ELECTRIC FIELD INFLUENCE ON ELASTICITY OF CRYSTALS

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Abstract

This paper presents the results of an experimental study of how an electric field affects the position of the plane of polarization of a transverse elastic wave and the acoustooptical interaction in the $LiTaO_3$ crystal. It is established that an electric field of $\pm 1\times 10^6$ V/m rotates the plane of polarization of a sound wave with frequency 580 MHz passing through 1 cm of a crystal by 2.5 degree. The variation of the resonance frequency of piezoelectric cavities subjected to a constant electric field is also demonstrated with longitudinal vibrations in $La_3Ga_5SiO_{14}$ crystal rods at temperatures between –60 and +80°C. For a field of $\pm 1{,}5\times 10^6$ V/m and α = 15°, the relative change of the frequency is $\pm 120\times 10^{-6}$. It is shown that $La_3Ga_5SiO_{14}$ crystals offer a set of physical parameters that makes them promising for electrically controlled and nonlinear acoustoelectronic devices.

Introduction

The study of polarization effects in piezoelectric crystals is a very interesting and complex specialization in solid-state physics because of the anisotropy and nonlinear properties of the crystals. The elliptical-polarization effect of a transverse elastic wave consists in a superposition of two plane-polarized transverse elastic waves propagating in the same direction with different phase velocities. The resulting wave has elliptical polarization. This polarization can arise either when the wave propagates in directions close to the optic axis or when an external effect acts on the crystal—for example, an electric field, induced elliptical polarization. The relative variation of the velocity $\boldsymbol{V}$ of plane-polarized transverse elastic waves is determined by the nonlinearity coefficient of the crystal and by the external electric field: $\boldsymbol{\Delta V/V = \gamma E}$, while the phase difference between the transverse waves is $\boldsymbol{\Delta\Phi = \omega\, l\, \Delta V/V^2}$; i.e., the angle of rotation of the polarization ellipse is proportional to the frequency $\boldsymbol{\omega}$ of the elastic wave and the electric field $\boldsymbol{E}$ [1].

The electric field influence on the resonance oscillation frequency or on the phase velocity of elastic waves in piezoelectric crystals makes it possible to determine the nonlinear properties of the crystals. This effect is widely used in different piezoelectric and acoustoelectric devices, such as resonators and tunable delay lines, harmonic generators, mixers, phase shifters, parametric amplifies, and transducers. The effect was first discovered by Tolman on the resonance frequency of contour vibrations in ***GT*** cut crystal oscillators. The pioneering study of this effect on longitudinal mode crystal resonators of different orientations was carried out by Hrushka, who came to a conclusion that this effect cannot be explained by a change in the linear sizes of the resonator because of the inverse piezoelectric effect and electrostriction; instead, the field dependence of the elastic constants of the crystal due to the nonlinear piezoelectric effect should be taken into account [2].

The search for new piezoelectric single-crystalline materials that combine a strong field dependence of the elasticity, low acoustic losses, and a weak temperature dependence of the elasticity is a topical problem in acoustoelectronics and piezoelectronics.

To 1986, more than 10 piezoelectric crystals had been investigated [2], none of them offering such a combination of properties. The crystals exhibit either high acoustic losses (ADP, KDP, and KIO_3), strong temperature dependence of the elasticity ($LiNbO_3$, $Cd_2(MoO_4)_3$, $Bi_{12}SiO_{20}$, and $Bi_{12}GeO_{20}$), or weak field dependence of the elasticity (α-quartz and $LiTaO_3$).

In 1983, the author discovered the temperature stability of the elastic properties of $La_3Ga_5SiO_{14}$ langasite crystals [3] and found that the acoustic Q factor of shear-mode and thickness-mode resonators made of *Y*-cut langasite may be as high as 10^5 [4]. Later, it was also found that an electric field has an effect on the longitudinal resonant vibration frequency in resonators made of XYs/**α**-cut langasite [5].

Experiment

The influence of the electric field on the polarization of transverse elastic waves with frequency 520–530 MHz in class-3 m piezoelectric crystals of lithium niobate and tantalate was investigated in [6, 7] by the reflected echo-pulse method and in $LiTaO_3$ crystal [8, 9] by the Bragg diffraction at a sound wave with frequency 580 MHz.

Lithium tantalate was chosen in the latter paper because, unlike lithium niobate, in it there are crystallographic orientations with zero temperature coefficient of variation of the propagation rate of elastic waves, and therefore it is more promising when developing acoustoelectronic devices that use the effect by which the polarization of an elastic wave is controlled by an electric field [9].

