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N° d'ordre :

ECOLE CENTRALE DE LILLE

UNIVERSITÉ NATIONALE VERNADSKY TAURIDA

THÈSE

présentée par

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Pour l'obtention du grade de

DOCTEUR

en Electronique

Doctorat delivré conjointement par l'Ecole Centrale de Lille
et l'Université Nationale Vernadsky Taurida

Parametrical Magnetoacoustical Effects in Magnetic Dielectrics and Composite Materials

soutenue le 13 Décembre 2007 devant le Jury d'Examen composé de :

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Thèse préparée dans le Laboratoire Européen en MagnétoACoustique nonlinéaire de la matière condensée (LEMAC) à l'Institut d'Electronique, de Microélectronique et de Nanotechnologie et à l'Université Nationale Vernadsky Taurida

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GENERAL DESCRIPTION OF THE WORK

Relevance. Wave phase conjugation (WPC) of ultrasonic waves for a long time draws to itself attention. Due to this phenomenon a new unique possibilities in physical researches, nondestructive tests, and medicine become available. One could say that mathematically WPC is a realization of time reversal principle, therefore all corresponding features such as phase and distortion compensation becomes available. One of realized applications of conjugated waves is the acoustic microscope. Using of WPC method gives the noise reducing effect in a case of heterogeneous distortions placed near tested object. Besides, WPC-amplification gives additional possibilities to regulate image brightness and contrast. Another application is a nondestructive test of titanium alloy structures. There was matrix of 121 transducers controlled by electronics, forming conjugated wave. With the noise reducing caused by medium granularity there were possible to detect accumulation of α-phase titanium. Hyperthermia is one of the very important medical testing methods. Autofocusing given by WPC allows to concentrate beams on brain-grows thru skull bones. Extension of one more medical method – lithotripsy – with WPC lets not only to destroy nephroliths and other stones in human body without high precision adjustment but with high resulting precision, but also to break the obtained fragments without additional tuning of apparatus.

For WPC systems development it is necessary to find materials with enough high dynamic magnetoelastic coupling. In antiferromagnets such as $\alpha\text{-}Fe_2O_3$ or $FeBO_3$ magnon-phonon coupling is amplified by exchange interaction that causes increase of modulation depth up to about hundred percents. In ultrasonic range WPC was realized in a sample of $\alpha\text{-}Fe_2O_3$, parametrical gain factor came to 36 dB at frequency 35 MHz. But it is impossible to use these materials in ultrasonic technique because of low level of wave's intensity, low aperture, high anisotropy, and difficulties in manufacturing of enough big samples. Exclusively high efficiency of parametrical wave conjugation of progressing wave was achieved with the sample of magnetostrictive ceramics based on nickel ferrite. Gain factor of inverted wave has exceeded 80 dB. Besides, modern technology allows manufacturing such active elements of any necessary shape. However, intensity level limits, threshold levels, conjugated pulse durations force us to search for new structures of parametrical active materials.

Except problems noted above for medical and hydroacoustic application great influence is given by acoustical match between water and active medium. Modern technologies provide manufacturing of wide range of material structures with different properties of materials. For increasing of acoustical match with water it is necessary to decrease both sound velocity and density active element. In a field of hydroacoustical and medical applications used piezotransducers one of the widely known method of acoustical match improvement is introduction of pores in active media. Within the limits of the thesis the synthesis and using of porous ferrites as an active media for WPC systems was suggested. Tested sample has shown transmitting coefficient with water of 50% unlike case of classic ferrite – 16%.

The alternative approach assumes the application of composite materials, which contain active components as particles distributed in vehicle matrix which acoustic parameters are intermediate between liquid and solid. The rare-earth intermetallic compounds of $Tb_{0.3}Dy_{0.7}Fe_2$ (Terfenol-D) are known as materials which demonstrate giant magnetostriction as well as the compensated magnetic anisotropy in the room-temperature range. The magnetoelastic coupling coefficient for these compounds can reach the value up to 80%. Strong magnetoelastic coupling for Terfenol-D is observed as giant magnetostriction and strong dependence of linear and non-linear elastic modules on the applied magnetic field. Such composites demonstrate workable magnetostriction in the frequency range of AC magnetic fields extended up to 1MHz as well as much better acoustical match with water and transmitting coefficient with water of 50%. Moreover, sound velocity in composite is very close to one in water, this noticeably improves the aperture.

For today samples of perspective media such a composites and porous ferrites has weak magnetoelastic coupling and there was no method to study these materials as active media for WPC systems. One of actual task of the work was to develop new method allowing studying parametrical interaction in materials with weak magnetoelastic coupling.

Parametrical interaction of ultrasonic wave with magnetic pumping is the 2^{nd} order nonlinear effect. In spite of the fact that nonlinear magnetoelastic phenomena of higher order are of both fundamental and applied interests, experimental investigation of such effects are presented very narrowly. Within the limits of the thesis the generation of three-phonon bounded states in antiferromagnet was studied theoretically and experimentally.

The main goal of the work was the searching for optimal materials and structures used as an active media for WPC systems, such as ferrites with different structure and porosity, magnetostrictive composites based on Terfenol-D, and searching for new magnetoelastic phenomena of higher orders of nonlinearity. For achievement of the formulated goal following **research tasks** were assigned:

- measurement of propagating longitudinal and transversal sound velocity dependence on external magnetic field with the purpose of determination of magnetoelastic coupling value and sensitivity of sound velocity to a magnetic field for samples of a different composition and structure.

- study of influence of external magnetic field on magnetoacoustical resonant modes frequency and Q-factor for the samples of ferrites and composites with the purpose of determination of magnetoacoustical parameters and analysis of possibility of using these materials as an active media for WPC systems.

- experimental and theoretical investigation of dependence of acoustical oscillations amplitude on parametrical pumping of double frequency with the purpose of determination of influence efficiency of variable magnetic field on sound velocity in composite materials and ferrites of different porosity.

- experimental and theoretical investigation of influence of parametrical pumping of triple frequency on acoustical resonant oscillations in α-Fe_2O_3 with the purpose of observation and description

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of subthreshold and superthreshold modes of three-phonon bounded states.

Object of investigations: magnetoelastic interactions in magnetics.

Subject of investigations: longitudinal and transversal sound velocity and resonant modes frequency and Q-factor dependencies on external magnetic field; parametrical interaction between pumping and magnetoelastic oscillations; dependence of magnetoacoustical parameters with structural parameters of materials.

Methods of investigations: At measurements of sound velocity dependence on magnetic field we used well known echo-pulse method. At measurements of resonant modes frequency and Q-factor dependence on external magnetic field we used both pulse method realized with ultrasonic pulse spectrometer developed and made within the limits of thesis; and the continuous method realized with HP4195A spectrometer. At investigation of parametrical subthreshold and superthreshold modes of interactions in ferrites, composite and porous materials; and for generation of three-phonon bounded states in Hematite we used original 2-pulse method presented in thesis.

Scientific and practical importance of obtained results is:

– obtained maximum value of magnetoelastic coupling constant of 59% in $Fe_{2,026}Ni_{0,95}Co_{0,024}$ ferrite for transversal waves allows, in particular, decrease threshold amplitude levels of dynamic magnetic field in superthreshold ultrasonic conjugation mode, increase parametrical instability increment, and decrease conjugated wave durations without decreasing of their intensity levels.

