STUDY OF THE ELASTIC PROPERTIES OF THE LITHIUM TANTALATE CRYSTAL BY THE BRILLOUIN LASER LIGHT SCATTERING

T. BŁACHOWICZ

Institute of Physics Silesian University of Technology (44-100 Gliwice, Krzywoustego 2, Poland) e-mail: tblachow@zeus.polsl.gliwice.pl

This article is dedicated to prof. J. Ranachowski

The preparation of an experiment is described and measured values of elastic constants of the piezoelectric LiTaO₃ crystal, which belongs to the rhomboedral symmetry system, are given. As the experimental method, the Brillouin laser light scattering was applied and the constants from the hypersonic range of frequencies were measured. Appropriate conditions for the experimental configurations were determined by the use of a formalism based on the looking for eigenvalues of a so-called "characteristic matrix" which is a function of direction of the acoustic wave propagation and the elastic constants of the medium. Not all the measurement results are in full agreement with calculations based on ultrasonic data. A dispersion in the velocity of the acoustic waves can be observed for some direction of propagation due to the elastic constant changes in the hypersonic frequency range.

1. Introduction

Brillouin light scattering experiments have been well known for many years as a very useful method for the observation of acoustic phonons in the hypersonic range, both in transparent (bulk phonons) [1–4] and nontransparent media (surface phonons) [5–6]. From the quantum point of view, the creation and annihilation processes of phonons by photons are responsible for the typical Brillouin spectrum in that lines of lowered and increased frequency can be observed.

The present calculations are based on the classical theory of elasticity and classical electrodynamics, and in particular on the Newton's second law and momentum conservation which connects the wave vector of the incident light \vec{k} with the wave vector of the scattered light \vec{k}' and the wave vector of the acoustic wave \vec{q}

$$\vec{q} = \vec{k}' - \vec{k}.\tag{1}$$

The equation of motion of the acoustic wave is given in the following form

$$T_{ij,j} = \rho \ddot{u}_i \,, \tag{2}$$

where T_{ij} is the stress tensor, ρ is the density of the medium, and u_i is the displacement at the given point caused by the acoustic wave. The left side of the above equation, which is the spatial derivative of the stress tensor, is equal to

$$T_{ij,j} = c^E_{ijkl} S_{kl,j} - i\chi_j e_{nij} E_{n,j}$$
(3)

and was derived from the formula

$$T_{ij} = c_{ijkl}^E S_{kl} - e_{nij} E_n \,, \tag{4}$$

where, both in (3) and (4), we can recognize the elasticity tensor c_{ijkl}^E obtained at the condition of a constant electric field, at the strain tensor S_{kl} , the piezoelectric tensor e_{nij} and the electric field E_n induced by the acoustic wave due to the piezoelectric effect. The χ_j are components of a unit-length-vector in the direction of the wave vector. This vector appears in both the displacement given by

$$u_i = u_{0i} \left[e^{i(\vec{\chi} \cdot \vec{r} - \omega t)} + e^{-i(\vec{\chi} \cdot \vec{r} - \omega t)} \right], \tag{5}$$

and in the stress-induced electric field

$$E_n = E_{0n} e^{i(\vec{\chi} \cdot \vec{r} - \omega t)}.$$
(6)

The equality of phases in the displacement and in the electric field formulas means that the electric field is coupled by a component parallel to the direction of the acoustic wave propagation. Taking into account the dependence of the dielectric displacement on strains in a piezoelectric medium

$$D_m = e_{mkl} S_{kl} + \varepsilon_{mn}^S E_n \,, \tag{7}$$

where ε_{mn}^S is the dielectric constant (at constant strain) and taking advantage of the first Maxwell equation $D_{m,m} = 0$, where D_m are the components of the electric induction vector, we have

$$e_{mkl}S_{kl,m} + \varepsilon_{mn}^S E_{n,m} = 0.$$
(8)

