Electromagnetic excitation of ultrasound in gadolinium

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An experimental and theoretical investigation of the processes of electromagnetic excitation of ultrasonics in gadolinium single crystals has been carried out. The main mechanisms of the electromagnetic-acoustic transformation have been established and the regions in which they appear most effective have been determined. Over a wide range of variation of the magnetic field and temperature the generation of longitudinal ultrasonics is achieved first through the paraprocess (near T_c), secondly through spin flip (at $T < T_c$) and finally, in high magnetic fields, through inductive interaction. From analysis of the temperature and field dependences of the efficiency of longitudinal ultrasound generation it was possible to establish the boundaries of the magnetic phases in gadolinium.

1. INTRODUCTION

Rare-earth metals and their compounds with 3d magnetic elements find ever widening application in devices for direct conversion of electromagnetic and acoustic waves.[1] There is, in particular, an interest in finding the values of frequency, magnetic field and temperature at which the processes of electromagnetic excitation of ultrasound occur most efficiently in these materials. On the other hand, a study of the effects of direct conversion can be used to obtain new information on the dynamical properties of magnetic materials and about the interaction in them of electromagnetic, spin, and elastic waves. The most intense interactions of these types of oscillations occur in the vicinity of magnetic phase transitions, when the magnetic ordering in the material is either changed or destroyed. Thus, when studying electromagnetic-acoustic conversion in nickel[2] and iron[3] a sharp rise was found in the generation amplitude of longitudinal ultrasonics at the Curie temperature. Such a clearly marked growth in the amplitude of longitudinal ultrasound was observed in dysprosium and terbium at the transitions from the paramagnetic to the antiferromagnetic and from the antiferromagnetic to the ferromagnetic state.[4,5] These results were mainly obtained on polycrystalline specimens of the magnetic materials, which appreciably complicates even a qualitative theoretical interpretation of the phenomenon.

In the present work the temperature and field dependences of the efficiency of electromagnetic excitation of longitudinal ultrasonics has been studied in gadolinium single crystals and a theoretical analysis has been carried out of the features of the conversion, observed in this metal in the region of the transition, from the paramagnetic to the ferromagnetic state and for spin-flip transitions in the ferromagnetic phase, both spontaneous and induced by a magnetic field.

Gadolinium has a hexagonal lattice and goes from the paramagnetic state to a ferromagnet of the "easy axis" type at $T_c = 293$ K. The magnetization vector **M** coincides in direction in the range $T_{sf} < T < T_c$ with the hexagonal crystal axis. At $T_{sf} = 235$ K a cone of easy magnetization axes is formed. On lowering the temperature, the aperture angle of the cone relative to the hexagonal axis first increases rapidly, reaches a maximum at $T \approx 180$ K and then decreases monotonically.[6] The magnetic anisotropy in gadolinium is small compared with other heavy rare-earth metals, so that spin flip transitions can be achieved in it in relatively weak magnetic fields.

2. THE EXPERIMENTS

The measurements were carried out on two gadolinium single crystals. Specimen No. 1 was in the form of a 0.95 cm diameter cylinder of height 0.82 cm. The normal to its plane surfaces coincided with the two-fold symmetry crystallographic axis b, the six-fold symmetry axis c was perpendicular to the axis of the cylinder. Specimen No. 2 was a rectangular parallelepiped with sides 0.41 cm, 0.56 cm, and 0.76 cm. The six-fold c axis coincided with the normal to the largest (the "working") face of the specimen. The quality of the specimens studied was characterized by the resistance ratio $RRR = 20$.

The specimens were placed in solenoidal induction coils to which radiofrequency pulses of voltage about 2 kV were applied, of ~ 1 μs duration, and with carrier frequency 5 MHz. The skin-layer thickness δ in gadolinium, over all the temperature range studied at this frequency, is less than the wavelength λ of the excited ultrasound. The strength of the alternating field h at the metal surface was ~ 100 Oe. Measurements in the range 80–300 K were carried out in a steady magnetic field $H < 10$ kOe and in the range 4–80 K in a field $H < 50$ kOe.

Ultrasound excitation by an electromagnetic wave falling on the boundary of the metal was studied in a tangential geometry $\mathbf{H} \| \mathbf{h} \perp \mathbf{n}$, where **n** is the normal to the plane of the specimen. Ultrasound pulses at the frequency of the incident electromagnetic wave were generated in both opposite faces of the specimen and propagated along the normal to them and, on reaching the opposite surface of the specimen, were recorded by the same coil because of the inverse electromagnetic-acoustic conversion. According to Gitis,[2] the efficiencies of the forward and reverse electromagnetic-acoustic conversion are equal. We confirmed this fact in control measurements using a quartz transducer as either the ultrasound source or receiver. The sequence of the echo signals, with intervals $t = d/s$ between them (where d is the thickness of the plate and s is the velocity of sound), were first fed to a wide-band amplifier and then to a pulse-height analyzer and a recorder. The amplitudes of separate (usually the third or fourth) echo signals could be recorded with the pulse-height

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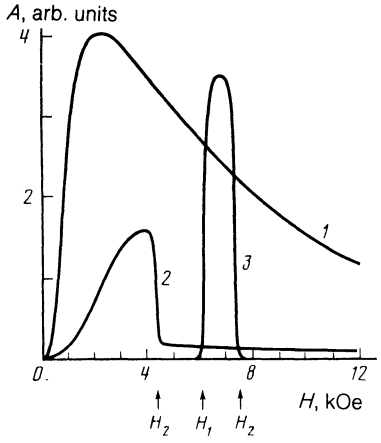