– measured parametrical amplification increment in Terfenol-D based composite shows that for realizing superthreshold mode in experiment it is enough to increase acoustic Q-factor from level Q = 40 up to Q = 150, that is technologically accessible. In case of porous ferrites is in interest synthesis of sample with porosity decreased from 47% down to 20-30%. Considering close value of an acoustic impedance of a composite and porous ferrite to an impedance of water, superthreshold parametrical magnetoacoustic effects in such materials undoubtedly are of interest for medical and hydroacoustic applications.

– possibility of generation of three-phonon bound states in magnets in application to progressing acoustic waves supposes a wide variety of possibilities of radiation bounded phonons of high intensity from the active magnetic media that can be of interest for applications.

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CONTENTS OF THE THESIS

INTRODUCTION

Introduction gives the bibliographic review on the given subjects, the modern condition of a problem is analyzed, are formulated the purpose and problems of theses.

CHAPTER 1

First chapter presents the physical and mathematical description of effects which are a basis of the dissertation, the review of works on the given theme is presented.

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CHAPTER 2

Second chapter presents the results of experimental investigation of magnetoacoustical properties of nickel ferrites made by hot pressing method, which are doped by Co, Cu, Bi, In and rare-earth ions of Sm, Tb, Yb и Tm. Ferrites of following structures were investigated:

№1 $Fe_{2,026}Ni_{0,95}Co_{0,024}O_4$

№2 $Fe_{1,960}Ni_{0,975}Co_{0,024}Bi_{0,001}In_{0,04}O_4$

№3 $Fe_{1,962}Ni_{0,97}Co_{0,026}Cu_{0,01}Tm_{0,032}O_4$

№4 $Fe_{1,96}Ni_{0,97}Co_{0,03}Cu_{0,01}Yb_{0,03}O_4$

№5 $Fe_{1,862}Ni_{1,092}Co_{0,025}Cu_{0,02}O_4$

№6 $Fe_{1,917}Ni_{1,044}Co_{0,024}Cu_{0,01}$ $Sm_{0,04}O_4$

№7 $Fe_{1,967}Ni_{0,957}Co_{0,006}Cu_{0,05}Tb_{0,02}O_4$

Measuring of sound velocity dependence on magnetic field was realized with echo-pulse method shown in Fig. 1. Necessary magnetic field was applied to a sample by the electromagnet. Exciting of longitudinal and transversal waves with frequency of 5 and 1.4 MHz was realized with piezotransducer of necessary polarization clued to one of the sides of a sample. Ultrasonic pulse produced by piezotransducer propagated along a sample, reflected, and received by the same piezotransducer. Measuring propagation time allows calculating sound velocity. Fig. 1. a, b, and c displays geometries of experiment. The displacement vector and the wave vector are indicated as $\vec{u}$ and $\vec{k}$ correspondently.

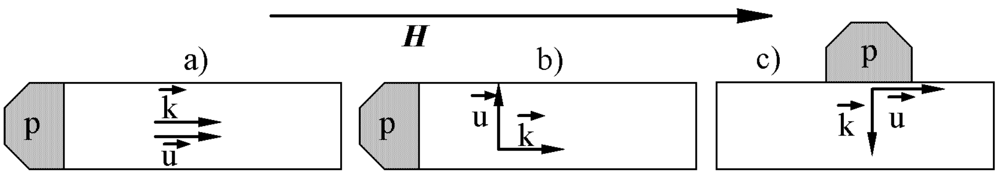

Fig. 1. Scheme of experiment. Possible geometries of experiment: (a) longitudinal wave, (b, c) transversal wave, *p* is piezotransducer, $\vec{k}$ is the wave vector, $\vec{u}$ is the displacement vector.

Fig. 2. displays both dependencies of normalized transversal sound velocity on magnetic field and hysteresis loop for two samples №4 and №1 which has shown the richest magnetoelastic coupling value. The difference between dependencies of transversal waves of different polarization is caused by different longitudinal and transversal magnetic susceptibility and dynamic demagnetizing fields in waves.

The main difference between samples №4 and №1 is the excess of Fe^{2+} ions which has strong spin-orbit coupling in sample №1. Values of magnetic field corresponding to a maximum of magnetoelastic coupling are in the range of 180-220 Oe for all samples. The sample №1 demonstrates maximum of sound speed renormalizing of 12%, unlike to case of sample №1 which has shown renormalizing of sound velocity of 19%. Measured saturation magnetostriction has made ~ $-4.2\cdot10^{-5}$ and ~ $-3.7\cdot10^{-5}$ for samples №1 and №4 correspondently.

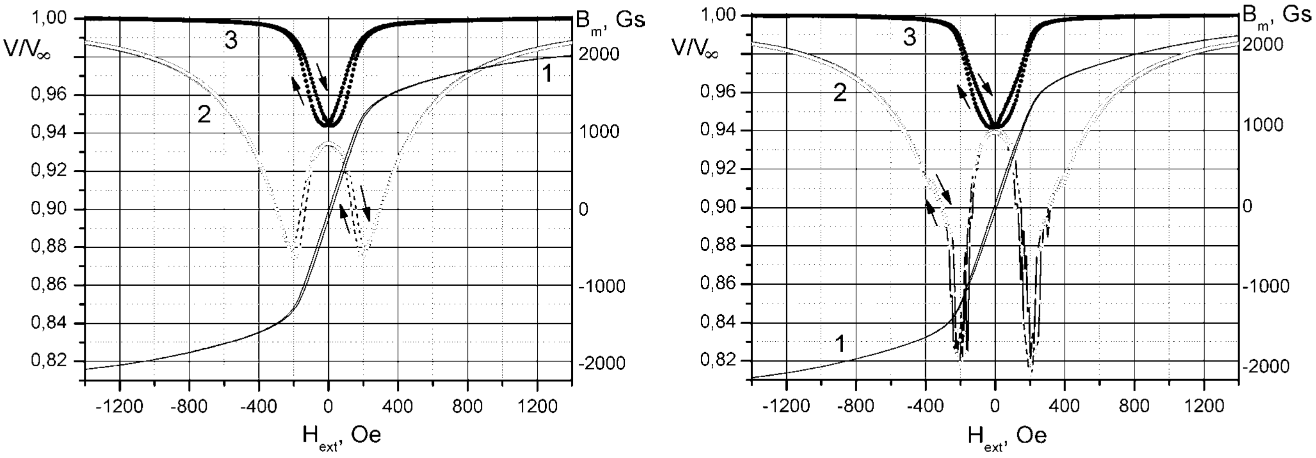

Fig. 2. Dependencies of normalized transversal sound velocity on external magnetic field for (a) sample №4 and (b) sample №1. Curve (2) corresponds to geometry $\vec{k} \parallel \vec{H}$ (see Fig. 1. b), curve (3) corresponds to geometry $\vec{k} \perp \vec{H}$ (see Fig. 1. c). (1) is the hysteresis loop for corresponding samples, $B_m = B(H_{ext}) - H_{ext}$, H_{ext} is the external magnetic field.

Position of a minimum of sound velocity correlates with the field of closure of the hysteresis loop. That confirms that the main influence on magnetoelastic coupling is given by the processes of rotation of magnetization in crystallites. Value of magnetomechanical coupling factor reaches 47% and 59% for samples №4 and №1 correspondently. A maximum of sound velocity slope which defines threshold of parametrical nonstability reaches 100%/kOe for sample №1 that is three times above than for any ferrites tested before including sample №4.

