\varepsilon_{\gamma s} - \omega_s|^2 + |\Gamma_{k\ell}^M|^2)} \}.
$$

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1. With consideration for the system symmetry we study the case of the antiferromagnetic of the “easy 
XOY plane” type when $H_0 \parallel z_0$,

\[ 0 < H_0 < 2\delta M_0 \equiv H_\delta, \quad (\delta/2 = \delta_{12}^{12} \approx \delta_{zz}^{12} \approx \delta_{yy}^{12}), \]
\[ \varepsilon_{z_0}^{zz} \equiv \cos \theta \approx H_0/H_\delta, \quad \varepsilon_{z_0}^{0} = 0, \quad \varepsilon_{y_0}^{zz} = (\delta_{1a} - \delta_{2a})\sin \theta. \]

1°. Consider the interaction of spin and elastic waves. In accordance with the direction of the wave 
vector $k$ following Eq. (3), in particular cases we find (here and in what follows we omit the coupling 
coefficients, which in our approximation are equal to zero): 

a) $k||z_0$

\[ \Gamma_{1t_1}^{M-EI} = 2i k \gamma_{t_1} (\lambda_{zzzz}^{11} + \lambda_{zzzz}^{12}) \cos \theta(u_{11} + v_{11}), \]
\[ \Gamma_{1t_2}^{M-EI} = -2i k \gamma_{t_2} (\lambda_{yyzz}^{11} \cos 2\theta + \lambda_{yyzz}^{12})(u_{11} - v_{11}), \]
\[ \Gamma_{2t_1}^{M-EI} = k \gamma_{t_1} (\lambda_{yyzz}^{11} - \lambda_{zzzz}^{11} - \lambda_{zzzz}^{12} - \lambda_{zzzz}^{12}) \sin 2\theta(u_{22} - v_{22}), \]
\[ \gamma_s = \left( \frac{\mu M_0^2}{\rho \omega_s} \right)^{1/2}, \quad s = l, t_1, t_2. \]

Since $u_{11} - v_{11} \approx (\mu M_0^2/\varepsilon_1)^{1/2}$, where $\varepsilon_1$ is the spin wave branch of the acoustic type, the parameter of 
interaction of the spin wave lower-lying branch with the second transverse elastic wave $\Gamma_{1t_2}^{M-EI}$ turns out to 
be $\delta^{1/2} \approx 30$ times enhanced in an exchange way[7]; 

b) $k||x_0$

no exchange strengthening of the magnetostriction takes place; 

c) $k||y_0$

\[ \Gamma_{1t_1}^{M-EI} = -2i k \gamma_{t_1} (\lambda_{yyzz}^{11} \cos 2\theta + \lambda_{yyzz}^{12})(u_{11} - v_{11}), \]
\[ \Gamma_{2t_2}^{M-EI} = -2i k \gamma_{t_2} (\lambda_{yyzz}^{11} - \lambda_{yyzz}^{12}) \sin \theta(u_{22} + v_{22}), \]
\[ \Gamma_{2t_1}^{M-EI} = k \gamma_{t_1} (\lambda_{yyzz}^{11} - \lambda_{zzzz}^{11} - \lambda_{zzzz}^{12} - \lambda_{zzzz}^{12}) \sin 2\theta(u_{22} - v_{22}); \]

here we have an exchange-enhanced interaction of the spin wave first branch with the first transverse elastic wave. 

In the absence of magnetoelastic and electroelastic interactions and bearing in mind that in the case of $k||y_0$ the coefficient of the magnetoelastic coupling $\Gamma_{1t_1}^{M-EI}$ is strengthened by the parameter of exchange 
interaction $\delta$, the approximate solutions of dispersion equation (6) at a distance from and near the region of
magnetoacoustic resonance can be represented as

\[ E_1 \cong \omega_1 \left(1 + \frac{2\varepsilon_1^2}{\omega_1^2 - \varepsilon_1^2} \zeta_{M-EI}^2\right), \quad E_2 \cong \varepsilon_1 \left(1 - \frac{2\omega_1^2}{\omega_1^2 - \varepsilon_1^2} \zeta_{M-EI}^2\right), \]

\[ E_1 \cong \omega_1(1 + \zeta_{M-EI}), \quad E_2 \cong \varepsilon(1 - \zeta_{M-EI}), \]

(9)

where \( \lambda' \) is the order of the components of the tensor \( \lambda_{ijnm}^{(3)} \). Thus, the difference between the coupled magnetoelastic wave and the corresponding free oscillation near the resonance is proportional to the dimensionless coupling parameter \( \zeta_{M-EI}^2 \), whereas at a distance from the resonance this difference is proportional to the square of the coupling parameter \( \zeta_{M-EI}^2 \). Using the values of the parameters \( \rho \approx 10 \text{ g/cm}^3, M_0 \approx 10^3 \text{ Gs}, \lambda' \approx 10 [8], \) and \( \delta \approx 10^3 [7] \) (typical of perovskites) near the resonance, when \( \varepsilon_1 \approx \omega_1 \approx 10^{12} \text{ s}^{-1}, k \approx 10^7 \text{ cm}^{-1}, \) we obtain \( \zeta_{M-EI} \approx 10^{-2} \).