In the experiment, a sample of single-crystal $LiTaO_3$ (0.8X1.2 X1.6 cm) was cut out along the ***X***, ***Y***, and ***Z*** axes. All the faces of the sample were processed according to optical standards: class-14 purity, flatness no worse than 0.5 ring. The inaccuracy in the dimensions was no more than $\pm10^{-2}$ mm; in the orientation, no more than 5 arc min.

A transverse elastic wave was excited in a $LiTaO_3$ sample by an external piezoelectric converter of pure transverse elastic waves—cut ***Y***/+163° $LiNbO_3$ in the form of a cylinder 12 mm long and 5 mm in diameter. When the end of the $LiNbO_3$ converter was placed in a coaxial quarter-wave microwave cavity, pure transverse elastic waves with frequency 580 MHz were excited in the converter in the form of 1 μs pulses with a repetition rate of 1 kHz

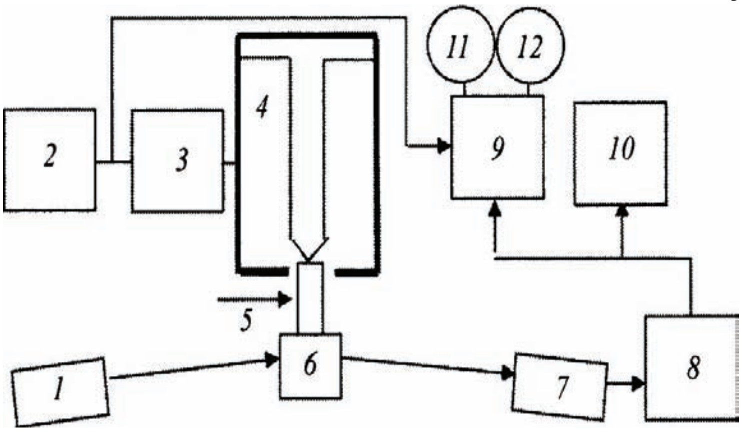

Fig. 1. Block diagram of measurement apparatus. *1*—laser, *2*—modulator, *3*—microwave generator, *4*—microwave cavity, *5*—$LiNbO_3$ converter, *6*— $LiTaO_3$ crystal, *7*—photomultiplier, *8*—amplifier, *9*—measurement unit, *10*—oscilloscope, *11* and *12*—electronic voltmeters.

The intensity of He–Ne laser radiation diffracted at the elastic pulses in $LiTaO_3$ was measured for this paper as a function of the magnitude and polarity of the voltage applied to the metallized ***ZX*** planes of the $LiTaO_3$ sample. An electronic circuit for discriminating and accumulating the video pulses arriving from an FEU-79 photomultiplier made it possible to determine the intensity variation of the diffraction pulses in the $LiTaO_3$ to within 1% at worst.

Figure 2 shows the interaction geometry of light, sound, and electric field in the $LiTaO_3$ crystal, for which the intensity of the light scattered at the sound wave in the Bragg regime is

$$I \sim p_{44}\cos^2(\phi + \varphi) = p_{44}\cos^2(\phi + \beta LE),$$

where p_{44} is the photoelastic constant; φ is the angle of the field-induced rotation of the plane of polarization, measured from the position of the plane for ϕ (+to the left and -to the right); and β is the constant of rotation of the plane of polarization of the incoming wave, which makes an angle of ϕ with the perpendicular to the plane of diffraction.