Except numerical difference in magnetoelastic renormalizing of sound velocity, one can clearly see that sample №1 shows curve brakes typical for interference of a modes in case of multimode sound propagation. Fig. 3. shows oscillograms of registered echo-pulse signal in the area of brakes with different values of a field: a) H=660 Oe; b) H=600 Oe; c) H=400 Oe; d) H=270 Oe. For defined threshold level (horizontal dotted line) the program measures delay duration (vertical dotted line). Curve brakes in Fig. 4. are caused by transfer (c)→(d) in Fig. 3. Comparing curves (1) and (2) in Fig. 4. one can note that these peculiarities and independent on sound frequency (5 and 1.4 MHz). Multimode propagation can be caused by strong nonlinearity of internal magnetic field allocation and strong magnetoelastic coupling. This can stipulate distortion of wave front and sound reflections from sides of a sample with further interference.

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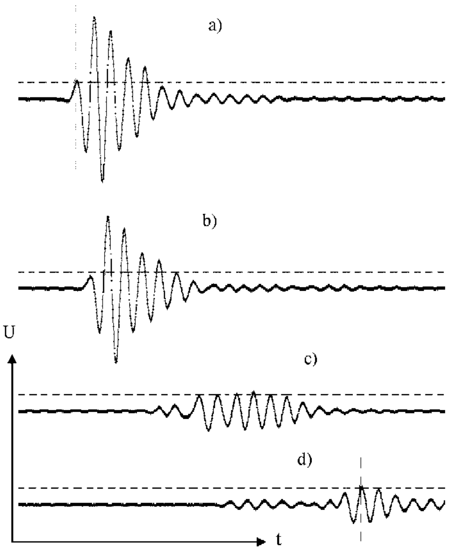

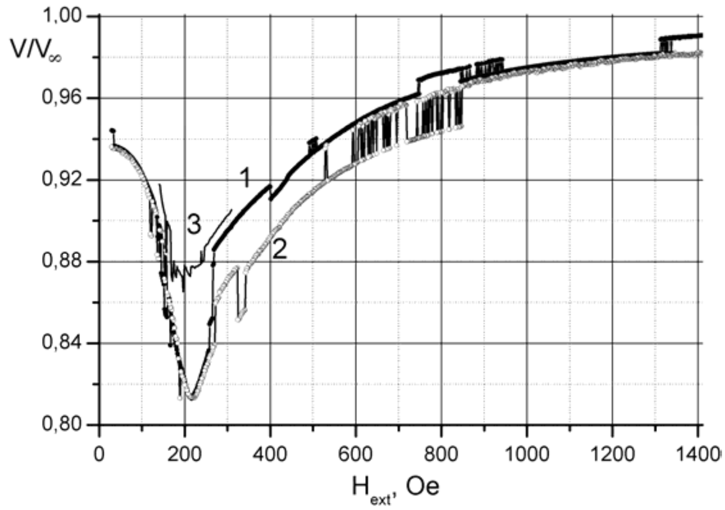

Fig. 3. Oscillograms of registered ultrasonic pulse for sample №1 with different values of external magnetic field: (a) H=660 Oe; (b) H=600 Oe; (c) H=400 Oe; (d) H=270 Oe. Horizontal dotted line is the sensitivity threshold, vertical dotted line is the defined delay duration.

Fig. 4. Dependency of normalized transversal sound velocity on external magnetic field for sample №1, geometry $\vec{k} \parallel \vec{H}$ (see Fig. 1.b). Curve (1) corresponds to 5 MHz sound, curve (2) corresponds to 1.4 MHz sound, curve (3) corresponds to 5 MHz sound with decreased sensitivity level which allows wathcing first of the divided pulses (see Fig. 4, d).

All studied samples demonstrate strong anisotropy of transversal sound on magnetic field direction relative to wave vector (see Fig. 2.). One of the reasons of that is the difference in demagnetization dynamic fields in waves propagating along and across the field. In the first case variable magnetization is mainly parallel to wave front and does not induce generation of a variable magnetic field whereas in the second case magnetization is mainly parallel to a wave vector. In this case opposite variations of the dynamic magnetization corresponds to opposite phases of deformations in the wave, this causes appearance of dynamic lattice of demagnetizing field which weaken magnetoelastic coupling. Estimates show that dynamic demagnetization corresponds to magnetic susceptibility of ~1 could decrease magnetoelastic coupling ten times. This is a good explanation for that big difference in values of sound velocity renormalizing for transversal waves of different polarization.

Fig. 5. displays results of investigation of sound velocity of longitudinal waves. Sound velocity renormalizing values for all tested samples are within the range of 2% that much lower than in case of transversal waves propagating along magnetic field.

The nature of longitudinal sound dependence is close to transversal one in case of wave vector perpendicular to a field. This peculiarity could be explained by influence of dynamic demagnetization too, because dynamic magnetization direction in longitudinal wave is mainly parallel to wave vector. In addition, longitudinal susceptibility in spinel is lower than transversal and decreases faster in process of approach to saturation of magnetization.

We have to note that in case of single domain ferromagnets the linear coupling between magnetic sublattice and longitudinal wave propagating along the magnetic field is absent because longitudinal

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susceptibility is equals to zero. In polycrystals this coupling is conditioned by disorientation of magnetic moment of crystallites relative to a magnetization field.

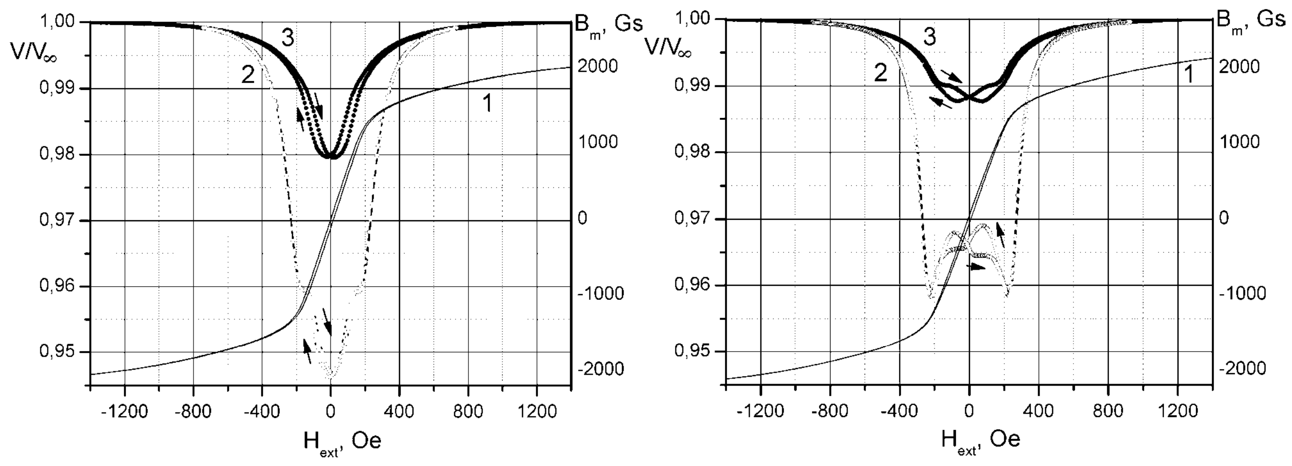

Fig. 5. Dependencies of normalized longitudinal sound velocity correspond to geometry $\vec{k} \parallel \vec{H}$ (3) and resonants mode frequency (2) on external magnetic field for (a) sample №4 and (b) sample №1. (1) is the hysteresis loop for corresponding samples, $B_m = B(H_{ext}) - H_{ext}$, H_{ext} is the external magnetic field.

