Nonlinear generation of ultrasound in PbTe(Ga)

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(Submitted 12 September 1994; resubmitted 12 October 1994)
Zh. Eksp. Teor. Fiz. **107**, 236–244 (January 1995)

A new mechanism for nonlinear electromagnetic excitation of ultrasound has been discovered in a narrow-gap semiconductor PbTe(Ga), which is characterized by a retarded photoconductivity. The excitation amplitude at twice the frequency of the electromagnetic wave is proportional to the static magnetic field, and does not depend on the conductivity of the material. This effect is explained under the assumption that localized magnetic moments exist at the impurity atoms in the presence of antiferromagnetic interaction between these moments. © *1995 American Institute of Physics.*

1. INTRODUCTION

Generation of acoustic oscillations in solids by an electromagnetic wave can occur in a linear regime, when the frequency of the incident wave is equal to the frequency of the excited wave, and in a nonlinear regime, when the frequency of the excited elastic waves is a multiple of the frequency of the electromagnetic wave. In insulators the linear excitation of ultrasound is attributable to the piezoelectric effect, while the nonlinear excitation stems from the electrostriction and higher-order piezoelectric interactions. In magnetic materials the linear excitation of ultrasound occurs as a result of the piezomagnetic effect, while the nonlinear excitation of ultrasound is caused by magnetostriction. In the presence of a static magnetic field, magnetostriction leads to an excitation of ultrasound at the frequency of the electromagnetic wave. In materials that are good conductors, both linear and nonlinear electromagnetic excitations of ultrasound occur as a result of interaction of the electromagnetic wave with the conduction electrons. The most comprehensively studied mechanism for excitation of ultrasound in conductors is the inductive interaction of an alternating current induced by an electromagnetic wave in the skin layer with the static magnetic field.[1] In the absence of a static magnetic field, the inductive interaction leads to an excitation of ultrasound at twice the frequency, while in the presence of a magnetic field, the ultrasound is excited at the electromagnetic wave frequency.[2,3] The characteristic feature of this excitation mechanism is the proportionality of the amplitude of the excited elastic vibrations of the static magnetic field. In insulators the induction mechanism for ultrasound excitation is inefficient, since the depth to which an electromagnetic wave penetrates a substance is greater than the wavelength of elastic vibrations.[4]

The use of electromagnetic excitation of ultrasound to study the elastic properties of a narrow-gap semiconductor PbTe(Ga), whose conductivity may vary by several orders of magnitude upon exposure to light at low temperature, has shown that even when this material is in the insulating state, ultrasound can be excited at twice the frequency of an electromagnetic wave whose amplitude is proportional to the static magnetic field. The characteristic features of conversion of the electromagnetic and acoustic waves in PbTe(Ga) indicated above cannot be described by any conversion mechanism mentioned above. In this article we report an experimental study and a theoretical analysis of a nonlinear electromagnetic–acoustic conversion in this material.

2. EXPERIMENTALLY STUDIED SAMPLE

Lead telluride is a narrow-gap semiconductor which crystallizes in an fcc lattice. The unsaturated electron bonds of the Pb and Te atoms account for the anomalously large dielectric constant of this compound.[5] Because of the deviation from stoichiometry, PbTe usually has a large hole concentration even at low temperatures. Doping of PbTe with gallium to a level $N_{Ga}=(1-3)\times10^{19}$ cm^{-3} causes the original material to become an insulator, while below $T_c=80$ K a retarded photoconductivity has been observed in PbTe(Ga).[6] Exposure of a crystal sample of this compound to a source of visible or infrared light at low temperatures is accompanied by an increase in its conductivity by seven or eight orders of magnitude, which persists after the removal of the light. PbTe(Ga) can be returned to the high-resistivity state only by warming it to a temperature above the critical temperature T_c and then cooling it in a shielded chamber.

