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Anisotropy of Attenuation of Acoustic Waves in Lanthanum Gallosilicate Crystals

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Abstract: Anisotropy of the attenuation of acoustic waves in lanthanum gallosilicate crystals have been studied by the Bragg light diffraction method in the frequency range from 0.4 to 1.6 GHz. The results were used to determine the real and imaginary elastic constants for these crystals, which can be used to construct the acoustic attenuation surfaces. It is shown that lanthanum gallosilicate crystals are characterized by relatively high anisotropy of attenuation coefficient of acoustic waves. The observed effect is explained by the strong dependence of the effective Grüneisen constant on the direction of the wave vector.

Keywords: Acoustic wave, Anisotropy of attenuation, Gruneisen constant, Lanthanum gallosilicate crystals, Real and imaginary elastic constants, Bragg diffraction method.

1. Introduction

The main material for creating sensors operating on the piezoelectric effect is thermostable and high-Q quartz, on which resonators are created in a wide frequency range from hundreds of hertz to 100 MHz. However, significant disadvantages of quartz are the relatively low value of the electromechanical coupling coefficient and the phase transition at a temperature of 573 ºC with the loss of piezoelectric properties [1].

Crystals of tantalate and lithium niobate are strong piezoelectrics, characterized by high thermal and chemical stability, but also have a phase transition at temperatures of 620 ºC and 1145 ºC, respectively. In addition, these crystals have a pyroelectric effect, due to which an electrical signal in the crystal arises not only from external pressure, but also from temperature [2, 3].

Compared to these materials used in sensors of physical quantities, single crystals of the langasite family have a number of advantages: thermal stability of piezoelectric characteristics, no pyroelectric effect, and no phase transitions up to a melting temperature of 1470 °C [4-7]. The effectiveness of the use of these crystals is associated with a high coefficient of electromechanical coupling that is 3-4 times higher than the coefficient of electromechanical coupling of quartz.

A well-known representative of this family is lanthanum hallosilicate crystals ($La_3Ga_5SiO_{14}$), which significantly exceeds quartz in piezoelectric coefficients, has thermostable cuts for bulk and surface acoustic waves, and is of great interest for modern acoustics, acoustoelectronics, and piezotechnics [7, 8].

One of the parameters limiting the use of crystals in acoustics and acoustoelectronics is the acoustic attenuation coefficient. The main mechanism of attenuation of acoustic waves in the region $\omega\tau \ll 1$ (ω is the circular frequency of the acoustic wave, τ is the relaxation time of thermal phonons) is the Akhiezer mechanism [9, 10]. This mechanism of acoustic attenuation is assumed that the frequency of acoustic wave is below that of any possible relaxations in the crystal. If the thermal phonon relaxation times are short with the period of acoustic waves, then the complex elastic tensor can be used to describe the loss of acoustic wave's energy [9, 11].

The real components of the complex elastic tensor are known for great number crystals [1, 4]). At the same time the imaginary components, describing attenuation of acoustic waves, were not practically investigated. The most detailed researches of imaginary components of elastic tensor have been made in [11-14].

For a complete description of the anisotropy of the acoustic properties of crystals, along with the characteristic surfaces of the propagation velocity of acoustic waves, it is convenient to use the acoustic attenuation surfaces. This approach was first proposed in the study of the anisotropy of acoustic attenuation in crystals of magnesium oxide [11] and lithium niobate [12] and was used in this work to analyze the anisotropy of attenuation of longitudinal and transverse acoustic waves in lanthanum hallosilicate crystals.

2. Samples and Experimental Method

Samples were prepared in the form of a cube with length of edges 12 mm. The faces of the cube have been oriented with accuracy no worse than 1° along the crystallographic axes [001], [100] and [010] or [001], [110] and [1-10], respectively. To excite longitudinal or transverse acoustic waves, lithium niobate plate transducers (X or rotated Y-cut) were used, which had a thickness of 40 or 70 μm.