In this way, we obtain a formula for the electric field

$$E_{n,j} = -\frac{e_{nkl}S_{kl,m}}{i\chi_m\varepsilon_{mn}^S} = -\frac{e_{mkl}\chi_k\chi_m u_l}{i\chi_m\varepsilon_{mn}^S},$$
(9)

in that we have taken into account Eq. (5) and the following simple formulas

$$S_{kl,j} = u_{k,lj} = -\chi_l \chi_j u_k \tag{10}$$

and

$$S_{kl,j} = u_{l,kj} = -\chi_k \chi_j u_l \tag{11}$$

derived from the definition of the strain tensor

$$S_{kl} = 0.5 \cdot (u_{k,l} + u_{l,k}). \tag{12}$$

By substituting (3), (9) and (11) into the equation of motion (2) the following relation can be obtained

$$-c_{ijkl}^E \chi_j \chi_l u_k - \frac{e_{nij} e_{mkl} \chi_n \chi_m}{\varepsilon_{mn}^S \chi_m \chi_n} \cdot \chi_j \chi_l u_k = -\omega^2 \rho u_k \delta_{ik}$$
(13)

which is equivalent to a set of three independent linear equations, corresponding to three acoustic waves of different polarization. The equations result from the following determinant

$$\left| \left(c_{ijkl}^E + \frac{e_{nij}e_{mkl}\chi_n\chi_m}{\varepsilon_{mn}^S\chi_m\chi_n} \right) \chi_j\chi_l - \omega^2\rho\delta_{ik} \right| = 0.$$
 (14)

The above equation defines the elastic constants modified by the piezoelectric effect and named in the literature as piezoelectrically stiffened elastic stiffness coefficients [7] or effective elastic constants [8]; an eigenproblem for the characteristic matrix defined as $Q_{ik} = c_{ijkl}^{ef} \chi_j \chi_l$ [8, 9, 10] can be recognized. Equation (14) can now be rewritten as follows

$$|Q_{ik} - X\delta_{ik}| = 0. \tag{15}$$

The eigenvectors $\vec{\gamma}$ of the Q_{ik} matrix describe states of polarization of the acoustic waves; a square root of the eigenvalues X, divided by the density of the medium, informs us about the speeds of sound.

The elastic constants can be expressed in a matrix form. Its well known symmetry for the rhomboedral crystal,

$$c_{ij} = \begin{bmatrix} c_{11}^E & c_{12}^E & c_{13}^E & c_{14}^E & 0 & 0 \\ c_{12}^E & c_{11}^E & c_{13}^E & -c_{14}^E & 0 & 0 \\ c_{13}^E & c_{13}^E & c_{33}^E & 0 & 0 & 0 \\ c_{14}^E & -c_{14}^E & 0 & c_{44}^E & 0 & 0 \\ 0 & 0 & 0 & 0 & c_{44}^E & c_{14}^E \\ 0 & 0 & 0 & 0 & c_{14}^E & 0.5 (c_{11}^E - c_{12}^E) \end{bmatrix},$$
(16)

is however changed for the effective elastic constants modified by the piezoelectric effect

$$c_{ij}^{ef} = \begin{bmatrix} c_{11}^{ef} & c_{12}^{ef} & c_{13}^{ef} & c_{14}^{ef} & 0 & 0\\ c_{12}^{ef} & c_{22}^{ef} & c_{23}^{ef} & c_{24}^{ef} & 0 & 0\\ c_{13}^{ef} & c_{23}^{ef} & c_{33}^{ef} & 0 & 0 & 0\\ c_{14}^{ef} & c_{24}^{ef} & 0 & c_{44}^{ef} & 0 & 0\\ 0 & 0 & 0 & 0 & c_{55}^{ef} & c_{56}^{ef}\\ 0 & 0 & 0 & 0 & c_{56}^{ef} & c_{66}^{ef} \end{bmatrix}.$$
(17)

The main purpose of this work was the measurement of the elastic constants of the $LiTaO_3$ piezoelectric crystal by Brillouin laser light scattering. This consists in measuring the changes of the photon frequencies by inelastic scattering on acoustic phonons near the origin of the first Brillouin zone.