Various physical models have been used to explain the retarded photoconductivity.[7–9] Among these models for PbTe(Ga), the hypotheses based on the assumption that the crystalline environment of an impurity changes as a result of the change in its charge state seem to be justifiable.[8,9] To verify this assumption, the temperature dependence of the velocity s and damping γ of the longitudinal ultrasound in PbTe(Ga) has been measured.[10] These experiments have shown that, first, the elastic moduli of PbTe decrease appreciably when it is doped with gallium and, secondly, the s and γ anomalies are present at the critical temperature T_c. Measurements of the elastic constants of PbTe(Ga) and analysis of the doping of PbTe produced by Ga have made it possible to formulate a conceptual explanation of the restructuring of the crystalline environment of the impurity by using models for the structure of the electron shells of Ga in PbTe. This explanation essentially reduces to the argument that the Pb atoms in the lead telluride are replaced by Ga atoms in the inverted electronic configuration s^1p^2. The electrons in the p orbitals participate in the establishment of ionic/covalent

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bonds with Te atoms, while the electron from the unfilled inner shell, driven by visible or IR light, can move to the conduction band, leaving the Ga atom in the s^0p^2 configuration. Since the atomic radius of Ga in the electronic s^1p^2 configuration is larger than in the s^0p^2 configuration, the transition from one state to another is accompanied by the displacement of Te atoms in the direction of Ga. The deformation of the immediate neighborhood of the impurity in the model proposed by us has a symmetry center. This deformation gives rise to an energy barrier which accounts for the slow relaxation processes in PbTe(Ga). According to the proposed model, the electronic s^1p^2 configuration based on Ga atoms has localized magnetic moments, between which short-range magnetic order can in principle be established.

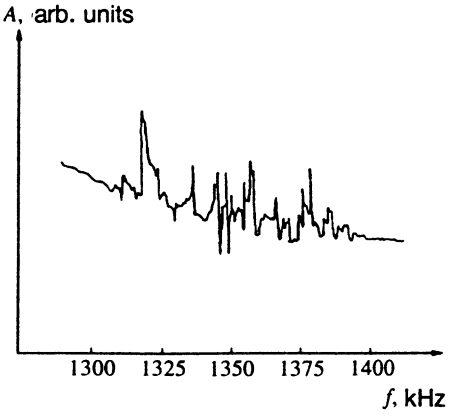

FIG. 1. Spectrum of acoustic resonances in a PbTe platelet. $T=4.2$ K, $H=70$ kOe, and $d=1.4$ mm.

3. EXPERIMENT

The measurements were carried out using PbTe and PbTe(Ga) single crystals. The Ga concentration in the doped sample amounted to 0.3% by number. The crystals were platelets with a cross section of about 1 cm and thickness of about 0.1 cm. The normals to their surfaces coincided with the crystallographic [001] axes. A platelet was placed inside two coaxial induction coils, one of which was used to excite elastic vibrations and the other recorded the surface impedance of the platelet when ultrasonic standing waves were set up in it. The signal from the output of an rf oscillator was sent to a given coil and the signal induced in the pickup coil was sent to a chart recorder after it was amplified and detected. The measurements were carried out in the frequency range 10^5–10^7 Hz.[11] The sample with coils enclosing it and the miniature incandescent lamp used to illuminate the sample at low temperatures were situated in a shielded chamber which in turn was placed in the warm-field channel of a superconducting solenoid. This arrangement allowed us to carry out the measurements in a magnetic field up to 80 kOe over the temperature range 4–100 K. Electrical contacts soldered to the platelet were used to monitor its resistance.

The conversion of electromagnetic waves to ultrasonic waves was studied by exciting the elastic standing waves in plane-parallel platelets.[12] When a platelet is inserted into an induction coil, the alternating electric fields at its surfaces, $z=d/2$ and $z=-d/2$, are antiparallel

$$e(d/2)=-e(-d/2), \tag{1}$$

and the alternating magnetic fields are parallel

$$h(d/2)=h(-d/2). \tag{2}$$

This corresponds to the case of antisymmetric excitation of elastic standing waves[13] which are set up in the interior of the platelet when

$$f=ns/2d, \quad n=1,3,\ldots . \tag{3}$$

We studied experimentally only the first ($n=1$) acoustic resonance, because of the increasing attenuation γ of ultrasound and the decreasing efficiency η of the electromagnetic–acoustic conversion resulting from the increase in the frequency. The resonance features of the surface impedance in such an experimental arrangement are

$$\Delta Z=\eta/\gamma. \tag{4}$$

For the induction mechanism for ultrasound excitation we have

$$\eta_{\text{ind}}=\frac{s}{c}\,\frac{H_0^2}{8\pi\rho s^2(1+\beta^2)}, \tag{5}$$

where H_0 is the static magnetic field, c is the velocity of light, ρ is the density of the material, $\beta=2\pi^2\delta^2/\lambda^2$, δ is the thickness of the skin layer, and λ is the length of an elastic wave.