Fig. 5 represents fundamental longitudinal mode frequency dependence on magnetic field for samples №1 and №4. Excitation and registering of resonant magnetoelastic oscillations was carried out with the solenoid winded along the long side of a sample parallel to the magnetic field. We have used HP4195A analyzer to observe and to analyze the resonance line. Renormalizing of longitudinal mode frequency is higher than longitudinal sound velocity, this fact is in good match with the assumption of strong influence of dynamic demagnetizing on magnetoelastic coupling (in case of long rod demagnetizing is much lower than in case of plane wave). Sample №1 demonstrates anomaly correlated with maximum of transversal waves that could be explained by strong influence of transversal elastic moduli on fundamental longitudinal mode frequency.

Fig. 6. represents signal waveform of conjugated waves recorded while testing samples in WPC system. Sample №4 demonstrates classical signal format unlike in case of the sample №1 which shows two conjugated waves with different delay and different increment of amplification. These signals correspond to different modes (see Fig. 3. c, d).

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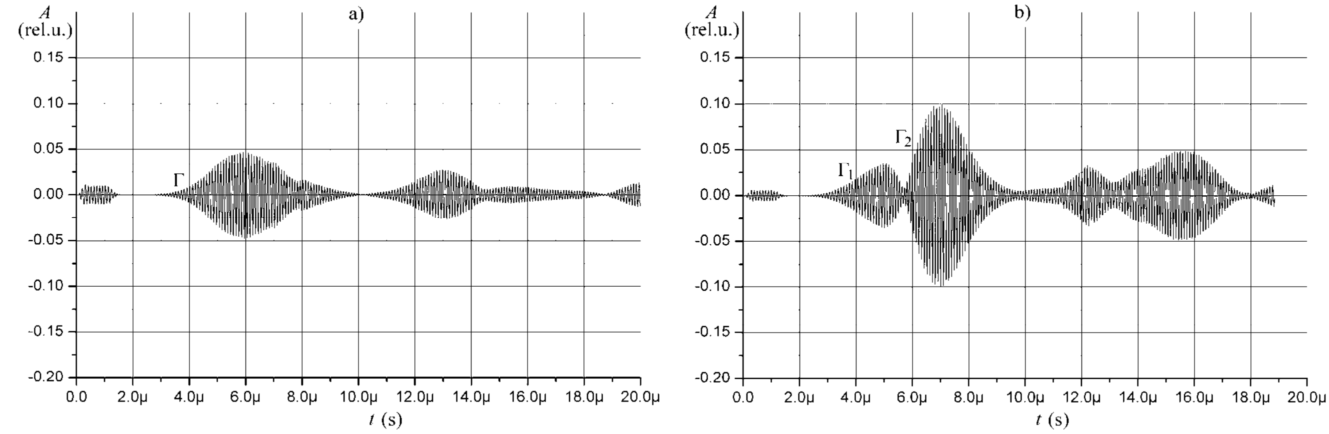

Fig. 6. Signal waveform of conjugated waves obtained with samples (a) №4 and (b) №1. Corresponding values of amplification increment: $\Gamma = 2.03\ \mu s^{-1}$, $\Gamma_1 = 2.4\ \mu s^{-1}$, $\Gamma_2 = 4.47\ \mu s^{-1}$.

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CHAPTER 3

Third chapter presents the results of experimental study of parametric magnetoelastic interaction in active materials with close low acoustic impedances: a porous nickel-cobalt ferrite and a composite based on $Tb_{0.3}Dy_{0.7}Fe_2$ (Terfenol-D) powder with giant magnetostriction. The original pulse technique developed for measurements of modulation depth of sound velocity under AC magnetic field is described. Sensitivity of sound velocity to AC magnetic field and critical amplitude of electromagnetic pumping providing parametric instability of ultrasound are defined for the materials under consideration. The results are giver in a comparison with ones obtained for polycrystalline nickel ferrite.

The sample of the $Fe_{1.943}Ni_{0.945}Co_{0.026}Sm_{0.059}O_4$ ferrite with porosity percentage of 43% prepared as a cylinder of 13.9 mm of base diameter and 34.7 mm in height has been used to perform experiments. The sample of composite material has been prepared as 50.2x10x5.6 mm parallelepiped which contained Terfenol-D particles of size 60 – 90 μm homogeneously distributed in the solidified epoxy. The volume concentration of active media was about 55 %. The measurements give the magnetostriction about $5 \cdot 10^{-4}$ in the external DC fields up to 4 kOe. The reference measurements have been made using the sample of polycrystalline nickel ferrite $Fe_{2.026}Ni_{0.95}Co_{0.024}O_4$ prepared as a 42x13.5x13.5 mm^3 parallelepiped with saturation magnetostriction of $-4.2 \cdot 10^{-5}$.

Exciting free decay oscillations it is possible to register both frequency and damping duration. Within this work the pulse ultrasonic spectrometer with induction excitation has been developed and made. The algorithm is following: radio frequency pulse is applied to a solenoid containing sample. After the pulse acting this or another solenoid gives us a free decay signal. If resonant frequency is close to exciting pulse frequency, amplitude of resonant oscillations increases. Changing frequency consequently it is possible to obtain spectra of resonant modes, see example on fig.7, b. Free decay signal of a ferrite sample is displayed in Fig. 7, a.

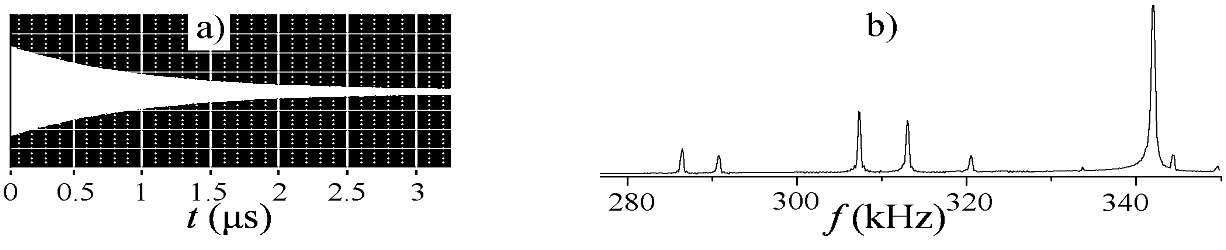

Fig. 7. (a) Free decay oscillation of magnetoelastic mode of a ferrite sample. (b) Spectrum of resonant modes of the same sample.

For measuring magnetoacoustical parameters of the samples the alternative method was used. The solenoid has been used to excite and to detect magnetoelastic oscillations using continuous technique. We have used HP4195A analyzer to observe and to analyze the resonance line at frequency of the fundamental mode of longitudinal oscillations along the longest side of the sample. The dependences of the resonance frequency and Q-factor for this mode on the external DC magnetic field for studied samples are given in fig.8.

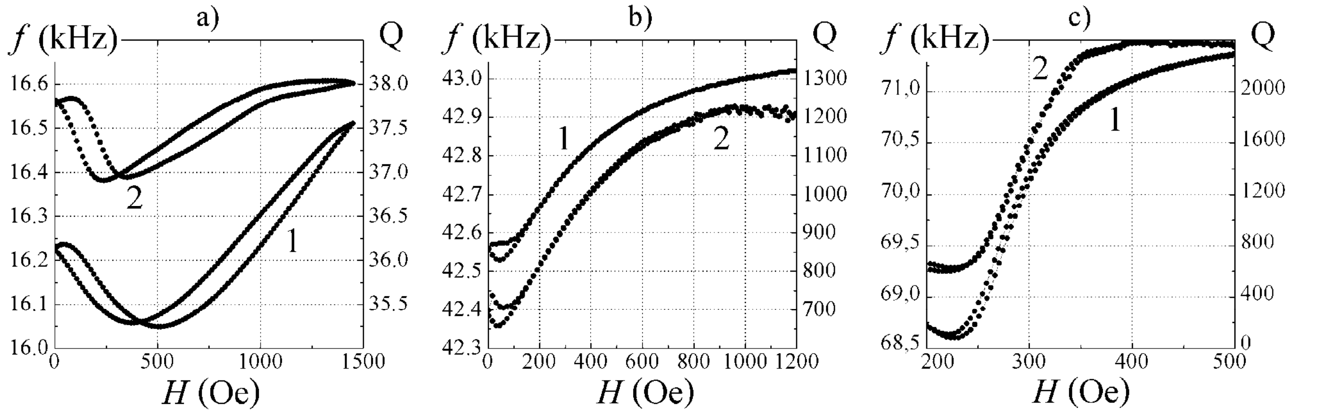

Fig. 8. Dependencies of the fundamental mode frequency (1) and Q-factor (2) on the DC magnetic field for the (a) composite, (b) porous ferrite, and (c) control ferrite samples.

