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Attenuation of Acoustic Waves in Single-domain and Polydomain $LiTaO_3$ Crystals

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Abstract: The frequency dependence of the attenuation of longitudinal and transverse acoustic waves along the [100] and [001] directions in single-domain and polydomain lithium tantalate crystals at room temperature has been investigated. In the frequency range 0.15–0.4 GHz, the measurements were carried out by the modified Williams-Lamb pulse interference method. At frequencies of 0.4–1.8 GHz, the studies were performed by the Bragg diffraction of light on the acoustic waves. It is shown that the quadratic dependence of the attenuation on frequency is fulfilled in single-domain $LiTaO_3$ samples. Based on the results all the six independent real and imaginary components of the complex elastic tensor for $LiTaO_3$ crystals have been determined. The obtained values of these components were used to determine the velocity and attenuation coefficient of acoustic waves along other directions. It is also shown, that there is an interrelationship between the orientation dependences of the velocity and attenuation of acoustic waves. A strong increase in the attenuation coefficient of acoustic waves and the violation of the quadratic dependence of the attenuation on frequency in polydomain crystals are ascertained. A resonant mechanism of the attenuation of transverse acoustic waves along the [100] axis associated with the domain microstructure of lithium tantalate crystals was discovered.

Keywords: Lithium tantalate crystals, Acoustic wave, Anisotropy of attenuation, Domain structure, Elastic constants.

1. Introduction

Lithium tantalate crystals are isomorphic with respect to lithium niobate crystals; however, they have a natural anisotropy that is almost an order of magnitude lower than that of lithium niobate crystals with the same electro-optical and elastic properties [1-3]. Lithium tantalate crystals, as well as lithium niobate crystals, have unique pyroelectric and piezoelectric properties combined with good mechanical and chemical stability. These crystals well suited for numerous applications including optical and acoustical waveguides, SAW substrates and piezoelectric transducers etc. [4].

Reference data on the elastic properties of lithium tantalate crystals can be found in a number of works [5, 6]. Nevertheless, at present, complete data on the attenuation of high-frequency acoustic waves in these crystals are lacking. In addition, lithium tantalate crystals are of interest for rapidly developing "domain engineering". Various acoustoelectronic devices such as controlled delay lines, filters, acoustic attenuators and deflectors are based on the interaction of acoustic waves with the regular domain structure of ferroelectrics [7-9].

Experimental studies of the propagation of high-frequency acoustic waves in polydomain $LiTaO_3$ crystals are practically absent in the literature. This is explained by the large value of the attenuation of acoustic waves in polydomain crystals, which limits the capabilities of the radio pulse method for measuring the acoustic attenuation [10]. A more universal research method is the method of light diffraction by acoustic waves, which allows experiments to be carried out at higher frequencies and higher values of acoustic attenuation [4].

2. Samples and Experimental Methods

Samples of single-domain and polydomain crystals of $LiTaO_3$ were prepared in the form of a parallelepiped oriented along the crystallographic directions [100], [001], and [110] with an accuracy of one degree. The average length of the samples was 12 mm. To excite longitudinal and transverse acoustic waves, lithium niobate plate transducers (X or rotated Y-cut) were used. The acoustic contact of the piezoelectric transducer with the sample was made through a thin layer of epoxy resin. The measurements were carried out at odd harmonics of the fundamental frequency of the piezoelectric sensor.

The domain structure of the $LiTaO_3$ samples was changed by heating above the Curie temperature (up to about 1000 K) and subsequent slow cooling at a rate of about 30 K/h. The temperature gradient on the sample did not exceed 2 K/cm.

To obtain a single-domain $LiTaO_3$ sample, platinum electrodes were deposited on two polar (opposite) planes of the crystal (001). The sample was placed in an oven and heated to a temperature above the Curie temperature. Then it was slowly cooled to room temperature. An external electric field exceeding the value of the spontaneous polarization of lithium tantalate was applied simultaneously. This procedure made it possible to obtain a $LiTaO_3$ single crystal with an almost perfect crystal structure.

In the frequency range from 400 MHz to 1.8 GHz, the studies were carried out at room temperature by the Bragg diffraction of light on acoustic waves [6]. The light source was a laser with a wavelength λ_0=0.63 mm, and the diffracted light was recorded using a photomultiplier tube. The velocity of acoustic waves V was measured by the angle of Bragg diffraction:

$$V = \lambda_0 \nu / 2\mathrm{Sin}\theta, \quad (1)$$

where ν is the frequency of the acoustic wave, θ is the external Bragg angle. The accuracy of determining the velocity of acoustic waves by this method depends mainly on the accuracy of determining the diffraction angle, and is ~ 0.2 %.