The control measurements were performed with a PbTe single crystal, which had a large concentration of free charge carriers and which was characterized by a metallic resistivity. A static magnetic field H_0 in the direction of the alternating magnetic field h was applied to the surface of the plate, which caused the longitudinal ultrasound to be excited. The trace of the acoustic resonances in PbTe is shown in Fig. 1. The location of the resonance group center, which is determined, according to (3), by the velocity of longitudinal sound, coincides within small error limits with the tabular data for s.[14] The appearance of a large number of resonance features in the frequency dependence of the surface impedance of PbTe reflects, on the one hand, the complex spectrum of the intrinsic elastic oscillations of a platelet. On the other hand, it indicates that the ultrasound is attenuated only slightly in this crystal. As the static magnetic field increases, the amplitudes of the acoustic resonances increase in proportion to the square of this field. These data indicate that the excitation of ultrasound in PbTe is attributable to the induction mechanism for conversion of the electromagnetic and elastic waves.

The excitation of ultrasound in PbTe(Ga) is fundamentally different in nature. In a chamber shielded from external light, a PbTe(Ga) sample exhibited semiconductive behavior of the resistivity, reaching values of $\rho\sim10^6$ $\Omega\cdot$cm at $T=4.2$ K. Exposure of the sample to light resulted in a reduction of ρ to values of $\sim10^{-2}$ $\Omega\cdot$cm. In either case, the parameter calculated on the basis of these data is $\beta\gg1$, which renders the induction mechanism of linear excitation ineffective. As is evident in Fig. 2, strong excitation of elastic standing

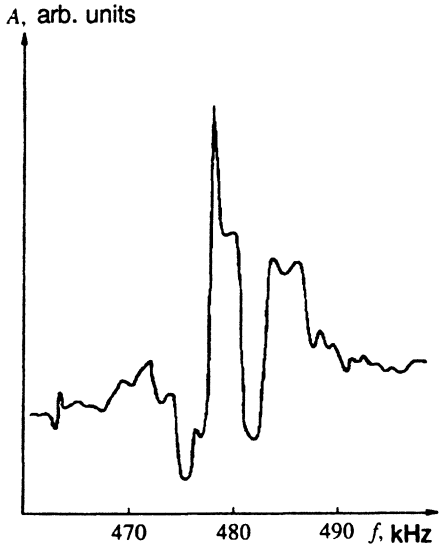

FIG. 2. Spectrum of acoustic resonances in a PbTe(Ga) platelet. $T=4.2$ K, $H=70$ kOe, and $d=0.86$ mm.

waves was observed nonetheless in PbTe(Ga) at low frequencies. The amplitudes of these resonances do not change upon exposure of the sample to light. The location of the center of the resonance group, in terms of the frequency, corresponds to the case in which $f=s/4d$. In the case of antisymmetric excitation this corresponds to the excitation of longitudinal ultrasound at twice the frequency of the electromagnetic wave. We see in Fig. 3 that the amplitudes of the acoustic resonances are proportional to the square of the static magnetic field.

We have thus observed experimentally a nonlinear excitation of ultrasound in a poorly conducting medium at an efficiency which depends on the magnetic field. The body of data which have been obtained cannot be described by any conversion mechanism mentioned above.

4. THEORY

To solve the problem of the electromagnetic excitation of ultrasound in a poorly conducting medium in a static magnetic field, we will use Maxwell's equations and the elasticity equation

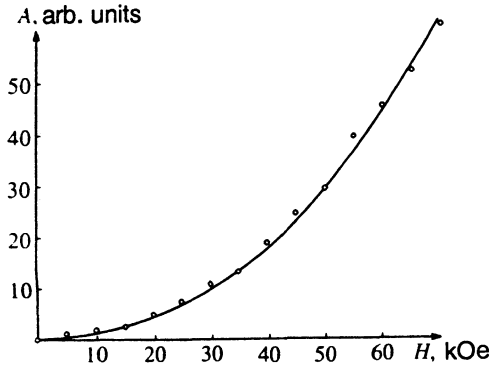

FIG. 3. Field dependence of the acoustic resonances in a PbTe(Ga) platelet.