The article provides information about four kinds of scattering configurations labeled by A, B, C, D (Fig. 1) in which the angle between the direction of the incident and scattered light was equal to $\pi/2$.



Fig. 1. Graphic definition of the experimental configurations useful for the determination of the elastic constants. Descriptions: \vec{k} – wave vector of the incident light, \vec{k}' – wave vector of the scattered light. The figure provides also components of the wave vector of the acoustic wave (χ_j).

2. The experimental-scattering configurations

The present section provides detailed information about the characteristic matrix for the mentioned experimental configurations and its eigenvalues. The eigenvalues contain information about frequency, polarization and velocity of the acoustic wave and, consequently, information about the investigated elastic constants.

The measurements were done on an arrangement the main elements of which are as follow: a single-mode ion-argon laser working at 514.5 nm with a power of about 100 mW, a scanned Fabry–Perot single-pass pressure interferometer and a device for single photon

counting (PTI-614 analog-digital unit from Photon Inc.) with a Hamamatsu R-4220P photomultiplier. The systematic error of the phonon frequency measurement, induced by the experimental arrangement and the numerical treatment of the data, was equal to 0.15 GHz. The statistical errors depended on the specific measurement and were in the range from 0.04 GHz to 0.27 GHz; in most cases, however they were equal to 0.08 GHz. The total error (standard deviation) for the measured frequency was calculated for the 0.7 level of confidence. All the spectra were achieved in the linear range of pressure changes [11, 12]. This means that the time scale is linearly proportional to frequency.

2.1. The A configuration — determination of the elastic constant c_{11}^E

The A configuration (see Fig. 2a) is suitable for the determination of the c_{11}^E elastic constant. It can be calculated from the Q_{11} element of the characteristic matrix because $Q_{11} = c_{11}^E$. The other values of the characteristic matrix elements are as follows:

$$Q_{22} = \left[0.5 \cdot \left(c_{11}^E - c_{12}^E \right) + \frac{e_{16}e_{16}}{\varepsilon_{11}^S} \right],$$

$$Q_{33} = c_{44}^E + \frac{e_{15}e_{15}}{\varepsilon_{11}^S},$$

$$Q_{23} = c_{14}^E + \frac{e_{15}e_{16}}{\varepsilon_{11}^S},$$

$$Q_{31} = 0, \qquad Q_{12} = 0,$$
(18)

where e_{ij} are the piezoelectric tensor elements written in the double-index formalism. The eigenvalues of the Q_{ij} matrix are equal to

$$X_{1} = c_{11}^{E},$$

$$X_{2/3} = 0.5 \left[\left(0.5(c_{11}^{E} - c_{12}^{E}) + c_{44}^{E} + \frac{e_{16}e_{16} + e_{15}e_{15}}{\varepsilon_{11}^{S}} \right) \\ \pm \left[\left(0.5(c_{11}^{E} - c_{12}^{E}) + c_{44}^{E} + \frac{e_{16}e_{16} + e_{15}e_{15}}{\varepsilon_{11}^{S}} \right)^{2} \right]^{1/2} \right]$$

$$- 4 \left[\left(0.5(c_{11}^{E} - c_{12}^{E}) + \frac{e_{16}e_{16}}{\varepsilon_{11}^{S}} \right) \left(c_{44}^{E} + \frac{e_{15}e_{15}}{\varepsilon_{11}^{S}} \right) - \left(c_{14}^{E} + \frac{e_{15}e_{16}}{\varepsilon_{11}^{S}} \right)^{2} \right]^{1/2} \right] \right].$$
(19)