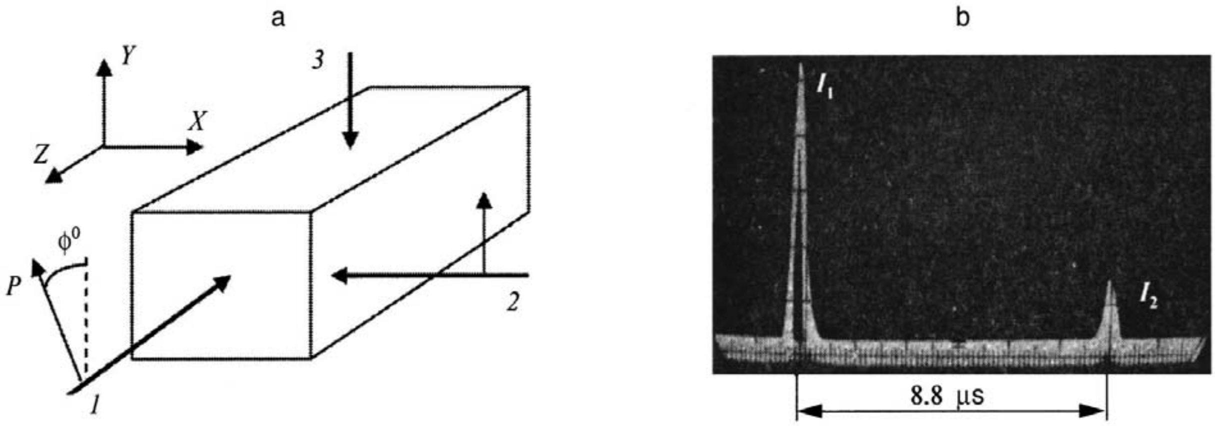

Fig. 2. (**a**)-Interaction geometry of transverse sound wave (**1**) with polarization **P**, light wave (**2**) polarized along **X**, and electric field **E**(**3**) in a $LiTaO_3$ crystal; (**b**)-diffraction pulses (**I₁**)and (**I₂**), induced by the incoming and reflected sound pulses, respectively.

The quantities measured in the experiment were the light intensity scattered at the elastic-wave pulse coming into the crystal, I_1, and that at the first reflected pulse, I_2, passing through distance $L=2l$, as a function of the field $E= u/ d$, where u is the voltage on the crystal, d is the size of the crystal along the field, and l is the length of the crystal.

As shown by experiment, varying the electric field in magnitude and sign in the range from 0 to ± 7.5 10^6 V/m had no effect on the light intensity diffracted at the elastic-wave pulse coming into the crystal. Varying the field in the indicated range affected only the intensity of the light diffracted at the reflected pulse and caused amplitude oscillations of the latter pulse. The pulse amplitude varied from zero when E= + 6 10^6 V/m to the maximum value when E=−3 10^6 V/m

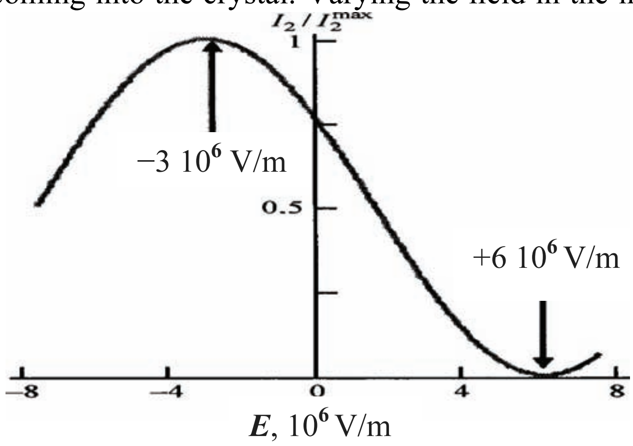

Fig. 3. Relative variation of light intensity I_2 diffracted at a reflected elastic pulse vs applied field in $LiTaO_3$.

The observed variation of the pulse amplitude I_2 for constant pulse amplitude I_1 can be explained by the dependence of the effective photoelastic constant **$P_{ef}= p_{44} \cos(\theta +2\beta lE)$** on the magnitude and sign of field E. The pulse amplitude I_2 actually must depend on the field if the observed effect is axial in the sense that the path difference of the polarized waves continues to increase when the elastic wave is reflected, as in the Faraday acoustic effect.

Two values must be observed for field E at which the pulse amplitude I_2 will be either maximal, when the field rotates the plane of polarization of the elastic wave by angle φ_m so that $\varphi_m=-\phi$, or zero, when the field rotates the plane of polarization by φ_0 so that $\varphi_0=\phi$.

The curve showing the dependence of I_2/I_2^{max} on E, where I_2 is the pulse amplitude for E=0− (±7.5) 10^6 V/m and I_2^{max} is the maximum pulse amplitude, is used to calculate the dependence of angle φ on field E, whence the constant of rotation of the plane of polarization of the transverse elastic wave at a frequency of 580 MHz is β =2.5 10^{-3}deg/V. For the case of sound along Z and the field along Y, γ =4.3·10^{-11} m/V, which coincides in order of magnitude with the γ value for $LiTaO_3$ and $LiNbO_3$.