$$\text{rot } \mathbf{h}=\frac{\varepsilon \partial \mathbf{e}}{c\partial t}, \tag{6}$$

$$\text{rot } \mathbf{e}=-\frac{\mu \partial \mathbf{h}}{c\partial t}, \tag{7}$$

$$\text{rot } \rho_M u_i=\frac{\partial \sigma_{ij}}{\partial x_j}. \tag{8}$$

Here h and $e \propto \exp(-i\omega t)$ are the magnetic and electric fields of the electromagnetic wave, u is the bias voltage, ε and μ are the dielectric constant and the permeability of the material, ρ_M is the density of the material, $\hat{\sigma}=\partial F/\partial \hat{U}$ is the stress tensor, F is the free energy, and $\hat{U}$ is the strain tensor.

In general, the stress tensor can be expanded in powers of the strain tensor $\hat{U}$, the polarizability tensor $\hat{P}$, and the magnetization tensor $\hat{M}$ (it is assumed that $\hat{P}$ and $\hat{M}$ are tensors of first rank, i.e., vectors; to simplify the notation, the caret above the tensor quantities is omitted below):

$$\begin{aligned}\sigma=&a_1^{\text{IV}}U+a_2^{\text{III}}P+a_3^{\text{III}}M+b_1^{\text{VI}}UU+b_2^{\text{V}}UP+b_3^{\text{V}}UM\\&+b_4^{\text{IV}}PP+b_5^{\text{IV}}PM+b_6^{\text{IV}}MM+c_1^{\text{VIII}}UUU\\&+c_2^{\text{VII}}UUP+c_3^{\text{VII}}UUM+c_4^{\text{VI}}UPM+c_5^{\text{VI}}UPP\\&+c_6^{\text{VI}}UMM+c_7^{\text{V}}PPP+c_8^{\text{V}}PPM+c_9^{\text{V}}PMM\\&+c_{10}^{\text{V}}MMM.\end{aligned} \tag{9}$$

Here the equilibrium elastic properties of the crystal are described by the term $a_1^{\text{IV}}U$, the terms $a_2^{\text{III}}P$ and $a_3^{\text{III}}M$ describe the piezoelectric and piezomagnetic effects, the terms $b_1^{\text{VI}}UU$ and $c_1^{\text{VIII}}UUU$ characterize the elastic nonlinearity of the crystal, the terms $b_2^{\text{V}}UP$ and $b_3^{\text{V}}UM$ determine the renormalization of the equilibrium elastic modulus which occurs as a result of the inverse piezoelectric and piezomagnetic effects, $b_4^{\text{IV}}PP$ and $b_6^{\text{IV}}MM$ determine the electrostriction and the magnetostriction, and the remaining terms describe the cross interactions and higher-order effects. Roman numerals in expression (9) denote the tensor rank.

This expression can be simplified using symmetry arguments, and by ignoring the renormalization of the equilibrium elastic modulus and the terms which do not contain the magnetic field, in conformity with the statement of the problem. The contribution to σ that is attributable to renormalization of the bulk moduli of the material as a result of the action of the electric and magnetic fields, is small in comparison with the piezoelectric and magnetostriction effects and effects similar to them. This circumstance allows us to disregard terms with the coefficients b_1^{VI}–b_3^{V} and c_1^{VIII}–c_6^{VI}. Terms with the coefficients a_2^{III}, b_4^{IV}, and c_7^{V} can be ignored compared with the terms that contain M. If a crystal has a symmetry center, then the coefficients multiplying the terms linear in the electric field are $b_5^{\text{IV}}=c_9^{\text{V}}=0$. Time reversal symmetry enables us to drop terms with the coefficients a_3^{III}, c_8^{V}, and c_{10}^{V} which contain odd powers of M.