The maximum value of sound velocity calculated from the resonance frequency, experimentally measured density, acoustic impedance, and transmission factor of the inter-face with water are given in the Table 1. For comparison purposes the parameters of the solid non-porous ferrites and water are given also in Table 1. Acoustic impedance $Z = \rho \cdot V_{max}$ has been calculated from experimental values of sound velocity and density.

Table 1

Acoustical parameters of the samples and water.

Material	V_{max} m/s	ρ kg/m^3	Z MRa	T
composite	1659	5336	8,9	0.50
Porous ferrite	2986	3017	9,0	0.49
Non-porous ferrites	~6000	~5600	33	0.16
Water	1500	1000	1.5	1

The scheme of the experiment to investigate the parametric interaction between electromagnetic pumping field and magnetoelastic oscillations in a sample is given in Fig. 9, a. Two radiofrequency pulses have been applied to the coil sequentially (Fig. 9, b). The carrier frequency *f* of the first pulse was equal to the frequency of the resonance mode. The pulse of parametric pumping *2f* has been applied just after the first pulse. The phase shift between two pulses ψ_0 was controlled. In the absence of pumping the first pulse excites a free decay oscillations (Fig. 9, b, curve A_0B_0). Depending on the phase ψ_0 the second pulse enhanced (Fig. 9, b, curve A_0B_1) or damped (Fig. 9, b, curve A_0B_2) oscillations.

The first analyzed parameter for the WPC purposes is the sound velocity sensitivity to the DC magnetic field k_{DC} which can be expressed in terms of resonance frequency ω and magnetic field *H* as $k_{DC} = \omega^{-1}\, \partial\omega/\partial H$. The chosen operating point H_m corresponds to a compromise between highest sensitivity

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k_{DC} and high quality factor Q.

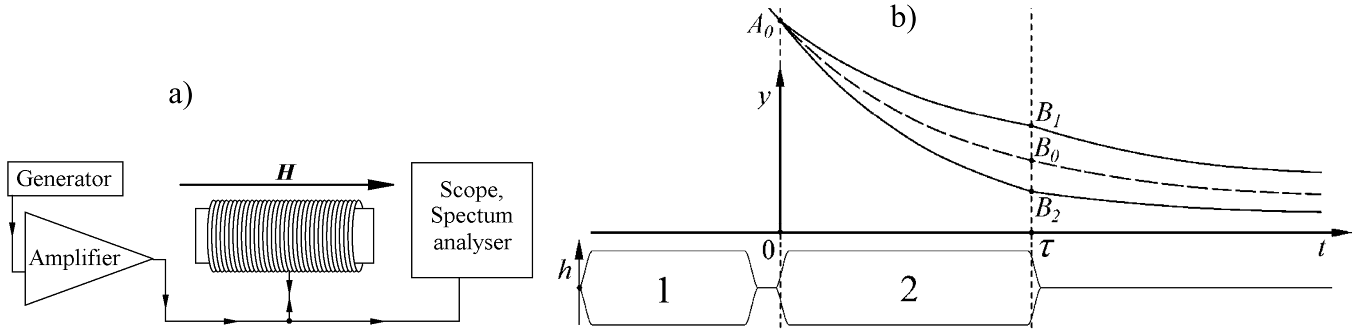

Fig. 9. (a) Scheme of the experiment, (b) time-diagram of the experiment: 1 – exciting pulse at resonance frequency, 2 – pulse of parametric pumping at double frequency, *h* – amplitude of pulses, *y* – amplitude of oscillations, A_0 – oscillation amplitude after first pulse, B_0 – free decay amplitude at moment *τ*, $B_{1,2}$ – decay amplitude at moment τ under parametric pumping, *τ* – duration of the parametric pumping pulse.

Fig. 10 represents the dependence of the oscillation relative amplitude B/B_0 on the pumping pulse phase. Experimental values are normalized on amplitude B_0 registered when the second pulse is absent. The experimental data correspond to the bias magnetic field H_m. Pumping duration for composite, porous ferrite and control ferrite made 1.3, 5.5, and 4.7 ms correspondently.

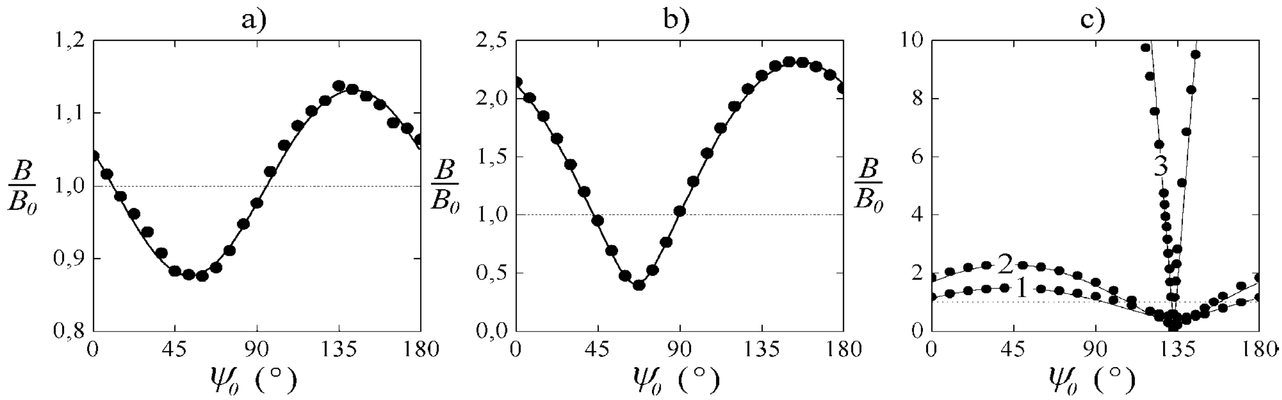

Fig. 10. Experimental (points) and calculated (curves) relative amplitudes of oscillations in the Terfenol-D based composite (a), the porous ferrite (b), and control ferrite (c) vs. phase shift between exciting and pumping pulses. Pumping field amplitude: the composite: h = 253 Oe, the porous ferrite: h = 35 Oe, and the control ferrite: 1.5 Oe (1), 2.9 Э (2), 15 Э (3).

Pumping field amplitude has made 253, 35, and 0–15 Oe for the composite, porous ferrite and for the control ferrite correspondently. The specified parameters allowed registering variations of damping factor in subthreshold modes in all samples and in superthreshold modes in control ferrite.