In this work, we also studied the dependence of the series resonance frequency of langasite resonators on applied field E in the case of the transverse electro-elastic effect (when the applied field is aligned with the weak exciting variable field and is directed normally to the longitudinal axis of the piezoelectric resonator). Fifteen langasite resonator of three (*XYs*/0, *XYs*/10°, and *XYs*/15°) orientations were studied. The dimensions of the resonators were 20 mm in the Y axis direction (the [1010] direction), 3.5 mm in the Z axis direction (the [0001] direction), and 0.5 mm in the X axis direction (the [1120] direction) (Fig. 4).

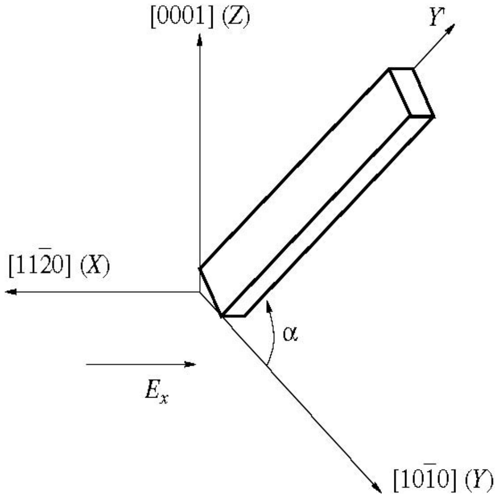

Fig. 4. Shape and crystallographic orientation of the langasite crystals.

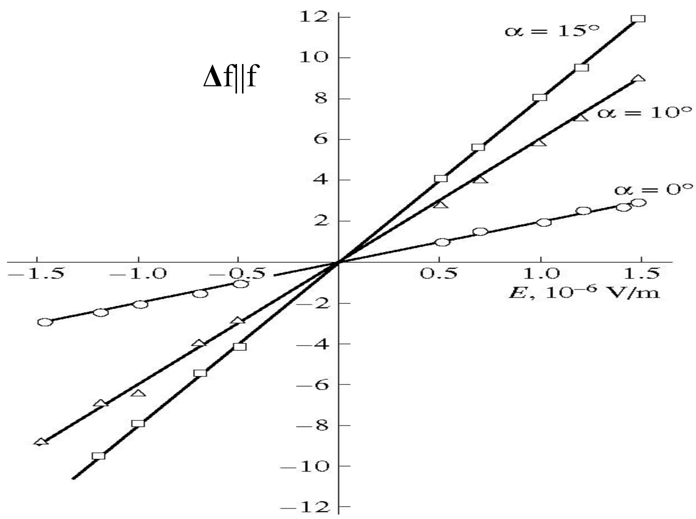

Fig. 5. Linear dependence of the relative change in the frequency on the constant electric field for three orientations of the langasite rods vibrating in the compression–tension mode along the length.

For these dimensions, the longitudinal vibration frequency was equal to ≈115 kHz. The inaccuracy in the dimensions was no more than ±10^{-2} mm; in the orientation, no more than 15 arc min. With the YZ faces nickel-plated, the mechanical **Q** factor was no less than 3×10^4. Langasite samples were inserted in the circuit of a TGK-1 generator (operating in the series resonance mode) through 4-μF blocking capacitors protecting the generator from high voltages (up to ±750V). Measurements were taken in the temperature interval from –60 to +80°C. The sensitivity to relative variation of frequency was 10^{-6} for a measurement time of 10 s. The power of the variable exciting signal that is dissipated by the resonators was no more than 0.1 mW.

It was found that the relative change in the longitudinal vibration frequency in thin langasite rods is a linear function of the applied field. The amount of the effect is sufficiently high: for a voltage applied to the electrodes of up to ±750 V (i.e., in electric fields of up to $\pm 1.5\times 10^{6}$ V/m), it reaches $\pm 120\times 10^{-6}$ for the *XYs*/15° rods (Fig. 5).

The orientation dependence of the effect for the longitudinal vibration mode is strong: when the propagation direction of the longitudinal mode changes by 15°, the amount of the transverse effect increases by four times, from $\pm 20\times 10^{-12}$ V/m to $\pm 80\times 0^{-12}$ V/m.

It was also found that the temperature variation from –60 to +80°C changes the amount of the effect according to the orientation. The temperature dependence of the polarization effect, is weak, depending on **α**, and varies from -2×10^{-5} to 5×10^{-5} °C.