The velocity and attenuation of acoustic waves were investigated with the help of Bragg diffraction of light on acoustic waves in the frequency range 0.4–1.2 GHz at room temperature [15]. The measurements were carried out using an acousto-optic setup consisting of a system for exciting acoustic waves and a system for recording diffracted laser light (wavelength 632.8 nm). The helium-neon laser with a wavelength of 632.8 nm was used as a light source. The intensities of diffracted light were measured with the help of photoelectric multiplier.

The acoustic wave velocity V was determined from the Bragg diffraction angle with the accuracy of 0.2 % [1]:

$$V = \lambda\nu / 2\sin\theta_B , \qquad (1)$$

where ν is the linear frequency of acoustic wave, θ_B is the external Bragg angle.

The attenuation coefficient was calculated with the accuracy ~ 5 % by the formula [15]:

$$\alpha = 10\lg\left(I_1/I_2\right) / 2L_0 \qquad (2)$$

Here L_0 is the length of the sample, I_1 and I_2 are respectively the intensity of the light diffracted on the direct acoustic wave and the intensity of the light diffracted on the wave, reflected from the free end of the sample, at the same point along the direction of propagation of acoustic waves. The corresponding distances and time intervals between the indicated signals were also measured.

The Formula (2) is easily derived from consideration of the dependence of the measured values of light intensities on the distance along the direction of propagation of the acoustic wave.

3. Results and Discussion

The experimentally obtained values of velocity and attenuation of longitudinal and transverse acoustic waves along the crystallographic directions [100], [010] and [001] in LGS crystals are shown at Table 1. In this table ***q*** and ***γ*** are the directions of wave vector and polarization vector, respectively.

Table 1. Velocity and acoustic attenuation in LGS crystals (ν= 1 GHz, T = 293 K).

q	γ	V, 10^3 m s^{-1}	α, dB·μs^{-1}
[100]	[1 0 0]	5.73	0.20
	[0 0.83 -0.56]	2.39	0.35
	[0 0.56 0.83]	3.32	0.23
[010]	[0 0.996 -0.092]	5.75	0.21
	[0 0.092 0.996]	3.02	0.31
	[1 0 0]	2.71	0.28
[001]	[0 0 1]	6.75	0.15
	[1 0 0]	3.06	0.31

The measurement results given in the Table 1 make it possible to determine first the effective values of the real and imaginary elastic constants for the studied directions of propagation of acoustic waves, which, in turn, can be represented as a certain combination of the elastic tensor components [11]:

$$c'_{eff} = c'_{ijkl}\kappa_j\kappa_l\gamma_i\gamma_k , \qquad (3)$$

$$c''_{eff} = c''_{ijkl}\kappa_j\kappa_l\gamma_i\gamma_k , \qquad (4)$$

where κ_j and γ_i are the components of the unit wave normal $\boldsymbol{\kappa}$ and the polarization vector $\boldsymbol{\gamma}$, respectively. c'_{ijkl} and c''_{ijkl} are the real and imaginary components of the complex tensor of elastic constants:

$$c_{ijkl} = c'_{ijkl} + ic''_{ijkl} \qquad (5)$$

It should be noted that some of the effective values of the elastic constants are equal to one of the components of the corresponding part of the elastic tensor, which facilitates their determination.

For example, for longitudinal waves propagating along the [100] direction, the following relations are valid:

$$c'_{eff} = c'_{11}, \tag{6}$$

$$c''_{eff} = c''_{11} \tag{7}$$

In the same way, for longitudinal waves propagating along the [001] direction, the following relations are valid:

$$c'_{eff} = c'_{33}, \tag{8}$$

$$c''_{eff} = c''_{33} \tag{9}$$

Symmetry properties and, accordingly, the number of independent components of the imaginary part of the elastic tensor are the same as for the real part, which is used to analyze the orientation dependence of the phase and group velocities of acoustic waves in crystals [1, 6].