FIG. 1. Experimentally recorded field dependences of the electromagnetic-acoustic conversion signal $A(H)$ in Gd for $\mathbf{H}\|\mathbf{h}\|\mathbf{a}$, $\mathbf{k}\|\mathbf{b}$. Curve 1) $T = 293$ K, 2) 260 K, 3) 127 K.

analyzer. By varying the magnetic field or the temperature, the field and temperature dependences of the efficiency of electromagnetic-acoustic conversion could be recorded. The measured signal in such an arrangement of the experiment is $A \propto K \exp(-N\Gamma d)$, where K is the efficiency of electromagnetic-acoustic conversion, defined as the ratio of energy fluxes falling on the face of the metal in the electromagnetic wave and in the elastic wave generated in it, Γ is the ultrasound attenuation, and N is the echo-signal number.

The measurements on the first specimen were carried out for two orientations of the field $\mathbf{H}$ relative to the crystallographic axes of gadolinium: $\mathbf{H}\|\mathbf{h}\|\mathbf{c}$ and $\mathbf{H}\|\mathbf{h}\|\mathbf{a}$, and on the second for $\mathbf{H}\|\mathbf{h}\|\mathbf{b}$. Characteristic field dependences of the recorded signal of the double electromagnetic-acoustic conversion $A(H)$ in the $\mathbf{H}\|\mathbf{h}\|\mathbf{a}$ orientation are shown in Fig. 1. Near the transition from the paramagnetic state to the ferromagnetic, the generation of ultrasound starts from zero magnetic field, reaches a maximum for $H \approx 2$ kOe and then decreases monotonically (curve 1). In the temperature interval $T_{sf} < T < T_c$ generation also starts in small magnetic fields, reaches a maximum and then falls rapidly and reaches a level weekly dependent on magnetic field (for $H < 10$ kOe)

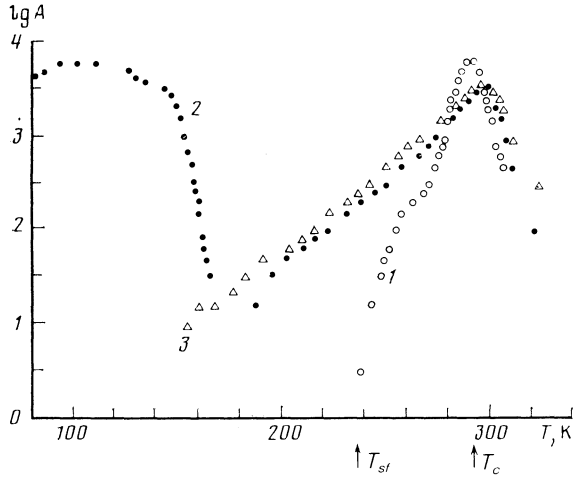

FIG. 3. Field dependences of the $A(H)$ signal in Gd for $\mathbf{H}\|\mathbf{h}\|\mathbf{c}$, $\mathbf{k}\|\mathbf{b}$. Curve 1) $T = 293$ K, 2) 260 K, 3) 80 K.

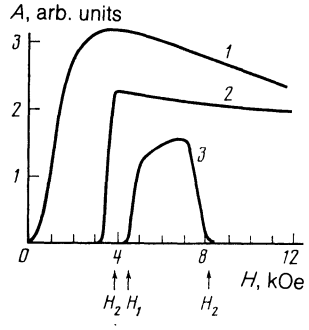

FIG. 2. Temperature dependences of the $A(T)$ signal in Gd for fixed values of the magnetic field $\mathbf{H}\|\mathbf{h}\|\mathbf{a}$, $\mathbf{k}\|\mathbf{b}$: ○) $H = 2.2$ kOe, ●) 4.4. kOe, △) 6.7 kOe.

(curve 2). Finally, for $T < T_{sf}$ the excitation of ultrasonics is only observed in a narrow range of magnetic fields H_1–H_2 (curve 3). As the temperature changes, both the width of this range and the values of the limiting fields H_1 and H_2 changed appreciably.

The $A(T)$ dependences for various mixed values of the field $\mathbf{H}\|\mathbf{h}\|\mathbf{a}$ are shown in Fig. 2. The efficiency of excitation of longitudinal ultrasound reached in all cases a clearly marked maximum in the region of the transition from the paramagnetic to the ferromagnetic state and fell with the distance from T_c. In addition, comparable generation efficiency was observed in some magnetic field range at low temperatures (curve 2 of Fig. 2). Similar temperature and field dependences of A were observed for $\mathbf{H}\|\mathbf{h}\|\mathbf{b}$.

The $A(H)$ dependences in the $\mathbf{H}\|\mathbf{h}\|\mathbf{c}$ orientation are shown in Fig. 3. Unlike the cases considered above, ultrasound generation in the range $T_{sf} < T < T_c$ started in a field $H_1 \neq 0$.

In the general case, electromagnetic ultrasound generation in magnetically ordered metals takes place not only on account of magnetoelastic interactions, but also under the action of inductive interaction which is universal for all conducting solids. For this mechanism of transformation, a quadratic dependence on magnetic field $A(H)$ is a characteristic feature.[7] The dependence of the amplitude of generation of longitudinal ultrasound in gadolinium over a wide magnetic field range is shown in Fig. 4 for $T = 8$ K. It can be seen that the efficiency of the inductive mechanism for the conversion only becomes comparable with the efficiency of the magnetoelastic mechanisms in sufficiently strong magnetic fields.