\( \text{2°} \). Consider the interaction of the magnetic and ferroelectric subsystems defined by expression (4). We study the following particular cases:

a) \( k \parallel z_0 \)

\[ \Gamma_{11}^{MF} = -\psi_1(a_{1zz}^{11} + a_{2zz}^{12})(u_{11} + v_{11}), \]

\[ \Gamma_{12}^{MF} = -i\psi_2(a_{yzy}^{11} + a_{yzy}^{12})(u_{11} - v_{11}), \]

\[ \psi_n \equiv \left(\frac{\mu M_0^3 \lambda}{\pi \Omega_n}\right)^{1/2}, \quad n = 1, 2. \]

(10)

We see that the coupling of the lower spin branch with the second ferroelectric branch is \( \delta^{1/2} \approx 30 \) times strengthened in an exchange way. Note that a similar effect of exchange strengthening of the magnetoelectric coupling was for the first time obtained in [8, 9].

b) \( k \parallel x_0 \)

\[ \Gamma_{11}^{MF} = -i\psi_1(a_{1yz}^{11} + a_{2yz}^{12})(u_{11} + v_{11}), \]

\[ \Gamma_{12}^{MF} = -i\psi_2(a_{yzy}^{11} + a_{yzy}^{12})(u_{11} - v_{11}), \]

\[ \Gamma_{22}^{MF} = -\frac{i}{2}\psi_2(a_{1zz}^{11} + a_{2zz}^{12}) \sin 2\theta(u_{22} - v_{22}). \]

(11)

In this case we observe an exchange strengthening of the coupling of the lower lying branches of the spin wave with both ferroelectric branches.

c) \( k \parallel y_0 \)

\[ \Gamma_{11}^{MF} = -i\psi_1(a_{1xz}^{11} + a_{2xz}^{12})(u_{11} - v_{11}), \]

\[ \Gamma_{12}^{MF} = -i\psi_2(a_{xzy}^{11} + a_{xzy}^{12})(u_{11} + v_{11})\cos \theta, \]

\[ \Gamma_{21}^{MF} = -\frac{i}{2}\psi_1(a_{1zz}^{11} + a_{2zz}^{12}) \sin 2\theta(u_{22} - v_{22}). \]

(12)

The interaction of the first branches of the spin and ferroelectric waves is strengthened in an exchange way.

Note that in strong fields \( H_0 > H_0 \), when the equilibrium magnetic parameters \( M_0^\alpha \) are oriented along the field, no exchange strengthening of the magnetoelastic and magnetoelectric couplings occurs, which is quite easily explained. As one can see from the above formulas, the possibility of exchange strengthening one or another coefficient of the magnetoelastic or magnetoelectric coupling depends on the combination of the \( u, v \)-transformation coefficients. The absence of the \( u, v \)-coefficients in the parameters of the above-mentioned couplings in the ferroelectroferromagnetics makes the exchange strengthening of coupling of the ferroelectric, magnetic, and elastic waves in these alloys impossible (see also [9]).

Another important conclusion is that only the coupling of the lower lying acoustic branch of spin oscillations with the ferroelectric and elastic modes can be exchange strengthened, which is in agreement with...
the data from [8, 9]. Note that the authors of [9] consider the smallness of the energy gap in the acoustic spin branch spectrum as the necessary condition for the exchange strengthening of the magnetoelectric coupling.

The relative corrections to the frequencies of ferroelectric and spin waves are determined (e.g., in the case of $k||y_0$) by the dimensionless parameter of the magnetoelectric coupling $\zeta_{MF}$ (similarly to (9)):

$$\zeta_{MF} = \frac{\left| \Gamma_{11}^{MF} \right|}{(\varepsilon_1 \Omega_1)^{1/2}} \approx \frac{a \mu M_0^2}{\Omega_1 \varepsilon_1} \left( \frac{\delta \lambda}{\pi} \right)^{1/2},$$

where $a$ is the order of the tensor components $a^{ij}_{\mu \nu}$. Substituting into (13) the values of the parameters $M_0 \approx 10^3$ Gs, $a \approx 3 \times 10^{-5}$ Gs$^{-1}$ [8], $\delta \approx 10^3$ [7], $\varepsilon_1 \approx \Omega_1 \approx 10^{12}$ s$^{-1}$, $\lambda \approx 10^{28}$ s$^{-2}$ typical of perovskite, we obtain $\zeta_{MF} \approx 10^{-1}$. Thus, in ferroelectroantiferromagnetics with orthorhombic symmetry the magnetoelectric coupling turns out to be approximately an order of magnitude larger than the magnetoelastic one.

Now if the field $H_0$ is directed along the $OY$ axis, then as a consequence of the anisotropy in the basal plane the exchange strengthening of the interaction of spin waves with elastic and ferroelectric ones occurs only when $2M_0 \sqrt{\delta(\beta_{yy}^2 - \beta_{yy}^1)} < H_0 < H_8$. Corresponding expressions for the coupling coefficients are obtained from formulas (7), (8), (10)–(12) by cyclic replacement of $x$ by $y$, $z$ by $y$, $y$ by $x$.

2. In the case of antiferromagnetic of the "easy Z axis" type when $H_0||z_0$, an exchange strengthening of the magnetoelastic and magnetoelectric interactions takes place only in strong enough fields, when $H_{s\theta} < H_0 < H_8$ ($H_{s\theta} = 2M_0 \sqrt{\delta(\beta_{zz}^1 - \beta_{zz}^2)}$). In this case the corresponding coupling coefficients are similar to the case 1.

As follows from the above consideration, the possibilities of exchange strengthening of magnetoelastic and magnetoelectric interactions in ferroelectroantiferromagnetics depend substantially on both the magnitude and the orientation of the external magnetic field with respect to the crystallographic axes.

REFERENCES


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