The quasi-linear approach for the parametric interactions in acoustic resonator yields the system of equations:

$$\begin{aligned} &\frac{\partial B}{\partial t} + \left(\delta - \frac{1}{2} m\omega \sin(2\varphi)\right) B = 0 \\ &\frac{\partial \varphi}{\partial t} - \frac{1}{2} m\omega \cos(2\varphi) = 0 \end{aligned} \quad , \qquad (1)$$

Here φ is the phase shift between displacement and double-frequency pumping field, B is the amplitude of elastic displacement, ω and $\delta = \omega/2Q$ are the frequency and damping factor of the magnetoelastic mode, $m = h\,(\omega^{-1}\partial\omega/\partial h)$ is modulation depth of the resonance frequency. Amplitude of elastic displacement after second pulse derived from Eq. (1) depends on pumping pulse duration τ as:

$$B = A_0 \exp\left\{-\delta\tau + \Gamma\int_0^{\tau}\cos\left[2\,\mathrm{arctg}\left(\mathrm{tg}\,\Psi_0 e^{-2\Gamma t}\right)\right]dt\right\}, \qquad (2)$$

where $B_0(\tau) = A_0 \exp(-\delta\tau)$, $\psi_0 = \varphi_0 - \pi/4$, A_0 and φ_0 – initial amplitude and phase of elastic displacement, $\Gamma = m\omega/2$ is increment of parametric enhancement.

The value of Γ was defined by fitting of function (2) with the experimental data Fig. 10. The solid lines in Fig. 10 represent calculated data. The corresponding values of the modulation depth derived from experimental data are given in Table 2 which represents also the sound velocity sensitivity to AC magnetic field $k_{AC} = \omega^{-1}\ \partial\omega/\partial h$ calculated from obtained values of the increment Γ. The data of experiment give a possibility to estimate the threshold values of increment of parametric instability $\Gamma_C = \delta$, modulation depth m_C and pumping field strength h_C (Table 2).

The difference between static and dynamic results can also be caused by the difference between the static and dynamic demagnetization fields. The latter is due to the difference of the static and high frequency longitudinal magnetic susceptibilities. While for ferrite in the bias field range above 200 Oe the reversible magnetization rotation processes take place that equalizes static and dynamic magnetic susceptibilities, dynamic magnetization of composite occurs by particular hysteretic cycles in all ranges of bias fields used in our experiment. In this case the experimental and calculated variations of amplitude are in good match with each other. In superthreshold mode amplitude is limited by nonlinear internal mechanisms.

Table 2

Parameters of Terfenol-D epoxi-bonded composite, porous $Fe_{1.943}Ni_{0.945}Co_{0.026}Sm_{0.059}O_4$ ferrite and control $Fe_{2.026}Ni_{0.95}Co_{0.024}O_4$ ferrite obtained by pulse and resonance experiments.

Material	H_m kOe	k_{DC} %/kOe	Q	h Oe	τ ms	Γ 10^3 s^{-1}	m 10^{-3}	k_{AC} %/kOe	Γ_C 10^3 s^{-1}	m_C 10^{-3}	h_C Oe
Composite	1.00	3.6	37	97.0	0.6	0.160	4.26	4.39	1.38	27	615
Porous ferrite	0.21	2.3	830	34.8	5.5	0.154	1,15	3.39	0.16	1.2	35.5
Control ferrite	0.3	28	1380	1.5, 2.9, 15	4.7	93, 186, 941	0.42, 0.84, 4.25	28	0.18	0.86	3

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CHAPTER 4

Fourth chapter presents the results of theoretical and experimental studies of the generation of three-phonon coupled excitations in an α–Fe_2O_3 single crystal. As the coupling mechanism, we consider the resonant nonlinear interaction of quasi-phonons with a homogeneous alternating magnetic field $h_{\perp}(t)$ applied in the basal plane of the crystal normally to the magnetization field *H*. At frequencies that are low compared to the frequency of the activation branch of the magnon spectrum, the dynamics of the spin system of the EPAF is reduced to rotations of the antiferromagnetic vector in the basal plane.

Writing the free energy of long-wave spin excitations for this case and solving the equation of motion for the angle φ of the antiferromagnetic vector rotation under the effect of strain and alternating field, it is possible to reduce the free energy density to the energy of the quasi-phonon system, which, accurate to the fourth-order terms, has the form

$$F = \sum_{m=2} \frac{1}{m!}\hat{C}^{(m)}(H)\hat{u}^m + h_{\perp}(t)(\hat{\Psi}\hat{u}^3), \tag{3}$$

where $\hat{C}^{(m)}$ is the *m*th-order elastic modulus tensor renormalized by magnetoelastic coupling and $\hat{\Psi}(H)$ is the amplitude of nonlinear interaction of quasi-phonons with the transverse pumping field:

$$\hat{\Psi}(H) = \left(\frac{2H_E}{M_0}\right)^2\left(\frac{\gamma}{\omega_{S0}}\right)^6 H_D(32\hat{B}_1^2 - 3\hat{B}_2^2)\hat{B}_2, \tag{4}$$

Here, γ is the magnetomechanical ratio, ω_{S0} is the antiferromagnetic resonance frequency, $(\omega_{S0}/\gamma)^2 = H(H+H_D)+2H_EH_{ms}$, and H_{ms} is the effective field of spontaneous magnetostriction. In Eq. (4), for simplicity, we assumed that $H \ll H_D$; for α-Fe_2O_3 and $FeBO_3$ crystals in the fields $H<1$ kOe, this condition is satisfied to a high accuracy.

The resonant interaction conditions for a three-phonon coupled excitation are achieved when the frequency of the pumping field is equal to the sum of the frequencies of three quasi-phonons and when the momentum conservation condition is satisfied. Considering a system of volume quasi-phonon excitations by representing the strain tensor in the form of a superposition of acoustic normal modes one can obtain a equations of motion for the acoustic oscillation amplitude and the phase ψ of the three-phonon correlator $G = |a_n|^3 \exp(i\psi)$:

$$\begin{aligned} &\frac{\partial |a_n|^2}{\partial t} + 2\delta_n |a_n|^2 - \kappa_n h_{\perp} |a_n|^3 \sin\psi = 0 \\ &\frac{\partial \psi}{\partial t} - 3\beta_n |a_n|^2 - \frac{3}{2}\kappa_n h_{\perp} |a_n| \cos\psi = 0 \end{aligned}, \tag{5}$$

where δ_n is the mode attenuation coefficient, $\kappa_n = 3\chi_n/4M_n\omega_n$ is the three-phonon coupling constant, and $\beta_n = 3\Phi_n^{(4)}/2M_n\omega_n$ is the mode's nonlinear frequency shift constant. Unlike the case of phonon pair generation, the characteristic feature of the generation of three-phonon coupled excitations in the case under consideration is the formation (at $\beta_n \rightarrow 0$ and $\psi = \pi/2$) of a singularity of the excitation amplitude $|a_n|$ within a finite time of pumping τ_c. The singularity arises when the following threshold condition is satisfied:

$$\Gamma = Q_n \kappa_n h_\perp |a_n|_0 / \omega_n > 1, \qquad (6)$$

where $|a_n|_0$ is the magnitude of the oscillation amplitude at the instant of the pumping onset and $Q_n=\omega_n/2\delta_n$ is the mode's Q factor. The characteristic time τ_c is determined by the quasi-phonon relaxation time and by the value of the supercriticality parameter Γ:

$$\tau_c = \delta_n^{-1} \ln\left(\frac{\Gamma}{\Gamma - 1}\right), \qquad (7)$$

The amplitude growth is stabilized by the mode's nonlinear frequency shift as the most pronounced intramode nonlinear effect in the quasi-phonon system of EPAFs. Below, results of modeling the generation of three-phonon coupled excitations on the basis of the system of equations (5) are compared with experimental data.