It is easy to see that the equation $c_{11}^E = X_1$ determines the investigated elastic constant. The acoustic wave frequency observed experimentally, associated with the X_1 eigenvalue, was equal to 33.74 ± 0.16 GHz. The other eigenvalues are smaller. This means that a quasi-longitudinal acoustic wave was responsible for the X_1 value. The X_2 and X_3 values provide information about the quasi-transverse waves of frequency f and the velocity v. The formulas adequate for these parameters are as follows:

$$f = \frac{n}{\lambda} \sqrt{\frac{2X}{\rho}} \tag{20}$$



Fig. 2. Example of the Brillouin spectrum of the LiTaO₃ crystal: a) A configuration — full spectral range FSR equal to 75 GHz. Descriptions: L – longitudinal wave, T1 – quasi-transverse wave, T2 – quasi-transverse wave, b) A configuration — full spectral range FSR equal to 37.5 GHz. Descriptions: T1 – quasi-transverse wave, T2 – quasi-transverse wave, c) B configuration — full spectral range FSR equal to 75 GHz. Descriptions: L – longitudinal wave, T – transverse wave, d) B configuration — full spectral range FSR equal to 37.5 GHz. Description: T – transverse wave.

and

$$v = \sqrt{\frac{X}{\rho}} \,, \tag{21}$$

where *n* is the refractive index of the medium for an ordinary beam, *X* is the eigenvalue of the characteristic matrix and ρ is the density of the medium. In the present paper, numerical results were obtained for the wave vector of the acoustic wave $|\vec{\chi}| = 1$. Figures 2a and 2b show examples of the Brillouin spectra for two different full FSR spectral ranges of the Fabry–Perot interferometer [12]. The spectrum in Fig. 2a shows all three lines arising from one longitudinal and two quasi-transverse acoustic waves. The FSR chosen for the next spectrum (Fig. 2b) enables a detailed observation of the two quasi-transverse frequency waves. The signal from the longitudinal wave is hidden in the strong

peak resulting from elastic scattering (the Rayleigh line). Table 1 gives the values of the calculated eigenvectors, eigenvalues, frequencies and velocities from the elastic constants of SMITH *et al.* measured ultrasonically [13], as well as a comparison with the results of the present measurements [14].

Table 1. Comparison of the acoustic wave velocity and frequency calculated from the values of the elastic constants measured ultrasonically with the velocity and frequency calculated from the hypersonic values of the elastic constants for the A configuration. Eigenvectors $(\vec{\gamma})$ and eigenvalues (X_1, X_2, X_3) calculated from the ultrasonic elastic constants.

Descriptions	$\begin{array}{c} \text{Longitudinal wave} \\ (X_1) \end{array}$	Quasi-transverse wave (X_2)	Quasi-transverse wave (X_3)
Calculated eigenvectors	[1, 0, 0]	[0, -0.6043, 0.7967]	[0, 0.7967, 0.6043]
Calculated eigenvalues (10^{10} Pa)	23.30	12.97	8.30
Calculated velocities (m/s)	5592	4172	3338
Calculated frequencies (GHz)	33.97	25.35	20.28
Measured velocities (m/s)	5554 ± 26	4214 ± 28	3352 ± 25
Measured frequencies (GHz)	33.74 ± 0.16	25.60 ± 0.17	20.36 ± 0.15
Measured eigenvalues (10^{10} Pa)	22.98 ± 0.21	13.23 ± 0.18	8.37 ± 0.13

2.2. The B configuration – determination of the elastic constants c_{33}^E and c_{44}^E

The Q_{33} element of the characteristic matrix provides information about the elastic constant c_{33}^E . The c_{44}^E value can be determined from the Q_{11} and Q_{22} elements which are equal to one another. All the elements of the characteristic matrix and its eigenvalues are written as follows:

$$Q_{11} = c_{44}^{E}, \quad Q_{22} = c_{44}^{E}, \quad Q_{33} = c_{33}^{E} + \frac{e_{33}e_{33}}{\varepsilon_{33}^{S}},$$

$$Q_{23} = 0, \quad Q_{31} = 0, \quad Q_{12} = 0,$$
(22)

$$X_{1} = c_{44}^{E},$$

$$X_{2} = c_{44}^{E},$$

$$X_{3} = c_{33}^{E} + \frac{e_{33}e_{33}}{\varepsilon_{22}^{S}}.$$
(23)