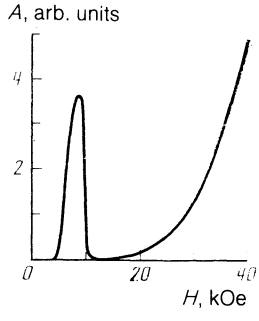

FIG. 4. Variation of the $A(H)$ signal in Gd for $\mathbf{H}\|\mathbf{h}\|\mathbf{b}$, $\mathbf{k}\|\mathbf{c}$, $T = 8$ K.

Attenuation of longitudinal ultrasound in gadolinium was measured by Anikeev *et al.*[8] From the analysis[8] of the temperature variation of Γ and the magnetic structure it was possible to confirm that all the important features observed in the $A(H,T)$ dependences are associated only with the changes in the efficiency of electromagnetic-acoustic conversion.

3. THEORY

1. The system of equations.

A study of generation of ultrasound by an electromagnetic wave falling on the surface of a magnetic material presupposes the solution of a coupled system of equations describing the propagation and interaction of electromagnetic, spin and elastic waves in the metal. Such a system includes the elastic equations, the Landau-Lifshitz equation for the magnetization, and the Maxwell equations

$$\rho \dot{U}_i = \frac{\partial \sigma_{ik}}{\partial x_k}, \qquad \dot{M} = g[M, H_{\text{eff}}] + R,$$
$$\operatorname{rot} E = -\frac{\partial}{\partial t}(H+4\pi M), \qquad \operatorname{rot} H = \frac{4\pi}{c}\sigma E. \tag{1}$$

Here ρ is the density of the metal, U_i is the displacement, $\sigma_{ik} = \delta F/\delta U_{ik}$ is the stress tensor, F the free energy density, U_{ik} is the strain tensor, M the magnetization, g the gyromagnetic ratio, $H_{\text{eff}} = -\delta F/\delta M$ is the effective magnetic field, E is the electric field, σ is the conductivity, and c the velocity of light. The relaxation term R can be put in the form

$$R = \frac{1}{\tau_2} H_{\text{eff}} + \frac{1}{\tau_1 M^2}[M, [M, H_{\text{eff}}]], \tag{2}$$

where τ_i are the relaxation times. In the system of equations (1) we have neglected attenuation in the elastic subsystem and the displacement current in the Maxwell equations.

The original system of equations must be supplemented by the standard boundary conditions for the tangential and normal components of the electric and magnetic fields at the surface, and by the condition that there be no elastic stresses at the free boundary of the metal, $\sigma_{ik|s} = 0$. The expression for the mean free energy density of the magnetic material has the form

$$F = {}^1/_2 aM^2 + {}^1/_2 bM^4 + {}^1/_2\beta_1 M_z^2 + {}^1/_2\beta_2 M_z^4 + {}^1/_2\beta_3 M_z^6 + {}^1/_2\gamma_1 M^2 U_{zz} + {}^1/_2\gamma_2 M^2 (U_{xx}+U_{yy}) + \gamma_{ijkl} M_i M_j U_{kl} + C_{ijkl} U_{ij} U_{kl} - M(H+h), \tag{3}$$

where a and b are the constants of homogeneous exchange, β_i are the magnetic anisotropy constants, γ_1 and γ_2 are the (bulk) magnetostriction exchange constants, γ_{ijkl} is the anisotropic relativistic magnetostriction tensor, C_{ijkl} is the elastic-moduli tensor. For simplicity we neglect inhomogeneous exchange in Eq. (3), as a result of which dispersion will be absent in the spin-wave spectrum. There is then no need to consider boundary conditions for the magnetization.

2. Excitation of longitudinal ultrasound in the temperature region $T \geqslant T_c$ in tangential geometry

We consider the case, corresponding to the experimental arrangement, of the following mutual orientation of **H** and **h** and of the direction of the wave vector in the excited waves: $\mathbf{H}\|\mathbf{h}\|\mathbf{a}\|\mathbf{x}$, $\mathbf{k}\|\mathbf{n}\|\mathbf{b}\|\mathbf{y}$.

In the paramagnetic region the anisotropic magnetostriction γ_{ijkl} and the constants of the magnetic anisotropy β_i can be neglected in the expression (3) for the free energy density. In the ground state the components M_y and M_z and the non-diagonal components of the strain tensor U_{ij} are equal to zero, while the diagonal components of U_{ij} and M_x are determined by the formulae

$$U_{zz} = \frac{1}{\Delta}\left[\gamma_2 c_{13} - \frac{1}{2}\gamma_1(c_{11}+c_{12})\right] M^2,$$
$$U_{xx} = U_{yy} = \frac{1}{\Delta}\left(\gamma_1 c_{13} - \frac{1}{2}\gamma_2 c_{33}\right) M^2, \tag{4}$$

where

$$\Delta = c_{33}(c_{11}+c_{12}) - 2c_{13}^2, \quad \tilde{b} = b + [\gamma_1\gamma_2 c_{13} - \gamma_1^2(c_{11}+c_{22})/4 - \gamma_2^2 c_{33}/2]/\Delta,$$
$$aM_x + 2\tilde{b}M_x^3 = H. \tag{5}$$

This yields the well known expression for the susceptibility of the magnetic material

$$\chi = (a + 6\tilde{b}M_x^2)^{-1}. \tag{6}$$

Near T_c the quantity a can be expressed in the form $a = a_0(1 - T_c/T)$.