For lanthanum gallosilicate crystals (point symmetry group 32), from the experimental data on the velocity and attenuation of acoustic waves, along with seven real elastic coefficients, seven independent components of the imaginary part of the elastic coefficient tensor should be determined (in the matrix notation: c''_{11}, c''_{12} c''_{13}, c''_{14}, c''_{33}, c''_{44}, c''_{66}).

The area of our research satisfies the condition ωτ<<1, where ω is the frequency of acoustic wave, τ is relaxation time of thermal phonons. The acoustic attenuation in this region can be expressed in terms of the real and the imaginary part of the complex elastic constants [11]:

$$\alpha = \frac{1}{2}\omega\frac{c''_{eff}}{c'_{eff}} \tag{10}$$

Expressions (3) and (4) can be written in terms of the real and imaginary components of the Green–Christoffel tensors, which are the convolution of the material tensor of elastic constants respectively real or imaginary, over the direction cosines of the wave vector [11, 14].

In particular, the imaginary effective values of elastic constants are determined by expression

$$c''_{eff} = \Gamma''_{ik}\gamma_i\gamma_k = \frac{2\alpha\rho V^2}{\omega}, \tag{11}$$

where ρ is the density of crystal.

Using Eq. (11), it is possible to determine the attenuation coefficient of acoustic waves in a crystal for any directions of the wave vector. These directions are included in the expressions for the effective elastic constants c'_{eff} (3) and c''_{eff} (4). It can be seen that the general form of these expressions is determined by the symmetry of the crystal and the direction cosines of the unit wave normal and the polarization vector.

The experimental results were used to calculate all the independent real and imaginary components of the complex tensor of elastic constants, with the help of which it is possible to determine the characteristics of acoustic waves along any arbitrary direction in crystal. It should be noticed that the obtained real elastic constants are in good coincidence with data from [5].

As a result we used the next real elastic constants:

c'_{11} =18.8·10^{10} Н/м^2, c'_{33} =26.2·10^{10} Н/м^2,

c'_{44} =5.37·10^{10} Н/м^2, c'_{66} =4.21·10^{10} Н/м^2,

c'_{13} =9.69·10^{10} Н/м^2, c'_{14} =1.41·10^{10} Н/м^2.

The accuracy of determining these real constants of the elasticity tensor was approximately 1 %.

The corresponding imaginary components of the elastic tensor were also calculated:

c''_{11} =2.78·10^6 Н/м^2, c''_{33} =2.90·10^6 Н/м^2,

c''_{44} =1.23·10^6 Н/м^2, c''_{66} =0.95·10^6 Н/м^2,

c''_{13} =0.26·10^6 Н/м^2, c''_{14} =-0.52 10^6 Н/м^2.

The accuracy of determining the imaginary constants of the elasticity tensor was worse due to the relatively large error in determining the attenuation coefficients of acoustic waves and amounted to about 10 %.

We considered the anisotropy of velocity and attenuation of all three modes of acoustic waves during their propagation in the crystallographic plane (100), orthogonal to the second-order symmetry axis. In this case, the direction cosines are κ_1=0, κ_2=cosφ, κ_3=sinφ.

The anisotropy of the propagation velocity of longitudinal and transvers acoustic waves in the (100) plane is shown in Fig. 1. It is seen, that the largest change in velocity by a factor of 1.4 is observed for pure transverse waves in this plane. While for other waves the change in the velocity value does not exceed 20 % for quasi-longitudinal waves and 16 % for quasi-shear waves.

It should be noted that in the investigated plane (100) there are two acoustic axes, one of which coincides with the symmetry axis of the third order [001] and the other is directed at an angle of 42 degrees to the Y axis. Transverse waves propagating along this acoustic axis are pure, since as the polarization vector of one of them is directed perpendicular to the (100) plane, and the polarization vector of the second transverse wave deflected by 90 degrees relative to the direction of the wave vector in the same plane.

