

MIXING OF ACOUSTIC WAVES WITH ALTERNATING MAGNETIC FIELD AND PARAMAGNETIC INTERACTION IN ANTIFERROMAGNETS IN THE "FLIPPED" PHASE

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This communication deals with hitherto unknown nonlinear effects that occur in an antiferromagnetic crystal to which electromagnetic and acoustic fields are applied.

1. The nonlinear effects produced in ferromagnets by interaction of electromagnetic and acoustic waves, particularly the Suhl parametric interaction of spin and acoustic waves, are well known. In antiferromagnets, however, in view of the much larger damping (line widths), the situation is essentially different. Parametric excitation of spin waves was observed in antiferromagnetic  $\text{CsMnF}_3$  [1] and  $\text{MnCO}_3$  [2] only recently, while nonlinear effects, that occur when electromagnetic and acoustic waves in antiferromagnets, have to the author's knowledge not been investigated before.

This paper deals with nonlinear interaction of electromagnetic and acoustic waves of commensurate frequencies in "easy-axis" antiferromagnets in the 'flipped' phase, described by a nonlinear magnetization proportional to the alternating field and quadratic in the acoustic wave. The proposed linearity leads, in particular, to two physical effects. The first is parametric amplification (generation) of two acoustic waves propagating opposite each other and induced by a homogeneous alternating magnetic pump field. The other is a mixing of the alternating magnetic field with the acoustic waves to form a nonlinear magnetization whose radiation power is recorded. This nonlinearity can be used to measure the magnon-phonon interaction, which determines the magnitude of the effect. At the same time, the interaction amplitude is determined for definite specified values of the quasimomenta, in contrast to relaxation effects, which contain interaction amplitudes that are integrated with respect to the quasimomenta. The investigated nonlinearity is quite appreciable in the 'flipped' phase because of two factors. The nonlinear response of the system is proportional to the interaction constant and contains frequency-dependent denominators. For antiferromagnets in the canted phase, when the angle  $2\theta$  between the sublattices differs from  $\pi$ , the magnon-phonon interaction for longitudinal acoustic waves is much larger than in the collinear phase; in the former it is determined by exchange interaction, whereas in the latter, where a weak nonrelativistic interaction occurs, the interaction is amplified by a factor  $H_0/M_0$  times for small  $\theta$  and by a factor  $\delta$  for  $\theta$  that are not small;  $H_0 > H_f$ ,  $H_f$  is the flipping field, and  $\delta = k\theta_c/\mu M_0$  is the exchange constant. In the collinear phase the frequency denominators contain the antiferromagnetic-resonance frequency, which is usually relatively large, and in the 'flipped' phase they contain only the frequencies of the external fields for the considered non-activated spin waves (for uniaxial crystals), and the frequency of the spin wave with wave vector on the order of the wave vector on the order of that of the acoustic wave, so that the frequency denominators can be much smaller.

2. We consider the nonlinear magnetization

$$M_{\alpha}^{NL}(k_0, t) = \chi_{\alpha\zeta}^{cd, fg}(\omega, \omega_1, \omega_2, q_1, q_2) H_{\zeta}(\omega, k) S_{cd}(\omega_1, q_1) S_{fg}(\omega_2, q_2) \times \exp[-i(\omega + \omega_1 + \omega_2)t]. \quad (1)$$

Here  $S_{cd}$  is the deformation tensor. Summation over repeated indices is assumed throughout. Following [3], we obtain the magnon-phonon interaction Hamiltonian; it is linear in the phonon operators  $b_q$  and nonlinear<sup>1)</sup> in the spin-wave operators  $c_k$

$$H_{s\zeta} = \sum_{k_1 k_2 k_3 q} A_{1,2,3,q} c_{k_1}^{\dagger} c_{k_2}^{\dagger} c_{k_3} b_q \Delta(k_1 + k_2 - k_3 - q) + \text{c.c.} \quad (2)$$

where  $A_{1,2,3,q} \sim \delta\gamma(q\mu M_0^2 \sin 2\nu/\rho_0\omega_q)$ ,  $\gamma$  is the dimensionless magnetostriction constant, and  $\rho_0$  is the density. Using the procedure of many-time retarded Green's functions (as applied to nonlinear effects [4]), we obtain the following equation for a definite<sup>2)</sup> component of the nonlinearity tensor