The attenuation coefficient was determined from measurements of the intensities of the diffracted light I_1 and I_2 at different points of the sample (x_1 and x_2) along the direction of sound propagation using the relation:

$$\alpha = \frac{V}{x_2 - x_1} 10 \lg \frac{I_1}{I_2} \quad (2)$$

The accuracy in determining the attenuation coefficient was about 5 %.

In the frequency range 100–400 MHz, the studies were carried out by the modified method of "pulse interference" Williams and Lamb [10], based on the superposition of high-frequency elastic pulses excited by two probing radio pulses. The developed acoustic measuring system, which makes it possible to simultaneously determine the propagation velocity and the attenuation coefficient of acoustic waves, is described in detail in [11, 12]. The advantage of our method is based on the use of an amplitude selector, with the help of which one or another pulse in a series of echo pulses is selected, and the subsequent measurement of its amplitude with a digital voltmeter. The attenuation coefficient of acoustic waves is determined from the measured values of the amplitudes of adjacent pulses A_1 and A_2 by the relationship:

$$\alpha = \frac{20\ell g(A_1/A_2)}{2L}, \quad (3)$$

where L is the length of the sample. The accuracy of determining the attenuation coefficient was ~ 5 %.

In the process of changing of the frequency of the generator, interference zeros or maxima of the signal amplitude are observed, the position of which on the frequency scale depends on the length of the sample and the velocity of the acoustic wave. By registering successive frequency values at which the amplitude of the resulting pulse passes through the minima, the acoustic wave velocity is determined from an enough accurate ratio [10, 12]:

$$V = 2L \cdot \Delta\nu, \quad (4)$$

where $\Delta\nu$ is the difference between two adjacent frequencies of the high-frequency generator, corresponding to antiphase interference. The indicated difference is determined from the readings of the frequency meter with an absolute accuracy of ±10 Hz. The accuracy of determining the velocity of the acoustic wave was limited by the accuracy of measuring the sample length and was ~ 0.01 %.

3. Results and Discussion

3.1. Single-domain Crystals

The measured values of the attenuation of acoustic waves with a frequency of 1 GHz in single-domain and polydomain lithium tantalate crystals at room temperature are given in Table 1. It can be seen that

the attenuation coefficient of acoustic waves in polydomain crystals increases strongly, especially for waves propagating along the axis [001]. Studies have also shown that the domain structure has practically no effect on the propagation velocity of acoustic waves in lithium tantalate crystals.

Table 1. Attenuation of acoustic waves (dB/μs) in single-domain and polydomain LiTaO3 crystals.

Direction of propagation	**q** ‖ [001]		**q** ‖ [100]		
Wave mode	*L*	*S*	*L*	S_F	S_S
Single-domain crystals	0.31	0.90	0.42	0.80	0.17
Polydomain crystals	5.1	8.0	2.7	0.91	0.48

The frequency dependences of the attenuation coefficient for longitudinal waves along the axes [100] and [001] and for transverse waves along the axis [001] are shown in Fig. 1. Straight lines in the figure are drawn assuming a quadratic dependence of the attenuation on frequency. As it is seen, in single-domain lithium tantalate crystals the attenuation of both longitudinal and transverse acoustic waves in the frequency range from 0.2 to 1.8 GHz increases with frequency according to the quadratic law. Such frequency dependence and the weak temperature dependence of attenuation are characteristic for Akhiezer attenuation mechanism in crystals due to the interaction of the acoustic wave with thermal phonons [13, 14].

It should be observed that the results of our measurements are in a good agreement with the results of [5], in which the attenuation of acoustic waves along the [001] axis was studied in the temperature range from 4 to 300 K.

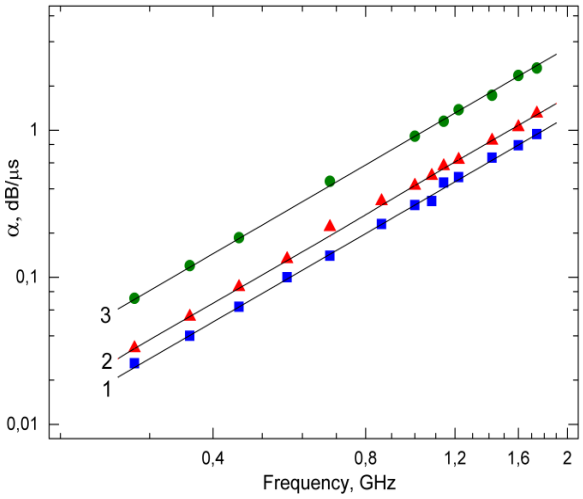


Fig. 1. Frequency dependence of acoustic attenuation in single-domain $LiTaO_3$ crystals for longitudinal waves along the axes [001] (1) and [100] (2) and transverse waves along the axis [001] (3).