The nonequilibrium correction to the components of the elastic stress tensor is then

$$\sigma_{ij}=b_{6\,ijkl}^{\text{IV}}M_kM_l. \tag{10}$$

This equality describes the bulk magnetostriction, which gives rise to only a linear excitation in the presence of a static magnetic field H_0:

$$\sigma = b_6^{IV} \chi_0 \chi H_0 h_0 e^{-i\omega t}, \tag{11}$$

where χ_0 and χ are the static and dynamic magnetic susceptibilities, and h_0 is the amplitude of the alternating magnetic field in vacuum.

In a crystal with a symmetry center, nonlinear excitation of ultrasound can be obtained by admitting the existence of localized magnetic moments with antiferromagnetic order. The elastic stress tensor σ in this case must be expanded in powers of the strain tensor U, of the polarization vector P, of the magnetization vector M, and of the antiferromagnetism vector L. Time reversal in this case does not eliminate the terms with the coefficients a_3^{III}, c_8^V, and c_{10}^V, while the expression for the nonequilibrium correction to the elastic stress tensor becomes

$$\sigma = a_3^{IV} LM + b_6^{IV} MM + c_8^{VI} PPLM + c_{10}^{VI} LMMM. \tag{12}$$

Allowance for the antiferromagnetism vector in the expansion of σ leads to a renormalization, which increases the rank of the corresponding tensors. The first term on the right-hand side of this expression describes piezomagnetism—the linear excitation of ultrasound in the absence of a static magnetic field. The second term, which describes magnetostriction, yields the nonlinear excitation of ultrasound only in the absence of a static magnetic field. Only the third and fourth terms therefore correspond to nonlinear excitation in a static magnetic field:

$$\sigma = (c_8^{VI} \chi_0 \kappa^2 L H_0 e_0^2 + c_{10}^{VI} \chi_0 \chi^2 L H_0 h_0^2) e^{-2i\omega t}, \tag{13}$$

where κ is the rf dielectric susceptibility, and e_0 is the amplitude of the alternating electric field in vacuum.

The problem of electromagnetic excitation of ultrasound involves the analysis of normally incident electromagnetic waves at the boundary of a half-space. The distribution of the electric field in the medium ($x>0$) can be determined from Maxwell's equations, (6)–(7) with allowance for the standard boundary conditions for the components of the electromagnetic wave h and e at $x=0$. Assuming that at $x<0$ the alternating magnetic field is $h=h_0 \exp(-i\omega t)$, we find

$$e = 2h_0 (\mu/\varepsilon)^{1/2} \exp(iqx - i\omega t), \tag{14}$$

where $q=(\varepsilon\mu)^{1/2}\omega/c$.

Equation (14) can be used to compare the terms on the right side of (13). If we assume that the coefficients c_8^{VI} and c_{10}^{VI} are of the same order of magnitude and $\varepsilon/\mu \gg 1$, then the first and second terms in (13) are related: $\kappa^2\mu/\varepsilon\chi^2 \gg 1$. We can then write the equation for longitudinal elastic waves,

$$\rho_M \frac{(\partial^2 u)}{\partial t^2} - \rho_M s^2 \frac{(\partial^2 u)}{\partial x^2} = 2c_8^{VI} \chi_0 \kappa^2 L H_0 e \frac{(\partial e)}{\partial x}. \tag{15}$$

Using the boundary condition for Eq. (8)

$$(\partial u/\partial x)|_{x=0} = 0, \tag{16}$$

which corresponds to the free surface of the material, we find the bias field in the ultrasonic wave

$$u = \frac{8ic_8^{VI} \chi_0 \mu \kappa^2 q h_0^2 H_0 L}{\rho_M s^2 \varepsilon (4q^2 - k^2)} \left(e^{2iqx} - \frac{2q}{k} e^{ikx} \right). \tag{17}$$

Here $k=2\omega/s$ is the wave vector of the ultrasound. The first term on the right side of Eq. (17) corresponds to elastic displacements which accompany the oscillation of the electromagnetic field in the medium. We are interested, however, in the component of the elastic displacement which propagates at the velocity of sound. Under the condition

$$q/k = (\varepsilon\mu)^{1/2} s/c \ll 1 \tag{18}$$

we obtain an expression for the amplitude of the ultrasound excited at twice the frequency of the electromagnetic wave:

$$|u| = \frac{2c_8^{VI} \chi_0 \kappa^2 \mu^2 h_0^2 s}{\rho_M c^2 \omega} H_0 L. \tag{19}$$