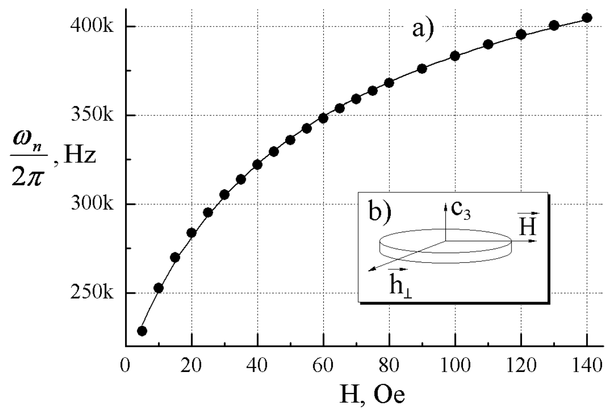

Fig. 11. (a) Frequency of the quasi-phonon mode versus the strength of the bias magnetic field. (b) Geometry of the experiment.

The experiment was performed on an α-Fe_2O_3 crystal shaped as a disk whose plane was parallel to the basal plane. The geometry of the experiment is shown in the inset in Fig. 11 (Fig. 11b). The single-mode excitation of quasi-phonons was performed at the frequency of the fundamental "contour shear" mode of the disk; this mode is characterized by an anomalously strong phonon–magnon coupling and a considerable separation in frequency from other quasi-phonon modes of the acoustic spectrum. The dependence of the resonant frequency of the chosen mode on the bias magnetic field is plotted in Fig. 11a. The curve shown in Fig. 11a can be approximated by the formula

$$\omega_n(H) = \omega_n^0 \sqrt{1 - \frac{H_{ms}^{(1)}}{H + H_{ms}^{(2)}}} \qquad (8)$$

where $H_{ms}^{(1)}$ = 74 Oe and $H_{ms}^{(2)}$ = 90 Oe. Note that the coupling coefficient of the mode under consideration contains the renormalized field value corresponding to the magnetoelastic gap: $H_{ms}^{(2)} < H_{ms}$. As the operating point used in the experiment, we choose the bias field value H = 60 Oe, which corresponds to the frequency $\omega_n/2\pi$ = 350 kHz. The generation of three-phonon excitations was observed as follows. Using an inductance coil, whose orientation was perpendicular to the bias field, two radio pulses were sequentially applied to the crystal. The first, initiating, pulse with a duration of 17 μs excited

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acoustic oscillations at a mode resonance frequency of 350 kHz. The envelope of the pulse had a Gaussian shape, which made it possible to avoid the stray excitation of higher-order modes. When the first pulse was terminated, the pumping pulse was switched on with duration of 200 μs at a tripled mode frequency of 1.050 MHz. At the end of the second pulse, the amplitude A of the signal induced in the coil by magnetoelastic oscillations of the sample at a frequency of 350 kHz was recorded. The measured amplitude was compared with the amplitude A_0 of the freely decaying oscillation signal, which was induced at the same instant of time in the absence of pumping. Depending on the phase shift of the second and first pulses, we observed either attenuation or amplification of oscillations due to the generation of three coupled quasi-phonons. Figure 12 shows the measured dependences of the amplitude ratio A/A_0 on the phase of pumping for different values of the excitation field h_0 with the mode frequency $\omega = \omega_n$ and different values of the pumping field $h_\perp$ of frequency $\omega_p = 3\omega_n$. One canclearly see the increase in the amplification factor with an increase in the pumping field, as well as with an increase in the initial phonon amplitude. When the initial amplitude value is too high (Fig. 12e), the amplification terminates.

The details of the process are revealed by the comparison of the experimental results with the solutions to the system of equations (5), which are displayed in Fig. 13. The parameters of the system were chosen as follows. The normalized initial excitation amplitude was taken to be equal to unity for h_0 = 256 mOe and varied in proportion with the variation of h_0. The value of the nonlinear frequency shift constant was taken to be $\beta_n = 0.45\cdot10^{-3}\omega_n$ according to the data of independent measurements. The supercriticality parameter for the pumping $h_\perp$ = 1.57 Oe at h_0 = 256 mOe was taken to be $\Gamma = 2.6$ and varied in proportion with the variation of $h_\perp$. The estimate of the supercriticality parameter by Eq. (7) for the α-Fe_2O_3 crystal ($2H_E = 18\cdot10^3$ kOe, H_D = 22 kOe, $B \approx 10^7$ dyn/cm^2, and $C \approx 10^{12}$ dyn/cm^2) at H = 60 Oe, $H_{ms}^{(2)}$ = 100 Oe, $h_\perp$ = 1.6 Oe, $|a_n|_0 \approx 10^{-5}$, and $Q_n = 10^3$ yields the value $\Gamma \approx 1$–10. The comparison of the experimental results with the data of numerical modeling shows that, at $|a_n|_0 \approx 10^{-5}$–10^{-6}, the superthreshold regime of three-phonon coupled excitation generation is realized already at the pumping fields $h_\perp$ from fractions to several oersteds. The amplitude stabilization and the generation suppression (Figs. 12 and 13, e) occur as a result of the nonlinear shift of the quasiphonon mode frequency. Under the generation suppression conditions, the phase of the three-particle correlator becomes nonstationary being shifted with time relative to the phase of pumping.

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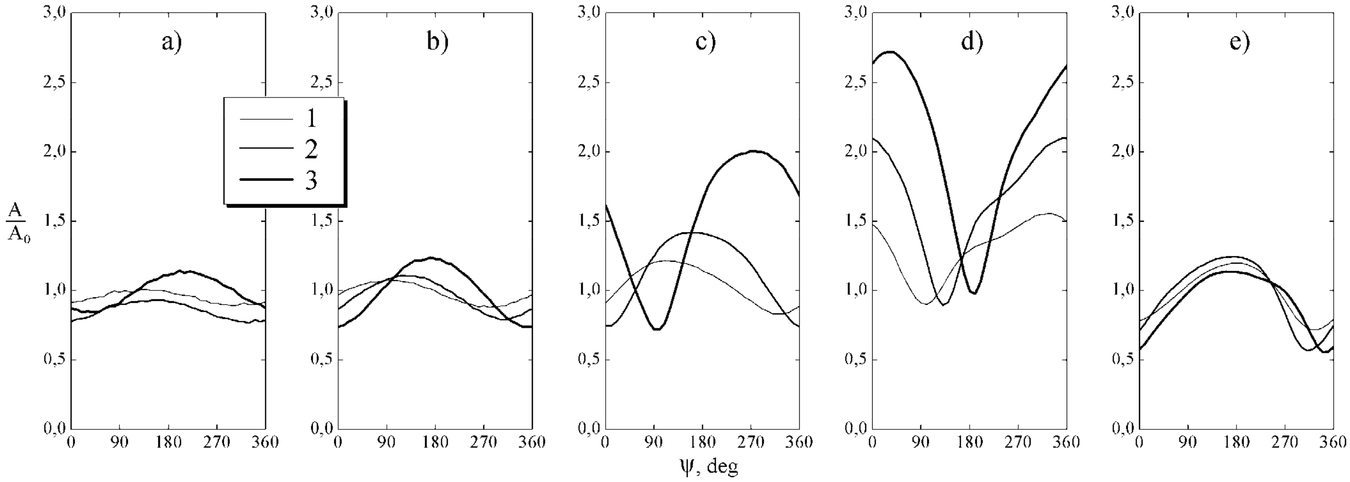

Fig. 12. Experimental dependences of the relative amplitude of magnetoelastic oscillations on the phase of pumping. The strength of the excitation field at the frequency ω_n is: (a) 65, (b) 127, (c) 256, (d) 387, and (e) 635 mOe. The strength of the pumping field at the frequency $3\omega_n$ is: (1) 0.5, (2) 0.86, and (3) 1.57 Oe.