It is obvious that the equation $c_{33}^E = X_3 - e_{33}e_{33}/\varepsilon_{33}^S$ is useful for the determination of the elastic constants c_{33}^E . The values of the piezoelectric e_{ij} constants were taken from Ref. [8] and from Ref. [9, 13] for comparison. There are no information about the experimental errors in these papers. Therefore the results of current calculations were doubled in this case. The experiment acoustic wave frequency, responsible for the X_3 eigenvalue measurement, was equal to 36.43 ± 0.19 GHz. The remaining eigenvalues are smaller. This means that a quasi-longitudinal acoustic wave was responsible for the X_3 value. The X_1 and X_2 provide information about frequencies and velocities of the quasi-transverse waves (23). Their frequencies are equal but possess perpendicular polarizations. The values are T. BŁACHOWICZ

equal to 20.94 ± 0.16 GHz. In this way, the waves are degenerated. Brillouin spectra similar to those of the A configuration can be found in Figs. 2c and 2d. Table 2 contains the values of the calculated eigenvectors, eigenvalues, frequencies and velocities from the elastic constants measured ultrasonically, as well as with the results of measurements for the B configuration for comparison.

Table 2. Comparison of the acoustic wave velocity and frequency calculated from the values of the elastic constants measured ultrasonically with the velocity and frequency calculated from the hypersonic values of the elastic constants for the B configuration. Eigenvectors $(\vec{\gamma})$ and eigenvalues (X_1, X_2, X_3) calculated from the ultrasonic elastic constants.

Descriptions	$\begin{array}{c} \text{Longitudinal wave} \\ (X_3) \end{array}$	Transverse wave (X_1)	Transverse wave (X_2)
Calculated eigenvectors	[0, 0, 1]	[1, 0, 0]	[0, 1, 0]
Calculated eigenvalues (10^{10} Pa)	28.45	9.40	9.40
Calculated velocities (m/s)	6180	3552	3552
Calculated frequencies (GHz)	37.54	21.58	21.58
Measured velocities (m/s)	5997 ± 31	3447 ± 26	3447 ± 26
Measured frequencies (GHz)	36.43 ± 0.19	20.94 ± 0.16	20.94 ± 0.16
Measured eigenvalues (10^{10} Pa)	26.79 ± 0.27	8.85 ± 0.14	8.85 ± 0.14

2.3. The C configuration – determination of the elastic constant c_{66}^E

The C configuration is suitable for the measurement of the elastic constant c_{66}^E . Its value is given by the Q_{11} element of the characteristic matrix. All the elements of the characteristic matrix and its eigenvalues are as follows:

$$Q_{11} = \frac{1}{2} \left(c_{11}^E - c_{12}^E \right), \qquad Q_{22} = c_{11}^E + \frac{e_{22}e_{22}}{\varepsilon_{11}^S}, \qquad Q_{33} = c_{44}^E + \frac{e_{15}e_{15}}{\varepsilon_{11}^S}, Q_{23} = -c_{141}^E + \frac{e_{22}e_{15}}{\varepsilon_{11}^S}, \qquad Q_{31} = 0, \qquad \qquad Q_{12} = 0,$$