$$\begin{aligned}
\chi_{\eta, \zeta}^{cd, fg}(\omega, \omega_1, \omega_2, \mathbf{k}_0, \mathbf{k}, \mathbf{q}_1, \mathbf{q}_2) &= a_1 \frac{\mu M_0}{3\pi^3} \frac{\Delta(\mathbf{k}_0 - \mathbf{k} - \mathbf{q}_1 - \mathbf{q}_2)}{[\omega + \omega_1 + \omega_2 - \omega_s(\mathbf{k}_0)]} \times \\
&\times \left\{ \frac{A_{\mathbf{k}_0, \mathbf{k}, -\mathbf{q}_1 + \mathbf{k}_0 - \mathbf{k}, \mathbf{q}_1}^{cd}}{\omega + \omega_1 - \omega_s(\mathbf{k}) - \omega_s(\mathbf{q}_1 - \mathbf{k} + \mathbf{k}_0)} A_{\mathbf{k}, -\mathbf{q}_1 + \mathbf{k}_0 - \mathbf{k}, \mathbf{q}_2, \mathbf{k}}^{fg} \right\} \times \\
&\times \{ (\omega_1 - \omega_s(\mathbf{q}_1 - \mathbf{k} + \mathbf{k}_0))^{-1} + (\omega - \omega_s(\mathbf{k}))^{-1} \} + \frac{A_{\mathbf{k}, \mathbf{k}_0, \mathbf{q}_1 - \mathbf{k}_0 + \mathbf{k}, -\mathbf{q}_1}^{fg}}{\omega + \omega_2 - \omega_s(\mathbf{k}) - \omega_s(\mathbf{q}_2 - \mathbf{k} + \mathbf{k}_0)} \times \\
&\times A_{\mathbf{k}, -\mathbf{q}_1 + \mathbf{k}_0 + \mathbf{k}, \mathbf{q}_2, \mathbf{k}}^{cd} \{ (\omega_2 - \omega_s(\mathbf{q}_2 - \mathbf{k} + \mathbf{k}_0))^{-1} + (\omega - \omega_s(\mathbf{k}))^{-1} \}.
\end{aligned} \tag{3}$$

Here  $a_1$  and  $a_2$  (below) are numerical coefficients of order unity,  $\omega_s(\vec{k})$  is the spin-wave frequency,  $A_{1,2,3,q}^{ab} \sim A_{1,2,3,q}(\rho_0 \omega_q / q)$  determines the interaction with the specified acoustic wave described by the deformation tensor and contains the polarization unit vectors. The sample dimensions are usually smaller than the electromagnetic-field wavelength, i.e.,  $\vec{k} = 0$ . We can then put also  $\vec{k}_0 = 0$ , for in this case the electrodynamic system (resonator or waveguide) is excited in the best manner because the spatial structure of the nonlinear magnetization is close to the structure of the field. The condition  $\vec{k} = \vec{k}_0 = 0$  leads to  $\vec{q}_1 = -\vec{q}_2$ , i.e., the geometry of the experiment should be such that the acoustic waves propagate in antiparallel fashion. For spin waves with an activationless spectrum [3]  $\omega_s(\vec{q}) = sq$  we obtain the estimate

$$\begin{aligned}
\chi(\omega, \omega_1, \omega_2) &\sim a_2 (gM_0)^3 (\gamma \delta \sin 2\theta)^2 (\omega + \omega_1 + \omega_2)^{-1} \{ [\omega^{-1} + (\omega_1 - \omega_s(q))^{-1}] \times \\
&\times (\omega + \omega_1 - \omega_s(q))^{-1} + [\omega^{-1} + (\omega_2 - \omega_s(q))^{-1}] \{ \omega + \omega_2 - \omega_s(q) \}^{-1} \}.
\end{aligned} \tag{4}$$