The imaginary components of the Green-Christoffel tensor can be are written as:

$$\Gamma''_{11} = c''_{66}\cos^2\varphi + c''_{44}\sin^2\varphi + 2c''_{14}\sin\varphi\cos\varphi, \tag{12}$$

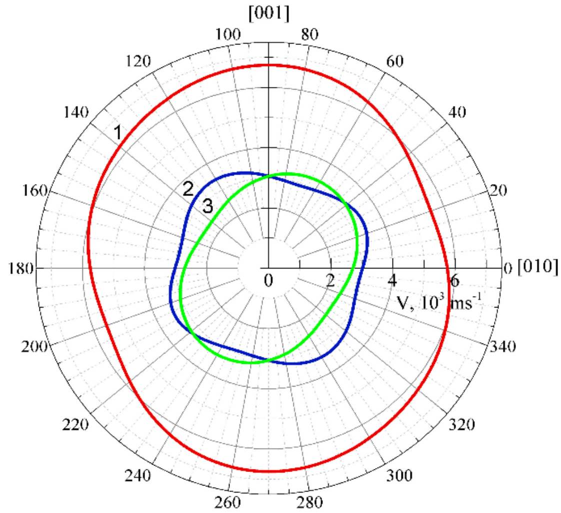

Fig. 1. Cross-section of the surface of the velocity of quasilongitudinal (1), quasi-transverse (2) and transverse acoustic waves (3) in $La_3Ga_5SiO_{14}$ crystals by the plane (100).

$$\Gamma''_{22} = c''_{11}\cos^2\varphi + c''_{44}\sin^2\varphi - 2c''_{14}\sin\varphi\cos\varphi, \quad (13)$$

$$\Gamma''_{33} = c''_{44}\cos^2\varphi + c''_{33}\sin^2\varphi, \quad (14)$$

$$\Gamma''_{23} = \Gamma''_{32} = (c''_{13} + c''_{44})\sin\varphi\cos\varphi - c''_{14}\cos^2\varphi, \quad (15)$$

$$\Gamma''_{12} = \Gamma''_{21} = \Gamma''_{13} = \Gamma''_{31} = 0 \quad (16)$$

As a result, the imaginary part of the effective elastic constant for acoustic waves, the polarization of which lies in the plane of their propagation (100), is determined by the expression [8]:

$$c''_{eff} = \frac{1}{2}\left(\Gamma''_{22} + \Gamma''_{33}\right) \pm \frac{1}{2}\sqrt{(\Gamma''_{22} - \Gamma''_{33})^2 + 4\Gamma''_{23}} \quad (17)$$

Note that in order to determine the real part of the effective elastic constant c'_{eff}, it is necessary to replace the imaginary components with real ones in equation (12). The imaginary part of the effective elastic constant for the pure transverse acoustic wave, the polarization of which is perpendicular to the plane (100), is equal $c''_{eff} = \Gamma''_{11}$ and changes with the direction in this plane according to Eq. (12).

First of all, our studies have shown that the attenuation of high-frequency acoustic waves in langasite is noticeably greater than in quartz, which is possibly due to the disordered structure of these crystals due to the deficiency of Ga + and Si + ions arising directly during their growth. According to [7], the introduction of TiO_2 and Al_2O_3 oxides into lanthanum hallosilicate crystals promotes the filling of the corresponding positions in the lattice with Al^{3+} and Ti^{4+} ions intended for Ga^{3+} and Si^{4+} ions, while the structure of the LGS+Al and LGS+Ti crystals becomes ordered and relaxation losses are reduced.

The anisotropy of the attenuation coefficients of longitudinal and transverse acoustic waves in the form of a cross-section of the attenuation surface by the plane (100) is shown in Fig. 2.

It is seen that lanthanum gallosilicate crystals are characterized by relatively high anisotropy of attenuation coefficient of acoustic waves. Especially it is true for pure transverse acoustic waves in examined plane. For these waves in the (100) plane, the maximum attenuation coefficient differs from the minimum attenuation by almost six times. For quasi-longitudinal and quasi-transverse acoustic waves, the ratio of the maximum attenuation value to the minimum value is approximately two.