$$(24)$$

$$\begin{aligned} X_{1} &= \frac{1}{2} \left(c_{11}^{E} - c_{12}^{E} \right), \\ X_{2} &= \frac{1}{2} \left[c_{11}^{E} + c_{44}^{E} + \frac{e_{22}e_{22} + e_{15}e_{15}}{\varepsilon_{11}^{S}} - \left(\left(c_{11}^{E} + c_{44}^{E} + \frac{e_{22}e_{22} + e_{15}e_{15}}{\varepsilon_{11}^{S}} \right)^{2} \right) \\ &- 4 \cdot \left(\left(c_{11}^{E} + \frac{e_{22}e_{22}}{\varepsilon_{11}^{S}} \right) \left(c_{44}^{S} + \frac{e_{15}e_{15}}{\varepsilon_{11}^{S}} \right) - \left(-c_{14}^{E} + \frac{e_{22}e_{15}}{\varepsilon_{11}^{S}} \right)^{2} \right) \right)^{1/2} \right], \end{aligned}$$
(25)
$$X_{3} &= \frac{1}{2} \left[c_{11}^{E} + c_{44}^{E} + \frac{e_{22}e_{22} + e_{15}e_{15}}{\varepsilon_{11}^{S}} + \left(\left(c_{11}^{E} + c_{44}^{E} + \frac{e_{22}e_{22} + e_{15}e_{15}}{\varepsilon_{11}^{S}} \right)^{2} \right) - 4 \cdot \left(\left(c_{11}^{E} + \frac{e_{22}e_{22}}{\varepsilon_{11}^{S}} \right) \left(c_{44}^{S} + \frac{e_{15}e_{15}}{\varepsilon_{11}^{S}} \right) - \left(-c_{14}^{E} + \frac{e_{22}e_{15}}{\varepsilon_{11}^{S}} \right)^{2} \right) \right)^{1/2} \right]. \end{aligned}$$

The acoustic wave frequency observed in the experiment and responsible for the measurement of the eigenvalue X_1 , was equal to 21.45 ± 0.19 GHz. The values of the calculated eigenvectors, eigenvalues, frequencies and velocities as well as a comparison with hypersonic results of measurements for the C configuration are given in Table 3.

Table 3. Comparison of the acoustic wave velocity and frequency calculated from the values of the elastic constants measured ultrasonically with the velocity and frequency calculated from the hypersonic values of the elastic constants for the C configuration. Eigenvectors $(\vec{\gamma})$ and eigenvalues (X_1, X_2, X_3) calculated from the ultrasonic elastic constants.

Descriptions	Quasi-longitudinal wave (X_3)	Quasi-transverse wave (X_2)	Transverse wave (X_1)
Calculated eigenvectors	[0, -0.9857, -0.1688]	[0, 0.1688, -0.9857]	[1, 0, 0]
Calculated eigenvalues (10^{10} Pa)	24.39	10.88	9.30
Calculated velocities (m/s)	5722	3821	3533
Calculated frequencies (GHz)	34.76	23.21	21.46
Measured velocities (m/s)	_	3946 ± 44	3530 ± 31
Measured frequencies (GHz)	_	23.97 ± 0.31	21.45 ± 0.19
Measured eigenvalues (10^{10} Pa)	_	11.60 ± 0.26	9.28 ± 0.16

2.4. Indirect determination of the elastic constant c_{12}^E from the A and C configurations

The elastic constant c_{12}^E was calculated from the following condition

$$c_{12}^E = c_{11}^E - 2 \cdot c_{66}^E \,, \tag{26}$$

where the c_{12}^E value was taken from the A configuration (19) and that of c_{66}^E from the C configuration (25).

2.5. The D configuration. Indirect determination of the elastic constant c_{14}^E from the C, B and D configurations and indirect determination of the elastic constant c_{13}^E from the A, B and D configurations