All the quantities in the present problem can be represented in the form

$$Q = Q_0 + q\exp(i\omega t - iky), \tag{7}$$

where Q_0 stands for M, U_{ik}, H, E; q stands for m, u_{ik}, h, e; $q \ll Q_0$. Using Eq. (7) the system of equations (1) is linearized relative to the equilibrium values Q_0, determined by expressions (4) and (5), in terms of the small quantities q. It then turns out that for longitudinal sound u_y is only associated with the components m_x, h_x and e_z. Transverse sound is not excited in the range $T \geqslant T_c$.

The generation of longitudinal ultrasound is described by the system of equations:

$$(\omega^2 - \omega_l^2) u_y = ik\gamma_2 M_x m_x/\rho,$$
$$(i\omega + 1/\tau_2\chi_\parallel) m_x = h_x/\tau_2 + ik\gamma_2 M_x u_y/\tau_2,$$
$$ikh_x = 4\pi\sigma e_z/c, \quad cke_z = \omega(h_x + 4\pi m_x), \tag{8}$$

where

$$\omega_l^2 = c_{11}k^2/\rho = s_l^2 k^2, \quad \chi_\parallel = (a + 6b^* M_x^2)^{-1},$$
$$b^* = b + [\gamma_1 U_{zz} + \gamma_2(U_{xx}+U_{yy})]/6M_x^2.$$

The dispersion equation for coupled electromagnetic, spin and elastic waves can be obtained from the system of equations (8):

$$\left(k^2 + \frac{2i\tilde{\mu}_\parallel}{\delta^2}\right)\left[k^2 - \frac{\omega^2}{s_l^2(1-\tilde{\zeta})}\right] + \frac{8\pi i k^2 \tilde{\zeta}\chi_\parallel}{\delta^2(1-\tilde{\zeta})} = 0, \tag{9}$$

where

$$\tilde{\mu}_\parallel = 1 + 4\pi\tilde{\chi}_\parallel, \quad \tilde{\chi}_\parallel = \chi_\parallel/(1 + i\omega\tau_2\chi_\parallel),$$
$$\tilde{\zeta} = \zeta_0\chi_\parallel/(1 + i\omega\tau_2\chi_\parallel), \tag{10}$$

$\zeta_0 = \gamma_2^2 M_x^2/c_{11}$ is the magnetoelastic coupling parameter while $\delta = c(2\pi\sigma\omega)^{-1/2}$ is the skin depth in the non-magnetic metal.

The following values of the wave number are the solution of the dispersion relation (9) under conditions of a skin depth small compared with the elastic-wave length

$$k_1 = \frac{1-i}{\delta}\left(\frac{\tilde{\mu}_\parallel - \tilde{\zeta}}{1-\zeta}\right)^{1/2}, \tag{11}$$

$$k_2 = k_2' - i\Gamma, \tag{12}$$

where

$$k_2' = \frac{\omega}{s_t}\left(\frac{\tilde{\mu}_\parallel}{\tilde{\mu}_\parallel - \tilde{\zeta}}\right)^{1/2}, \quad \Gamma = \frac{\delta^2\omega^2 k_2'}{4s_t^2}\frac{(\tilde{\mu}_\parallel - 1)\zeta}{(\tilde{\mu}_\parallel - \tilde{\zeta})^2}.$$

The solution (11) coresponds to a quasielectromagnetic wave and (12) to a quasielastic wave.

The boundary conditions at the free surface of the metal ($y = 0$) for the linearized system of equations (8) is written in the form

$$h_{1x} + h_{2x} = h_{0x} + h_{3x}; \quad e_{1z} + e_{2z} = e_{0z} + e_{3z} - ik_1 c_{11} u_{1y} + \gamma_2 M_x m_{1x} - ik_2 c_{11} u_{2y} + \gamma_2 M_x m_{2x} = 0, \tag{13}$$

where h_{0x}, e_{0z} and h_{3x}, e_{3z} are the components of the electromagnetic field of the waves falling on and reflected from the surface of the magnetic material. The indices 1 and 2 refer to waves propagating within the bulk of the metal and correspond to the solutions of the dispersion relation (11) and (12).

Solution of the system of equations (8) with the boundary conditions (13) lead to the following expression for the amplitude of the excited ultrasound:

$$u_{2y} = \frac{ik_2'\gamma_2 M_x \tilde{\chi}_\parallel h_{0x}}{\rho s_t^2 (1-\tilde{\zeta})(k_1^2 - k_2'^2)}. \tag{14}$$

The wave u_{1y} is attenuated within the depth of the skin layer and will not be considered in what follows.

In the geometry with $\mathbf{H}\|\mathbf{h}\|\mathbf{b}$ and $\mathbf{k}\|\mathbf{c}$, Eq. (14) serves as the solution of the initial system of equations (1), in which it is only necessary to carry out a replacement of variables: γ_2 by γ_1, u_{2y} by u_{2z}, and y by z.