The attenuation of acoustic waves in crystals due to their interaction with thermal phonons can be calculated using the real and imaginary components of the complex elastic tensor [15–17]:

$$c_{ijkl} = c'_{ijkl} + ic''_{ijkl}, \tag{5}$$

which enter into the expression of the real and imaginary effective elastic constants:

$$c'_{eff} = c'_{ijkl}\kappa_j\kappa_l\gamma_i\gamma_k \tag{6}$$

and

$$c''_{eff} = c''_{ijkl}\kappa_j\kappa_l\gamma_i\gamma_k \tag{7}$$

Here κ_j and γ_i are the direction cosines of the wave vector and the displacement vector, respectively.

If the thermal phonon relaxation times are short with the period of acoustic waves, then the components of the complex elastic tensor can be used to calculate the loss of acoustic wave's energy. The real components of elastic tensor are known for great number crystals [4, 6]. At the same time the imaginary components, describing attenuation of acoustic waves, were not practically investigated. The detailed studies of imaginary components of elastic tensor for cubic crystals BGO and BSO have been made in [14, 18].

Based on the received results and the value of density ρ [6] all the six independent real c'_{ijkl} and imaginary c''_{ijkl} components of the complex elastic tensor for $LiTaO_3$ crystals have been calculated. The obtained values of these components were used to determine the velocity and attenuation coefficient of acoustic waves by following relations [15, 16]:

$$V = \left(\frac{c'_{eff}}{\rho}\right)^{1/2}, \tag{8}$$

$$\alpha = \frac{\omega}{2}\cdot\frac{c''_{eff}}{c'_{eff}} \tag{9}$$

In these formulas ω is the angular frequency of acoustic wave, ρ is the density of crystal.

By relations (8) and (9) the velocity and attenuation of the acoustic waves along any direction in crystal can be determined. As a result, it is possible to calculate and construct the characteristic surface of attenuation of acoustic waves in a certain crystal, which will clearly describe the anisotropy of the acoustic attenuation in this crystal. In practice, the anisotropy of acoustic attenuation in crystals can be clearly represented by the dependence of the attenuation coefficient of longitudinal and transverse acoustic waves on the direction of propagation in a certain crystallographic plane, for example, in the (001) plane. Such the cross-section of the surface of the attenuation of longitudinal and transverse

acoustic waves in lithium tantalate crystals by the plane (100) is shown in Fig. 2.

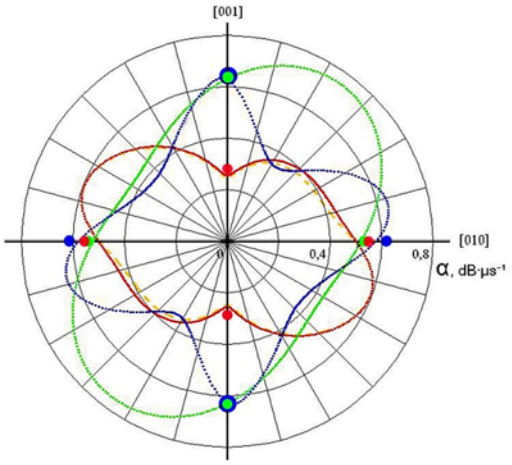


Fig. 2. Cross-section of the attenuation surface of longitudinal (red), transverse (blue) and pure transverse (green) acoustic waves by the plane (100) in $LiTaO_3$ crystals. Dots are experimental values of attenuation.

It should be noted that lithium tantalate crystals are characterized by relatively low anisotropy of attenuation of acoustic waves. This result can apparently be explained by the fact that transverse and longitudinal acoustic waves in lithium tantalate crystals interact mainly with the same branches of the thermal phonon spectrum. The only exception is slow shear waves along the axis [100].

The analysis indicates that the most noticeable change in the acoustic attenuation for all types of waves occurs when the direction of propagation of acoustic waves turns from the axis [111] to the [001]. Our results also show that there is a relationship between the velocity anisotropy and the attenuation anisotropy of acoustic waves, which depends on the anisotropy of the real and imaginary effective elastic constants.

3.2. Polydomain Crystals

The results of measurements of the frequency dependence of the attenuation of longitudinal and slow transverse acoustic waves (S_S wave) along the axis [100] in polydomain lithium tantalate crystals are shown in Fig. 3. Straight lines in this figure are drawn under the assumption of a quadratic (solid lines) and linear (dashed line) dependences of the attenuation on frequency.