In the experiment on the excitation of ultrasonic standing waves in a platelet, the amplitudes of the acoustic resonances are proportional to the coefficient representing the conversion of electromagnetic waves into ultrasonic waves, η. This quantity is generally defined as the ratio of the acoustic energy flux $P=\rho_M \omega^2 |u|^2 s/2$ to the electromagnetic energy flux $W=c|h_0|^2/8\pi$, at the boundary of the material:

$$\eta = \frac{P}{W} = 16\pi \left(\frac{s}{c} \right)^5 \frac{(c_8^{VI})^2 \chi_0^2 \mu^4 \varkappa^4 h_0^2}{\rho_M s^2} H_0^2 L^2. \tag{20}$$

Expressions (19) and (20) describe the generation of longitudinal ultrasound in an insulator at twice the frequency of an electromagnetic wave incident on its surface. The strength of this wave is proportional to the square of the static magnetic field.

5. DISCUSSION

In deriving the equations describing the nonlinear excitation of ultrasound we assumed that, first, localized magnetic moments, which are ordered antiferromagnetically, are present in the substance. In addition, in choosing the term that makes the dominant contribution in the elastic stress tensor, we based our argument on the assumption that the permeability of the material is small in comparison with its dielectric constant. Finally, evaluating the amplitude of the ultrasound that is excited, we ignored the bias-field component, which propagates at the velocity of the electromagnetic wave in the medium.

Let us determine the extent to which these assumptions are justified. We note, first of all, that PbTe is characterized by an anomalously large static dielectric constant ($\varepsilon_0 \sim 3000$) and dynamic dielectric constant ($\varepsilon_\infty \sim 30$) (Ref. 5). On the other hand, the permeability of this material does not differ markedly from unity. The inequality $\varepsilon/\mu \gg 1$ is therefore always valid. The inequality (18) is also valid even for such large values of ε. Doping of lead telluride with gallium, which causes it to become an insulator at low temperatures, cannot disturb this relation, although the permeability of PbTe(Ga) can, in our view, increase slightly. The assertion that localized magnetic moments can exist at the gallium atoms is based on the model proposed by Vasil'ev *et al.*[10] for the retarded photoconductivity in this material. At a low dop-

ing level (N_{Ga}=0.3%), however, it is difficult to admit the existence of antiferromagnetic order over the entire crystal. It is easier to assume that the magnetic structure of PbTe(Ga) corresponds to the spin glass state.[1)] Antiferromagnetic short-range order can be established in spin glass.

Let us first determine the factors responsible for the nonlinear excitation of ultrasound in PbTe(Ga). Worth noting here is the strong power-law dependence of η, $\eta \sim \kappa^4$. The value of κ rapidly decreases with increasing frequency. This behavior accounts for the maximum efficiency of the proposed mechanism for excitation occurring at low frequencies. The high conversion efficiency stems from the quadratic dependence on the static magnetic field. Finally, this mechanism for nonlinear conversion of electromagnetic and acoustic waves can be realized only in a material with nonzero components of the tensor of cross interaction of the electrostriction and piezomagnetism.

Let us compare the excitation of ultrasound produced as a result of the mechanism (20) described by us here and the excitation of ultrasound produced by the Lorentz force (5):

$$\eta/\eta_{\text{ind}}=128\pi^2(2\pi\omega\rho)^2(c_8^{\text{VI}})^2\chi_0^2\mu^4\kappa^4h_0^2L^2. \qquad (21)$$

Setting $\omega \sim 3\times10^6$ s^{-1}, $\rho \sim 10^{-12}$ esu, $\chi_0 \sim 10^{-6}$, $\mu \sim 1$, $\kappa \sim 10^2$, $h_0 \sim 10^2$ Oe, and $L \sim 10$ Oe, we find that $\eta > \eta_{\text{ind}}$ for $c_8^{\text{VI}} > 10^{-2}$. The components of the tensor c_8^{VI} have not, to the best of our knowledge, been measured previously. The electromagnetic excitation of ultrasound can be utilized to determine these quantities.

We wish to thank K. N. Baranskii, B. A. Volkov, and M. I. Kaganov for many discussions.

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Translated by S. J. Amoretty