3. Excitation of ultrasonics in the ferromagnetic region in a single-domain magnetic material

The constants γ_{ijkl} and β_i in gadolinium are non-zero below the Curie temperature and depend on temperature in a complicated way. This manifests itself in the complicated temperature dependence, described above, of the direction of the easy magnetization axis relative to the crystallographic axes. In a magnetic field perpendicular to the hexagonal c axis the magnetization vector lies in the plane passing throught the c axis and the vector $\mathbf{H}$. As the magnetic field is increased, a spin-flip transition takes place which, depending on the temperature, can be a transition of either first or second order.[6]

We shall consider the experimental geometry $\mathbf{H}\|\mathbf{h}\|\mathbf{b}$ and $\mathbf{k}\|\mathbf{n}\|\mathbf{c}$ and neglect the third magnetic-anisotropy constant. In this case the equlibrium components M_x and M_z of the magnetization in the canted phase of gadolinium are determined by the equations

$$2\tilde{\beta}_2 M_x^3 - (2\tilde{\beta}_2 M^2 + \tilde{\beta}_1) M_x = H, \quad M_z^2 = M^2 - M_x^2, \tag{15}$$

where β_i are the renormalized magnetostriction constants of the magnetic anisotropy.

The linearized system of equations (1) takes near the equilibrium position Eq. (15) the following form:

$$\begin{aligned}
i\omega m_{x,z} &= -gM_{z,x}h_y + \frac{M_{z,x}}{M}\omega_1 m_y + g\gamma_{44}M_z M_{z,x}\frac{\partial u_y}{\partial z} + r_{x,z},\\
i\omega m_y &= gM_z h_x - gM_x h_z + \frac{M_x}{M}\omega_2 m_z - \frac{M_z}{M}\omega_3 m_x\\
&\quad + 2g(\gamma_{33}-\gamma_{31})M_x M_z\frac{\partial u_z}{\partial z} + g\gamma_{44}(M_x^2 - M_z^2)\frac{\partial u_x}{\partial z} + r_y,\\
(\omega^2 - \omega_t^2)u_x &= -\frac{\gamma_{44}}{\rho}\left(M_x\frac{\partial m_z}{\partial x} + M_z\frac{\partial m_x}{\partial x}\right),\\
(\omega^2 - \omega_t^2)u_y &= -\frac{\gamma_{44}}{\rho}M_z\frac{\partial m_y}{\partial z},\\
(\omega^2 - \omega_t^2)u_z &= -\frac{2M_z}{\rho}\left(\gamma_{33}-\gamma_{31}+\frac{\gamma_1}{2}\right)\frac{\partial m_z}{\partial z} - \frac{\gamma_1 M_x}{\rho}\frac{\partial m_x}{\partial z},\\
-i[k,h] &= (4\pi\sigma/c)e, \quad c[k,e] = \omega(h + 4\pi m),
\end{aligned} \tag{16}$$

where

$$\omega_t^2 = c_{44}k^2/\rho = s_t^2 k^2, \quad \omega_l^2 = c_{33}k^2/\rho = s_l^2 k^2,$$

$$\omega_1 = \frac{g}{M}\left(M_x H - \beta_1 M_z^2 - 2\beta_2 M_z^4 + \frac{2(\gamma_{11}-\gamma_{12})^2}{c_{11}-c_{12}}M^2 M_x^2 + \frac{\gamma_{44}^2}{c_{44}}M^2 M_z^2\right),$$

$$\omega_2 = gM(4\beta_2 M_z^2 + \gamma_{44}^2 M^2/c_{44}), \quad \omega_3 = gM(H/M_x + \gamma_{44}^2 M^2/c_{44}), \tag{17}$$

with r_i the components of the linearized relaxation term [Eq. (2)].

Analysis of the system of equations (16) shows that in the range $T < T_c$ the efficiency of electromagnetic excitation of longitudinal ultrasound is determined by two mechanisms. The first is brought about by bulk magnetostriction of the paraprocesss which is manifested most effectively for $T \sim T_c$. The second is due to anisotropy of the relativistic magnetostriction.

In order to consider the longitudinal ultrasound generation due to the paraprocess in the region $T < T_c$, we must put $M_z = 0$ in the system of equations (16). This corresponds to the state of the magnetic material after the spin-flip transition along H, when $\mathbf{M}\|\mathbf{H}$. The system of equations (16) then breaks up into two subsystems relative to the variables m_x, u_z, h_x, e_y and m_z, m_y, u_y, u_x, h_y, e_x. For the first group of variables describing the excitation of longitudinal ultrasound, the system (16) takes the form of Eq. (8) with replacement of u_y by u_z; e_z by e_x, γ_2 by γ_1, y by z. The susceptibility is then defined as

$$\chi_\parallel = [a + 6bM_x^2 + b_2(U_{xx}+U_{yy}) + b_1 U_{zz} + 2(b_{11}-b_{12})U_{xx}]^{-1}, \tag{18}$$

where U_{ij} are the equilibrium strains due to magnetostriction.

In the region $T < T_c$ the formulae (9)–(14) remain valid for the ultrasound generation produced by the paraprocess. As the temperature is lowered the efficiency of generation of longitudinal ultrasound via the paraprocess decreases, corresponding to the decrease in the constants γ_i and of the susceptibility (18).

In the canted phase an electromagnetic wave falling on a face of the metal excites in it both longitudinal and transverse elastic waves. The complete system of equations (16) is very complicated to solve exactly. We limit ourselves here to a discussion of the case when the relaxation terms can be omitted. Ultrasound generation under this condition is due to anisotropic magnetostriction.