All the elements of the characteristic matrix (not equal to zero) and their eigenvalues for the D configuration are as follows:

$$Q_{11} = \frac{1}{2} \left(c_{66}^E - c_{44}^E \right) + c_{14}^E,$$

$$Q_{22} = \frac{1}{2} \left(c_{11}^E + \frac{e_{22}e_{22}}{\varepsilon_{11}^E + \varepsilon_{33}^S} \right) + \frac{1}{2} \left(c_{44}^E + \frac{e_{15}e_{15}}{\varepsilon_{11}^E + \varepsilon_{33}^S} \right) + \frac{1}{2} \left(-c_{14}^E + \frac{e_{22}e_{15}}{\varepsilon_{11}^E + \varepsilon_{33}^S} \right),$$

$$Q_{33} = \frac{1}{2} \left(c_{44}^E + \frac{e_{15}e_{15}}{\varepsilon_{11}^E + \varepsilon_{33}^S} \right) + \frac{1}{2} \left(c_{33}^E + \frac{e_{33}e_{33}}{\varepsilon_{11}^E + \varepsilon_{33}^S} \right),$$

$$Q_{23} = \frac{1}{2} \left(-c_{14}^E + \frac{e_{22}e_{15}}{\varepsilon_{11}^E + \varepsilon_{33}^S} \right) + \frac{1}{2} \left(c_{13}^E + c_{44}^E + \frac{e_{22}e_{33} + e_{15}e_{15}}{\varepsilon_{11}^E + \varepsilon_{33}^S} \right),$$

$$Q_{23} = \frac{1}{2} \left(-c_{14}^E + \frac{e_{22}e_{15}}{\varepsilon_{11}^E + \varepsilon_{33}^S} \right) + \frac{1}{2} \left(c_{13}^E + c_{44}^E + \frac{e_{22}e_{33} + e_{15}e_{15}}{\varepsilon_{11}^E + \varepsilon_{33}^S} \right),$$

$$X_{1} = Q_{11},$$

$$X_{2} = \frac{1}{2} \left[Q_{22} + Q_{33} + \sqrt{Q_{22}^{2} + 4Q_{23}^{2} - 2Q_{22}Q_{33} + Q_{33}^{2}} \right],$$

$$X_{3} = \frac{1}{2} \left[Q_{22} + Q_{33} - \sqrt{Q_{22}^{2} + 4Q_{23}^{2} - 2Q_{22}Q_{33} + Q_{33}^{2}} \right].$$
(28)

The elastic constant c_{14}^E was calculated from the following formula

$$c_{14}^E = X_1 - \frac{1}{2} \cdot (c_{66}^E + c_{44}^E), \qquad (29)$$

where the X_1 value was taken from the D configuration (28) and the remaining values, c_{66}^E and c_{44}^E , were taken from the C (25), and B configurations (23), respectively.

Table 4. Comparision of the acoustic wave velocity and frequency calculated from the values of the elastic constants measured ultrasonically with the velocity and frequency calculated from the hypersonic values of the elastic constants for the D configuration. Eigenvectors $(\vec{\gamma})$ and eigenvalues (X_1, X_2, X_3) calculated from the ultrasonic elastic constants.

Descriptions	Quasi-longitudinal wave (X_2)	Quasi-transverse wave (X_3)	Transverse wave (X_1)
Calculated eigenvectors	[0, -0.9857, -0.1688]	[0, 0.1688, -0.9857]	[1, 0, 0]
Calculated eigenvalues (10^{10} Pa)	24.39	10.88	9.30
Calculated velocities (m/s)	5722	3821	3533
Calculated frequencies (GHz)	34.76	23.21	21.46
Measured velocities (m/s)	-	3946 ± 44	3530 ± 31
Measured frequencies (GHz)	-	23.97 ± 0.31	21.45 ± 0.19
Measured eigenvalues (10^{10} Pa)	-	11.60 ± 0.26	9.28 ± 0.16

Table 5. Summary of the measurements of the elastic constants of the rhomboedral LiTaO₃ crystal. Comparision of the measured (hypersonic) and ultrasonic values of the elastic constants.