It can be seen that, in polydomain crystals, not only a strong increase in the attenuation coefficient of acoustic waves is observed, but also a deviation of the frequency dependence of the attenuation of longitudinal waves along the axis [100] from the quadratic law towards linear at the frequencies above 0.8 GHz, as well as the anomalous behavior of the attenuation of slow transverse waves along this direction.

The influence of the interaction of acoustic waves with domain boundaries on the propagation of acoustic waves in ferroelectrics can be reduced to two main mechanisms. The first of them is due to oscillations of domain walls under the action of an acoustic wave, which can lead to the additional attenuation of acoustic waves [3]. The second mechanism is associated with the reflection, refraction and transformation of an acoustic wave as it passes through the domain boundaries. These effects can lead to a noticeable attenuation of acoustic waves [4].

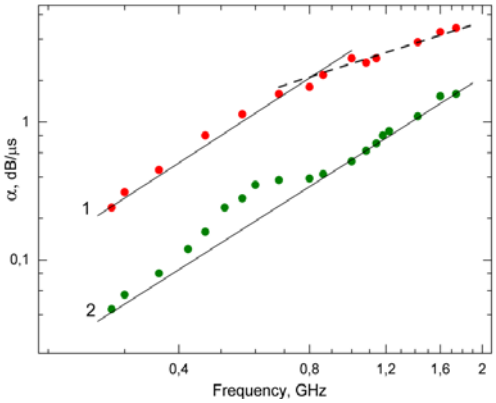


Fig. 3. Frequency dependence of the attenuation of acoustic waves along the axis [100] in polydomain $LiTaO_3$ crystals. 1 - longitudinal, 2 - transverse waves.

The main reason for the increase in the attenuation of acoustic waves can be additional energy losses during their reflection from domain boundaries. However, lithium tantalate crystals are centrosymmetric in the paraphase, and therefore the crystallographic axes in the adjacent 180 degree domains are directed in the same way. This, according to [3], leads to the fact that the reflection of acoustic waves from domain walls, at normal incidence on them, should not occur.

However, the observed increase in the attenuation of acoustic waves indicates that the domain walls separating neighboring domains are not oriented strictly perpendicular to the crystallographic axes [100] and [001]. This conclusion is consistent with the results of [8], in which is detected the motion of the domain walls and the independent growth of domains by their merging due to the difference in the orientation of the walls of adjacent domains.

The anomalous section of the frequency dependence of the attenuation of transverse waves along the axis [100] is shown separately in Fig. 4.

The observed maximum of the resonance-type attenuation may be due to the domain structure of $LiTaO_3$. It can be seen that the maximum attenuation is experienced by waves with a frequency of about 0.68 GHz, which corresponds to a wavelength of ~ 5 μm. The obtained value is in good agreement with the domain widths found in [8, 9] in order of magnitude.

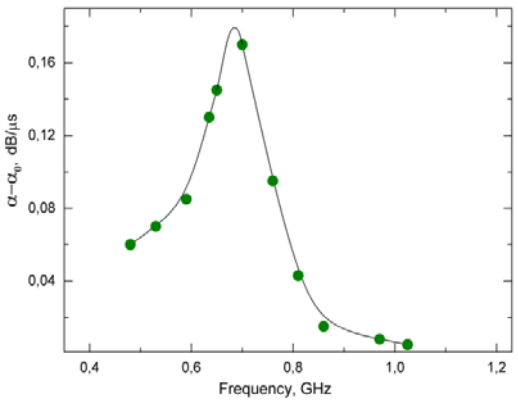


Fig. 4. Frequency dependence of attenuation of slow transverse acoustic waves along the axis [100] in polydomain $LiTaO_3$ crystals.

4. Conclusions

Both real and imaginary independent components of the complex elastic tensor of single-domain lithium tantalate crystals have been determined for the first time. It is shown, that in full accordance with the expressions (8) and (9) there is an interrelationship between the orientation dependences of the velocity and attenuation of acoustic waves. With a decrease in the velocity of acoustic wave, the attenuation coefficient of this wave is increased. In the same way, the opposite dependence is observed.

It is also shown that the quadratic dependence of the attenuation on frequency is fulfilled in single-domain $LiTaO_3$ samples.

A strong increase in the attenuation coefficient of acoustic waves and the violation of the quadratic dependence of the attenuation on frequency in polydomain crystals are ascertained. A resonant mechanism of the attenuation of transverse acoustic waves along the [100] axis associated with the domain microstructure of lithium tantalate crystals was discovered.

To establish a more accurate dependence of the acoustic properties of polydomain lithium tantalate crystals on the domain microstructure, further studies are needed on samples with known sizes and orientations of domains.

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