The dispersion-equation system (16) in the frequency region $\omega \ll \omega_{s0}$, where ω_{s0} is the frequency of homogeneous oscillations of the magnetization, has the form

$$\begin{aligned}&[\omega^2-s_l^2k^2(1-\zeta_l)][\omega^2-s_t^2k^2(1-\zeta_t)](ik^2\delta^2/2-\mu_\parallel)\\&+[\omega^2-s_l^2k^2(1-\zeta_t)]4\pi\chi_\parallel s_l^2k^2\zeta_l+[\omega^2-s_l^2k^2(1-\zeta_l)]4\pi\chi_\parallel s_t^2k^2\zeta_t\\&-(ik^2\delta^2/2-\mu_\parallel)s_l^2s_t^2\zeta_t\zeta_lk^4-8\pi\chi_\parallel s_l^2s_t^2\zeta_l\zeta_tk^4=0,\end{aligned} \tag{19}$$

where

$$\begin{aligned}&\omega_{s0}^2=\omega_1\tilde{\omega}_2,\quad \tilde{\omega}_2=(M_x^2\omega_2+4\pi gMM_x^2+\omega_3M_z^2)/M^2,\\&\mu_\parallel=1+4\pi\chi_\parallel,\quad \chi_\parallel=gM_z^2/M\tilde{\omega}_2,\quad \zeta_t=\gamma_{44}^2\chi_\perp(M_x^2-M_z^2)/c_{44},\\&\zeta_l=4\chi_\parallel M_x^2(\gamma_{33}-\gamma_{31})^2/c_{33},\quad \chi_\perp=g(M_x^2-M_z^2)/M\tilde{\omega}_2.\end{aligned} \tag{20}$$

It follows from an analysis of the expressions for the susceptibilities $\chi_\parallel$ and $\chi_\perp$ and for the magnetoelastic interaction parameters ζ_t and ζ_l that efficient excitation of longitudinal and transverse ultrasound takes place for different values of the external magnetic field. Since the maximum value of the parameter ζ_t is reached either at $M_x = M$ or for $M_z = M$, while for the parameter ζ_l it is at $M_x = M_z$, a separate consideration of the generation of longitudinal and transverse ultrasound is permissible. Therefore in studying the excitation of longitudinal ultrasound we will neglect the parameter ζ_t in Eq. (19). The boundary conditions for Eq. (16) in this case have the form:

$$\begin{aligned}&h_{1x}+h_{2x}=h_{0x}+h_{3x},\quad e_{1y}+e_{2y}=e_{0y}+e_{3y},\\&-ik_1c_{33}u_{1z}+\gamma_1(M_xm_{1x}+M_zm_{1z})+2(\gamma_{33}-\gamma_{31})M_zm_{1z}\\&-ik_2c_{33}u_{2z}+\gamma_1(M_xm_{2x}+M_zm_{2z})+2(\gamma_{33}-\gamma_{31})M_zm_{2z}=0.\end{aligned} \tag{21}$$

A calculation of the amplitude of the ultrasound excited in the ferromagnetic phase of a metal via anisotropic magnetostriction leads to the following expression:

$$u_{2z}=-\frac{2ik_2'(\gamma_{33}-\gamma_{31})M_x\chi_\parallel h_{0x}}{\rho s_t^2(1-\zeta_l)(k_1^2-k_2'^2)}. \tag{22}$$

The wave numbers k_1 and k_2 in Eqs. (21) and (22) are determined by Eqs. (11) and (12) with the replacement of $\tilde{\mu}_\parallel$ by $\mu_\parallel$, $\tilde{\zeta}$ by ζ_l, and $\tilde{\chi}_\parallel$ by $\chi_\parallel$. The quantities $\mu_\parallel$, ζ_l, and $\chi_\parallel$ are determined by Eq. (20).

In the geometry when $\mathbf{H}\|\mathbf{h}\|\mathbf{a}$ and $\mathbf{k}\|\mathbf{n}\|\mathbf{b}$, Eq. (22) remains valid for the ferromagnetic phase of gadolinium and a corresponding change or variables has to be carried out.

4. DISCUSSION OF THE RESULTS

Over a wide range of variation of magnetic field and temperature, the generation of ultrasound in gadolinium proceeds both via isotropic magnetostriction of the paraprocess and via anisotropic magnetostriction associated with spin-flip transitions. To first-order approximation these magnetoelastic interactions can be considered independent of one another, while their contributions to the processes of electromagnetic-acoustic transformation are additive.

The signal measured in the experiment is proportional to the ultrasound generation efficiency, defined as the ratio of the fluxes of acoustic ($W_{ac} = \rho\omega^2u^2S/2$) and electromagnetic ($W_{em} = ch^2/4\pi$) energy onto the face of the magnetic material. Using the expressions for the amplitude of elastic waves excited on account of the paraprocess, Eq. (14) or due to spin-flip, Eq. (22), we obtain in the general case

$$K=\frac{W_{ac}}{W_{em}}=\pi\frac{S}{c}\frac{\zeta(1-\zeta)(1+4\pi\chi)^{3/2}}{(1+4\pi\chi-\zeta)^{7/2}}\left(\frac{\omega\delta}{s_l}\right)^4\chi. \tag{23}$$

The values of the parameters of magnetoelastic ζ and magnetic susceptibility χ which are to be used here are determined by Eq. (10) or Eq. (20).