Resonant amplification of the nonlinearity tensor takes place both in the region of magneto-acoustic resonance  $\omega(q) \sim \omega_s(q)$ , and off resonance, at  $\omega + \omega(q) \sim \omega_s(q)$  for the tensor  $\chi(\omega, \omega_1, \omega_2)$  or  $\omega \sim \omega(q) + \omega_s(q)$  for the tensor  $\chi(\omega, -\omega_1, -\omega_2)$ . In this case we get for small  $\theta$

$$\chi_{\text{res}} \sim 0.5 [\gamma \delta (H_0 / H_E)]^2 (M_0' / \Delta H_0) [g(M_0 / \omega)]^2, \tag{5}$$

where  $g$  is the gyromagnetic ratio,  $\Delta H_0$  is the damping, and  $H_E$  is the exchange field. It is easily seen that parametric interaction of two acoustic waves of frequencies  $\omega_1$  and  $\omega_2$  propagating opposite to each other is induced by an alternating magnetic pump field of frequency  $\omega$  under the condition  $\omega_1 + \omega_2 \approx 2\omega$ , and that this effect by the nonresonant component of  $\chi(\omega, -\omega_1, -\omega_2)$ .

The nonlinearity in question can be registered in a displacement experiment in which the crystal is placed in a two-frequency resonator fed at a frequency  $\omega$ , and antiparallel acoustic waves are applied to the sample. For the emitted power  $P$  of nonlinear magnetization of frequency  $\omega_\Sigma$  in a waveguide coupled to the resonator (coupling coefficient  $k$ ) it is easy to obtain the estimate

$$P \sim (k Q_\Sigma \omega_\Sigma \chi^2 H^2 \Pi_{\text{ac}}^2 a V_0) / (\rho_0 v_e^3)^2, \tag{6}$$

where  $Q_\Sigma$  is the quality factor at frequency  $\omega_\Sigma$ ,  $\Pi_{\text{ac}}$  is the sound power flux,  $a$  is the resonator filling factor,  $V_0$  is the volume of the working medium, and  $v_e$  is the speed of sound. For parametric effects to exist it is necessary to satisfy the condition  $q\chi H^2/c > \alpha$ , where  $C$  is the modulus of elasticity and  $\alpha$  is the sound absorption coefficient.

3. We present estimates for the uniaxial antiferromagnet  $\text{MnF}_2$ , in which  $H_f \approx 92$  kOe. According to data given in [5],  $M_0 \approx 500$  Oe,  $H_E \approx 5.5 \times 10^5$  Oe; according to [6],  $C \approx 3 \times 10^{11}$  erg/cm<sup>3</sup>, and from the data of [6] we have  $\gamma \sim 8$ ; from [7] we have  $\Delta H_0 \sim 10$  Oe. Then  $\chi_{\text{res}} \sim 10^9$  for external-field and acoustic-wave frequencies  $\omega \sim \omega_1 \sim \omega_2 \sim 5 \times 10^9$  sec<sup>-1</sup>, and for the power emitted at the frequency  $\omega_\Sigma = \omega + \omega_1 + \omega_2 \sim 1.5 \times 10^{10}$  sec<sup>-1</sup> from a resonator with  $Q_\Sigma \sim 10^3$ ,  $k \sim 0.9$ ,  $aV_0 \sim 10^{-3}$  cm<sup>3</sup> for  $\Pi_{\text{ac}} \sim 10^{-2}$  W/cm<sup>2</sup> and  $H \sim 1$  Oe we obtain  $P \sim 0.5 \times 10^{-3}$  W. For parametric generation of sound waves by an alternating magnetic pump field at a frequency  $\omega \sim 5 \times 10^9$

$\text{sec}^{-1}$  we have  $\chi \sim 2 \times 10^7$  and  $q\chi H^2/c \sim 1 \text{ cm}^{-1}$  at  $H \sim 1 \text{ Oe}$ , i.e., at  $\alpha < 1 \text{ cm}^{-1}$  the parametric condition is satisfied for fields  $H \geq 1 \text{ Oe}$ .

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1) There exist  $H_{S\ell}^i$  that are quadratic and of fourth order in the spin-wave operators, but none of these terms contain exchange parts, and are therefore omitted.

2) The coordinate system  $\zeta\eta\xi$  was chosen here in accord with [3]: the unit vector  $\zeta$  is directed along the magnetic moment of one of the sublattices,  $\xi$  is perpendicular to the plane of the sublattices, and  $\eta$  is perpendicular to the other two unit vectors.

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