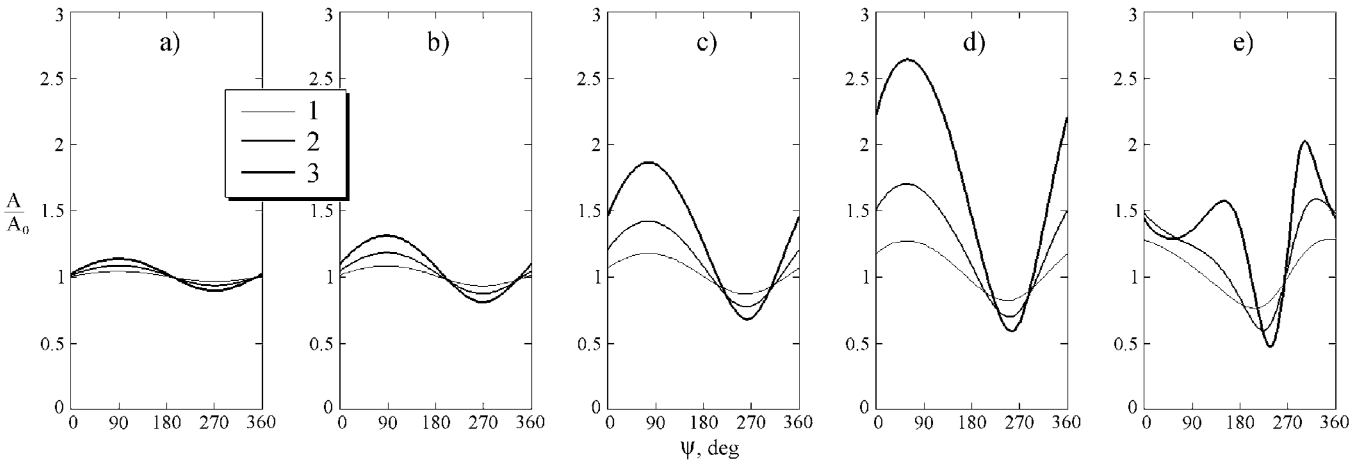

Fig. 13. Calculated dependences of the relative amplitude of magnetoelastic oscillations on the phase of pumping for the conditions identical with those of Fig. 12.

The results presented above demonstrate the possibility of the generation of three-phonon coupled excitations in magnets in a relatively weak alternating magnetic field. When the initial amplitude of magnetoelastic waves is sufficiently high, three-phonon excitations can be generated in a specific superthreshold regime, which is accompanied by the formation of a singularity of the acoustic field within a finite time of pumping. The development of singularity is stabilized by internal nonlinear processes in the quasi-phonon system. As applied to traveling acoustic waves, the phenomenon under consideration allows a wide variety of possibilities for the emission of high-intensity coupled phonons from an active magnetic medium, which may be of interest for practical applications.

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MAIN RESULTS AND SUMMARIES

1. For the first time it is experimentally obtained huge magnetoelastic coupling constant of 59% in $Fe_{2,026}Ni_{0,95}Co_{0,024}$ ferrite for transversal waves. Corresponding sensitivity of sound velocity to a magnetic field makes 100%/kOe, that three times higher than for any ferrites tested before. In particular that allows decreasing threshold amplitude levels of dynamic magnetic field in superthreshold ultrasonic conjugation mode, increase parametrical instability increment, and decrease conjugated wave durations without decreasing of their intensity levels. Within the limits of thesis this sample was tested in WPC system. For the first time the value of amplification increment has made ~ 4.5 μs^{-1} unlike case of ferrites studied before with amplification increment of ~2 μs^{-1}.
2. Within the limits of the thesis the new original method was developed and realized which allows investigating parametrical interactions in magnetoelastic media including materials with weak magnetoelastic coupling. This method was used not only for investigation of parametrical interactions in composites and porous ferrites, but also for experimental studying of three-phonon bound states in antiferromagnet α-Fe_2O_3.
3. For the first time sound velocity dependencies on an alternating magnetic field in Terfenol-D ($Tb_{0.3}Dy_{0.7}Fe_2$) based composite and porous nickel ferrites are recorded. Estimates show that for realizing superthreshold mode in experiment it is enough to increase acoustic Q-factor from level Q = 40 up to Q = 150, this is technologically accessible. In case of porous ferrites is in interest synthesis of samples with porosity decreased from 47% down to 20-30%. Considering close value of an acoustic impedance of a composite and porous ferrite (~ $9\cdot10^3$ $n\cdot s/m^3$) to an impedance of water, superthreshold parametrical magnetoacoustic effects in such materials undoubtedly are of interest for medical and hydroacoustic applications.
4. For the first time the generation of three-phonon bound states in antiferromagnet under influence of relatively weak alternating magnetic field was demonstrated experimentally and theoretically. Evolution of this process was described mathematically and modeled numerically. In case of enough high levels of initial exciting of magnetoelastic waves the three-phonon bound state could be generated in specific superthreshold mode accompanied by formation of acoustical field singularity within a finite duration. These features in application to progressing acoustic waves supposes a wide variety of possibilities of radiation bounded phonons of high intensity from the active magnetic media that can be of interest for applications.

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SUMMARY

Rudenko V. V. Parametrical magnetoacoustical effects in magnetic dielectrics and composite materials

Influence of a structure of the polycrystalline nickel based ferrites on magnetoelastic coupling in ultrasonic range is investigated experimentally. Sound velocity dependencies on magnetizing field are given. Maximum magnetoelastic coupling coefficient of 59% and sensitivity of shear sound velocity a magnetic field of 100% was obtained for $Fe_{2,026}Ni_{0,95}Co_{0,024}O_4$ ferrite sample. The sample was tested in wave phase conjugation system, for the first time gain increment make 4.5 μs^{-1} that twice higher than values obtained earlier.

Due to original high sensitivity pulse method the parametric interaction between electromagnetic pumping field and magnetoelastic oscillations is studied experimentally in perspective materials with acoustical impedance close with water: the $Tb_{0.3}Dy_{0.7}Fe_2$ (Terfenol-D) based composite and porous ferrite of $Fe_{1.943}Ni_{0.945}Co_{0.026}Sm_{0.059}O_4$ structure. Sound velocity sensitivity on pumping AC magnetic field was found as 4.39%/кOe and 3.39%/кOe for the composite and porous ferrite respectively. Application of studied materials in ultrasonics for acoustic matching of parametric wave phase conjugators with water is discussed.

The results of experimental observation of generation process of three-phonon bound states in high-temperature antiferromagnet α-Fe_2O_3 under transversal pumping field influence executed for the first time are presented. Sub-threshold and over-threshold modes of three-bound states with controlled changing of the phase of pumping relative to the phase of three-phonon correlator of an acoustical field are investigated with original pulse method. Specificity of above-threshold mode is the forming of singularity in a finite pumping duration. Singularity progress is stabilized by internal nonlinear processes in a quasiphonon system. Experimental results are in a good match with the theory of resonant nonlinear interaction of quasiphonon with an electromagnet field.

Keywords: nickel ferrite, composite, hematite, magnetoelastic wave, wave phase conjugation, parametrical gain increment.

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