We will first consider the features of the transformation due to the paraprocess. This conversion mechanism is most efficient in the region of the transition from the paramagnetic to the ferromagnetic state. The magnetoelastic coupling parameter ζ is small compared with unity over the whole temperature range, so that we can simplify Eq. (23):

$$K=\text{const}\cdot\frac{\zeta\chi}{(1+4\pi\chi)^2}. \tag{24}$$

Far from the Curie temperature the paramagnetic susceptibility $\chi \ll 1$; hence $K \propto \zeta\chi$. For a fixed value of the magnetic field $H \neq 0$ the magnetization M changes with temperature monotonically, while χ increases rapidly on approaching T_c. This explains the observed sharp growth in the amplitude of generation of longitudinal ultrasound in the vicinity of the Curie temperature (Fig. 2).

By starting from Eq. (24) the field dependences of the efficiency of excitation of longitudinal ultrasonics in the region of the transition from the paramagnetic to the ferromagnetic state (curve 1 of Figs. 1 and 3) can be explained in the following way. In weak fields near the Curie temperature the homogeneous exchange constant $a \approx 0$ and the magnetic susceptibility $\chi_\parallel > 1$. Since $\chi_\parallel$ depends weakly on H, it follows that $K \propto M_x^2$, i.e., it increases with increasing magnetic field. In strong fields the susceptibility $\chi_\parallel \ll 1$. Then $K \propto M_x^{-2}$, i.e., it decreases with increasing magnetic field. Thus, the efficiency of electromagnetic generation should reach a maximum at some value of the magnetic field. A dependence of this type is observed near T_c not only in the paramagnetic but also in the ferromagnetic region. For $T < T_c$ Eqs. (9)–(14) which describe the paraprocess remain valid, but expression (18) for the susceptibility must be used in them.

We shall now consider the efficiency of electromagnetic-acoustic transformation via spin-flip. The contribution of this transformation mechanism is determined not only by the magnitude but also by the orientation of the steady magnetic field relative to the crystallographic axes of the specimen. The excitation of ultrasound through spin-flip can be

interpreted in the following way. In the ferromagnetic phase the equilibrium orientation of the magnetization **M** relative to the crystallographic axes is determined by the values of the magnetic anisotropy constants β_i for a given temperature and strength of the field H. In the tangential geometry $\mathbf{H}\|\mathbf{h}\perp\mathbf{n}$ the alternating field h modulates the field H and, consequently, the direction of the vector **M**. In conformity with Eq. (22), a change in the direction of **M** is accompanied by a deformation of the crystal lattice; the amplitude of the elastic waves excited is then determined by the coresponding component of the anisotropic magnetostriction tensor.

The structure of Eqs. (23) and (24) is preserved for this case, but it must be borne in mind that spin-flip takes place in a limited magnetic-field range. The generation of ultrasound by the spin-flip transition in a single-domain specimen should start from zero magnetic field, reach a maximum, and end in a field at which the vector **M** assumes the direction of **H**. The experimentally observed dependences, however, show appreciable differences. First, generation usually starts from some field H_1, not equal to zero (curve 3 in Fig. 1 and curves 2 and 3 in Fig. 3); second, the generation in the region of relatively high temperatures does not end at a field H_2, but reaches a level that depends weakly on the magnetic field (curves 2 in Figs. 1 and 3).

In order to explain the experimentally observed dependences in the ferromagnetic region it is essential to take account of the real domain structure and of the demagnetization factor of the specimens, the orientation of the field, and also of the paraprocess magnetostriction. The paraprocess is insignificant far from the Curie temperature and generation takes place only in some interval of magnetic fields limited from above and below, $H_1 < H < H_2$. The absence of generation of ultrasound in fields $H < H_1$ can be explained by the fact that so long as the specimen contains domains unfavorably distributed relative to H, the internal magnetic field H_{in} in the magnetic material is zero. In fields $H < H_1$ an increase in the strength of the steady magnetic field and its modulation by the field of the electromagnetic wave only leads to a shift of the domain walls and to a change in the ratio of domains with different directions of the spontaneous magnetization vector. The magnetostriction of processes associated with a shift in domain walls is apparently small, which leads to a low efficiency of the elastic-wave excitation. In the range of magnetic fields $H_1 < H < H_2$ the vector **M** rotates away from the direction of spontaneous magnetization to the direction of the external field **H**. This process is accompanied by a sharp increase in the efficienicy of electromagnetic-acoustic conversion. Finally, in fields $H > H_2$ the magnetic moments of the atoms are oriented along **H** and generation of ultrasound due to anisotropic magnetostriction does not take place.

As the temperature increases, isotropic magnetostriction of the paraprocess is added to the process of electromagnetic-acoustic transformation. However, unlike the situation in the paramagnetic region, generation of ultrasound in the ferromagnetic phase via the paraprocess takes place in fields $H > H_1$. In the magnetic field interval $H_1 < H < H_2$ ultrasound is excited both on account of the isotropic and the anisotropic magnetoelastic interactions, and finally in fields $H > H_2$ it is due only to the paraprocess.

The case $T_{sf} < T < T_c$ is interesting. If **H** is directed along the easy-magnetization axis C, then in the field H_1 the specimen goes over into the monodomain phase in which $\mathbf{M}\|\mathbf{H}$, i.e., H_1 coincides with H_2. In this case, although generation also starts at H_1, it takes place only through magnetostriction of the paraprocess (curve 2 of Fig. 3). If the field H lies in the basal plane of the crystal, spin-flip starts from zero magnetic field (curve 2 of Fig. 1).