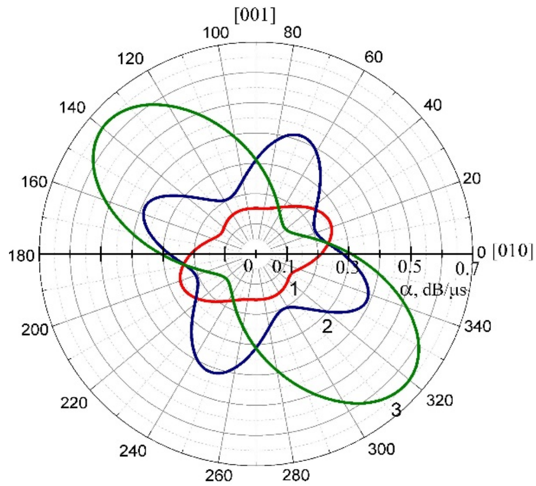

Fig. 2. Cross-section of the attenuation surface of quasilongitudinal (1), quasi-transverse (2) and transverse acoustic waves (3) by the plane ZOY in $La_3Ga_5SiO_{14}$ crystals.

The results of our investigations showed also that, in lanthanum hallosilicate crystals, there is no relationship between the velocity anisotropy and the attenuation anisotropy of acoustic waves. Although, according to Expression (10), the attenuation anisotropy is determined by the anisotropy of both real and imaginary effective elastic constants.

At the same time, there is a clear connection between the anisotropy of attenuation and the anisotropy of the imaginary part of the effective elastic constant. Fig. 3 shows the orientation dependence of three quantities in relative units, including the real effective elastic constant, the imaginary effective elastic constant and the attenuation coefficient for longitudinal acoustic waves in the crystallographic plane (100).

It can be seen that the anisotropy of the acoustic attenuation coefficient almost completely coincides with the orientation dependence of the imaginary effective elastic constant.

The observed regularity is well explained by the fact that the anisotropy of acoustic attenuation is primarily due to the dependence of the Gruneisen anharmonicity constant on the direction of the wave vector of the acoustic wave [9, 10, 14]. This constant enters into the expression determining the value of the attenuation coefficient of acoustic waves by the Akhiezer mechanism [9, 10] and, according to Eq. (11), determines also the orientation dependence of the imaginary part of the effective elastic constant.

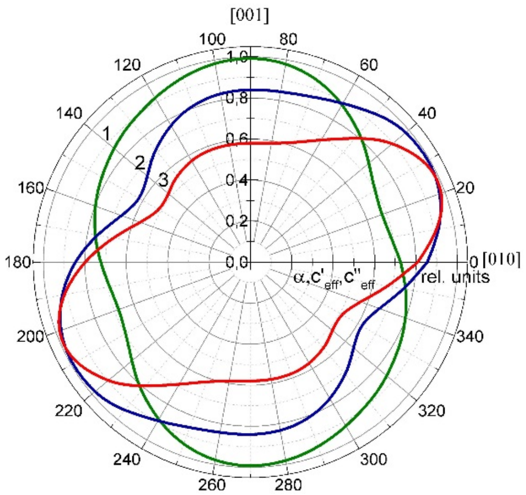

Fig. 3. Orientation dependence of the real effective elastic constant (1), the imaginary effective elastic constant (2), and the attenuation coefficient (3) for longitudinal acoustic waves in the plane (100).

4. Conclusions

Our studies have shown that the cavities of the attenuation coefficient of quasi-transverse and pure transverse waves intersect in two directions, one of which at an angle of about 10 degrees to the [010] axis in the (100) plane does not coincide with the direction of the acoustic axis, known from studies of the transverse wave velocities in lanthanum hallosilicate crystals. And this interesting result requires further detailed study of the propagation of transverse acoustic waves in other planes of symmetry.

The obtained results on the anisotropy of acoustic attenuation in LGS crystals will be useful in the development of acoustic and acoustoelectronic devices, including sensors and piezoelectric resonators based on lanthanum hallosilicate crystals.