Discussion

The observed electric-field dependence of the optical radiation intensity diffracted at a sound pulse passing through a double path along a crystal is hard to explain as being caused by any other effect. For example, the effect of the electric field on the photoelasticity of lithium tantalate via the piezoelectric and electrooptic effects is expected to affect the amplitude of the first pulse, this was not observed for the given interaction geometry of sound, light, and field [9].

The observed difference of the way how an external electric field affects the light intensity diffracted at a sound pulse at the distance through which the pulse passes in the volume of the crystal is thus in a good agreement with the electric field effect on the rotation of the plane of polarization of a transverse acoustic wave, i.e., with the effect of induced elliptic polarization.

The linear temperature dependence of the polarization effect observed in the langasite crystals indicates that not only the field dependence of the elastic compliance but also the field dependences of the piezoelectric moduli and permittivity are responsible for the effect. It is specific that in the *XYs*/(0–15°) rods, the temperature dependence of the longitudinal vibration frequency is nonlinear and is described by a parabola of the second order with an extremum at 10–30°C [3]. Such a temperature dependence of the frequency control coefficient, which is proportional to the effective nonlinear piezoelectric modulus was observed earlier for the L-vibration in KDP and $NaNH_4SeO_42H_2O$ crystals [2].

The results obtained in this work (the strong field dependence of the elastic properties, the low internal friction, and the presence of crystal orientations with the zero temperature coefficient of frequency (TCF) for longitudinal and thickness modes) suggest that langasite crystals are promising materials for electrically controlled and nonlinear acoustoelectric devices. After study [9], the field dependence of the elastic properties of langasites was investigated in [10, 11]. In particular, the shear mode velocity in the [100] direction of langasites was found to depend on the field to a greater extent than the longitudinal mode velocity and to be a record value for crystals with low sound attenuation [11].

Crystals for field-controlled devices of acoustoelectronics.

Acoustoelectronic devices that are based on the field dependence of the elastic properties are resonators, filters, and delay lines based on volume and surface acoustic waves

(VAWs and SAWs). Before 1986, only two crystals, lithium niobate ($LiNbO_3$) and bismuth silicate ($Bi_{12}SiO_{20}$), had been viewed as promising media for field-controlled acoustoelectric devices. The disadvantage of bismuth silicate is a high sound attenuation and high negative values of the first-order temperature coefficients of elastic moduli. Therefore, there is lack of cross sections with the zero temperature coefficients of propagation velocity and delay time of VAWs and SAWs in $Bi_{12}SiO_{20}$ (the temperature coefficient of velocity is -1.5×10^{-4}°C).

Crystals for nonlinear acoustoelectronic devices.

The efficiency of nonlinear interactions between acoustic waves in acoustoelectric devices depends on the nonlinear properties of a piezoelectric crystal serving as an acoustic line. In accordance with the equation of motion for strain wave $\boldsymbol{S_1}$ propagating in a piezoelectric crystal placed in variable (pumping) electric field [9], parametric interaction between strain wave $\boldsymbol{S_1}$ and pumping electric field takes place. If the frequency of the pumping field equals the double frequency of the strain wave $\boldsymbol{\omega}$, parametric resonance is observed, which generates strain wave $\boldsymbol{S_2}$ at the same frequency $\boldsymbol{\omega}$, which propagates toward wave $\boldsymbol{S_1}$. The backward-to-forward wave power ratio, $\boldsymbol{P_2/P_1}$, is proportional to the parametric interaction efficiency $\boldsymbol{Q^2}$, which depends on a combination of physical parameters of the crystal, $\boldsymbol{Q^2 = g^2/c^2\varepsilon}$ [9], where $\boldsymbol{c}$ is the elastic modulus.

Conclusion

Langasite crystals with control coefficient γ much higher than that of $LiNbO_3$ and comparable to γ of $Bi_{12}SiO_{20}$ have a low sound attenuation and, in addition, possess orientations with the zero temperature coefficients of velocity. Therefore, they are more promising materials for controlled piezoelectric devices.

The $La_3Ga_5SiO_{14}$ crystals offering a high parametric interaction efficiency and a high temperature stability are a promising material for nonlinear acousoelectronic devices, especially at high frequencies

Thus, investigation into the properties of piezoelectric crystals is a topical issue. It seems that the most interesting results will be found in the nonlinear electromechanical properties of isomorphic langasites, which have an ordered structure and therefore offer extremely low acoustic losses, such as $Ca_3NbGa_3Si_2O_{14}$ and $Ca_3Ta\ Ga_3Si_2O_{14}$ crystals [12].

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