In situations when both the paraprocess and spin-flip show up with comparable efficiency in the processes of transformation of waves, the values of the field H_2 were determined from the sharp break in the $A(H)$ curve past the maximum value (curves 2 in Figs. 1 and 3).

The analysis of the $A(H)$ dependences explains the temperature variation of the generation efficiency shown in Fig. 2. The plots 1–3 were obtained respectively in fields $H < H_1$, $H_1 < H < H_2$ and $H > H_2$. In weak and strong magnetic fields generation of longitudinal ultrasonics is efficient only in the region of the transition of the metal from the paramagnetic to the ferromagnetic state. In the intermediate region $H_1 < H < H_2$ generation of comparable efficiency is observed near the Curie temperature (paraprocess) and at low temperatures (spin-flip).

5. *H-T* PHASE DIAGRAMS

If the interpretation of the experimental results which has been carried out above is correct, then the *H-T* phase diagrams of gadolinium can be established from the temperature dependences of the magnetic fields H_1 and H_2 in which sharp changes in the efficiency of electromagnetic-acoustic conversion take place, i.e., the regions of existence in it of monodomain, canted (the magnetization M not parallel to the field H), and collinear ($\mathbf{M}\|\mathbf{H}$) phases can be determined.

We shall first consider the case of the magnetic field parallel to the hexagonal axis $\mathbf{H}\|\mathbf{h}\|\mathbf{c}$. The processes of the shift in domain walls in the specimen ends at $H_1 = DM\cos\theta$, where θ is the angle between the direction of the magnetization **M** and the **c** axis, and D is the demagnetization factor. In the temperature interval $T_{sf} < T < T_c$ the angle θ is equal to zero and the specimen immediately goes over from the monodomain into the collinear phase (i.e., $H_1 = H_2$). For $T < T_{sf}$ the angle θ is non-zero and is temperature dependent. A reorientation of the magnetic moments of the atoms into the diretion of H starts from the field H_1 and gadolinium is then in the canted phase. The reorientation ends at the field H_2 and gadolinium goes over into the collinear phase. In order to find $H_2(T)$ we write out that part of the free energy density of the crystal which depends on the angle θ:

$$F = {}^1/_2\beta_1 M^2\cos^2\theta + {}^1/_2\beta_2 M^4\cos^4\theta + {}^1/_2\beta_3 M^6\cos^6\theta - H_{in}M\cos\theta. \qquad (25)$$

By minimizing F with respect to θ and assuming that $\theta = 0$, we find an expression for H_2:

$$H_2 = DM + (\beta_1 + 2\beta_2 M^2 + 3\beta_3 M^4)M.$$

In the situation when the steady magnetic field lies in the basal plane $\mathbf{H}\|\mathbf{h}\|\mathbf{a}$, the processes of the shift of the domain walls ends at the field $H_1 = DM\sin\theta$. In the interval $T_{sf} < T < T_c$ we have $\theta = 0$ and $H_1 = 0$ unlike the case considered above ($\mathbf{H}\|\mathbf{c}$), in this geometry the reorientation of the spins occurs over the whole temperature range. The free

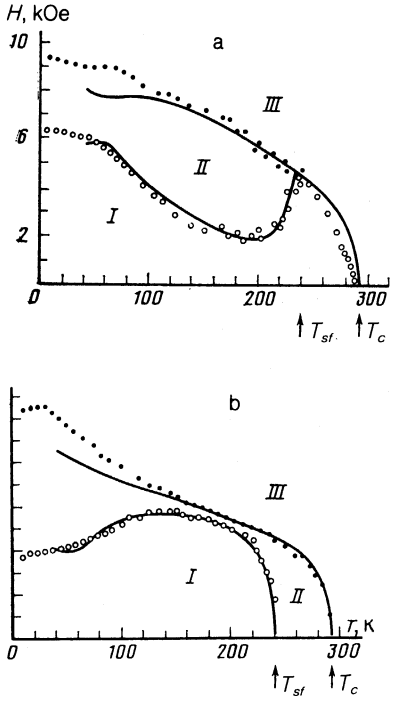

FIG. 5. a) H-T phase diagrams for GD for **H**||**c**. Points ○) H_1 ●) H_2 are experimental; the lines are calculated. Regions I, II, and III correspond to single-domain, canted and collinear phases. b) the same for **H**||**a**.

energy density is written in the form of Eq. (25), similar to that before, with the substitution in the last term on the right hand of cos θ by sin θ. Rotation of the magnetic moments ends in the field

$$H_2 = DM - \beta_1 M.$$

The fields H_1 and H_2 are evaluated by using results on β_i and M, given by Belov *et al.*[6] Since the electromagnetic-acoustic conversion takes place in a thin sub-surface layer, the calculation of the demagnetizing factor D was carried out for the field acting on the surface of the crystal. For the field orientations considered for specimen No. 1 the demagnetizing factor $D = 3.2$. The calculated and experimental $H_1(T)$ and $H_2(T)$ dependences for two orientations are shown in Fig. 5. It can be seen that fairly good agreement is observed between the experimental and theoretical dependences which then determine the boundaries of the magnetic phases of gadolinium.

An H-T phase diagram of gadolinium in the temperature range 180–290 K in a magnetic field **H** perpendicular to the hexagonal axis was constructed by Martin *et al.*[9] starting from an analysis of the temperature and field dependences of the elastic modulus c_{33}. In the overlapping temperature region, the H-T diagrams obtained by us (Figs. 5 and 6) and by Martin *et al.*[9] practically coincide.

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