References

[1]. E. Dieulesaint, D. Royer, Elastic waves in solids, Application for treatment of signals, *Nauka*, Moscow, 1982.

[2]. Andrushchak A. S., Mytsyk B. G., Laba H. P., *et al.*, Complete sets of elastic constants and photoelastic coefficients of pure and MgO-doped lithium niobate crystals at room temperature, *Journal of Applied Physics*, Vol. 106, 2009, pp. 073510-1 – 073510-6.

[3]. Chezganov D. S., Shur V. Ya., Baturin I. S., Akhmatkhanov A. R., *et al.*, Polarization reversal in crystals of congruent lithium tantalate at elevated temperatures, *Ferroelectrics*, Vol. 439, Issue 1, 2012, pp. 40-46.

[4]. Andreev I. A., Single crystals of the langasite family: An intriguing combination of properties promising for acoustoelectronics, *Technical Physics*, Vol. 51, Issue 6, 2006, pp. 758-764.

[5]. Sotnikov A. V., Kunze R., Schmidt H., Weihnacht M., Hengst M., Gotze J., Piezoelectric and elastic properties of $Sr_3NbGa_3Si_2O_{14}$ (SNGS) single crystals, *Physics of the Solid State*, Vol. 51, Issue 2, 2009, pp. 275-279.

[6]. Kazantsev S. G., Ovcharenko T. N., Calculated and experimental study of the anisotropy of elastic properties of piezoelectric single crystals of the langasite group, *Electromechanics*, Vol. 123, 2011, pp. 51-59.

[7]. I. A. Andreev, Anisotropy and dispersion of the velocity and absorption of elastic waves in piezoelectric crystals, *Experimental Physics*, Issue 6, 2015, pp. 27-43.

[8]. F. R. Akhmedzhanov, T. Sh. Mustafaev, J. T. Nazarov, Attenuation of Acoustic Waves in Langasite Crystals, in *Proceedings of the 3rd IFSA Frequency & Time Conference (IFTC '2021)*, Palma de Mallorca, Mallorca, Spain, 22-24 September 2021, pp. 40-43.

[9]. Yu. A. Logachev, B. Ya. Moyzhes, On the theory of sound absorption according to Akhizer, *Solid State Physics*, Vol. 16, Issue 8, 1974, pp. 2219-2223.

[10]. R. Nava, M. P. Vecchi, J. Romero, B. Fernandez, Akhiezer damping and the thermal conductivity of pure and impure dielectrics, *Phys. Rev. B.*, Vol. 14, Issue 2, 1976, pp. 800-807.

[11]. F. R. Akhmedzhanov, V. V. Lemanov, A. Nasyrov, Surfaces of acoustic attenuation in crystals, *Letters Journal Technical Physics (SU)*, Vol. 6, Issue 10, 1980, pp. 589-592.

[12]. I. L. Bajak, A. McNab, J. Richter, C. D. Wilkinson, Attenuation of acoustic waves in lithium niobate, *J. Acoust. Soc. Am.*, Vol. 69, Issue 3, 1981, pp. 689-695.

[13]. Wenhua Jiang, Wenwu Cao, High-frequency dispersion of ultrasonic velocity and attenuation of single crystal 0.72 $Pb(Mg_{1/3}Nb_{2/3})O_3$ – 0.28 $PbTiO_3$ with engineered domain structures, *Appl. Phys. Lett.*, Vol. 80, 2002, pp. 2466-2468.

[14]. F. R. Akhmedzhanov, S. Z. Mirzaev, U. A. Saidvaliev, Parameters of Elastic Anisotropy in Bismuth Silicate Crystals, *Ferroelectrics*, Vol. 556, Issue 01, 2020, pp. 23-28.

[15]. F. R. Akhmedzhanov, J. O. Kurbanov, A. F. Boltabaev, Attenuation of Acoustic Waves in Single-domain and Polydomain $LiTaO_3$ Crystals, *Sensors & Transducers*, Vol. 246, Issue 7, 2020, pp. 43-47.