Elastic constant	Experimental configuration	$\begin{array}{c} \text{Measured elastic} \\ \text{constant } c^E_{ij} \\ (10^{10}\text{Pa}) \end{array}$	Ultrasonically measured elastic constant c_{ij}^E [13] (10^{10} Pa)	Ultrasonically measured elastic constant c_{ij}^E [8] (10^{10} Pa)
c_{11}^{E}	А	22.98 ± 0.21	22.98	23.3
c_{33}^{E}	В	$\begin{array}{c} 26.48 \pm 0.27^{\rm a} \\ 25.84 \pm 0.26^{\rm b} \end{array}$	27.98	27.5
c^E_{44}	В	8.85 ± 0.14	9.68	9.4
c^{E}_{66}	С	9.28 ± 0.16	9.29	9.3
c_{12}^{E}	A, C	4.42 ± 0.53	4.40	4.7
c_{14}^{E}	D, B, C	0.45 ± 0.29	-1.04	-1.1
c_{13}^E	D, A, B	$\begin{array}{c} 5.36 \pm 0.47^{\rm a} \\ 5.05 \pm 0.44^{\rm b} \end{array}$	8.12	8.0

^a – The piezoelectric constants e_{ij} were taken from Reference [13] and permittivities ε_{ij}^S were taken from Reference [9].

^b – The piezoelectric constants e_{ij} and permittivities ε_{ij}^S were taken from Reference [8].

τ,

The c_{13}^E value is hidden in the X_3 eigenvalue (28), and in the Q_{23} element of the characteristic matrix. To solve this problem, the values of the elastic constants c_{33}^E and c_{44}^E must to be taken from the B configuration and the c_{11}^E is available from the A configuration, so that the c_{13}^E elastic constant is calculated indirectly from 4 values. Therefore their experimental error is relatively large and equal to 8.8%. Table 4 shows the values of the calculated eigenvectors, eigenvalues, frequencies and velocities from the ultrasonic data as well as a comparison with results of the measurements. Table 5 contains values of the measured elastic constants, their experimental errors as well as a comparison with ultrasonic values.

3. Conclusions

A description of the appropriate choice of configurations required for the measurement of the elastic constants of a rhomboedral piezoelectric crystal was given above. As an example, the LiTaO₃ crystal was investigated. The appropriate configuration means that calculated quantities, such as eigenvalues of the characteristic matrix, frequencies and velocities of hypersonic acoustic waves, possess a simple interpretation. This means that the velocities and frequencies depending on the elastic constants in an evident form and not only by pure numerical values. The discussion of a contrary example can be found in Ref. 10.

The general conclusion is that the elastic constants c_{33}^E , c_{44}^E , c_{14}^E , c_{13}^E , for the hypersonic range stayed weaker, if to compare their values with values measured ultrasonically, then the subsequent values of velocities stayed lower. The elastic constants c_{11}^E , c_{13}^E , c_{66}^E are not changed.

It was shown that the formalism based on the determination of the eigenvectors and eigenvalues is very effective and provides a simple physical interpretation. The eigenvectors describe states of polarization of the acoustic wave and the square root of the eigenvalues divided by the density of the medium informs about the speeds of sound. However the presented calculations, based on the classical theory of elasticity and a comparison with the Brillouin scattering measurements can not describe the divergences obtained. More theoretical investigation is required to explain these facts in details.

We hope that the considerations and data given here are detailed enough to provide an adequate description of the nature of the phenomenon.

References

- G. LI, X.K. CHEN, N.J. TAO, H.Z. CUMMINS, R.M. PICK and G. HAURET, Brillouin scattering studies of the transverse acoustic modes of incommensurate K₂SeO₄, Phys. Rev., B44, 13, 6621– 6629 (1991).
- [2] L.E. MCNEIL and M. GRIMSDITCH, *Elastic constants of* As₂S₃, Phys. Rev., B44, 9, 4174–4177 (1991).
- [3] J.J. VANDERWAL, P. ZHAO and D. WALTON, Brillouin scattering from the icosahedral quasicrystal Al_{63.5}Cu_{24.5}Fe₁₂, Phys. Rev., B46, 1, 501–502 (1992).

- [4] U. SCHÄRER and P. WACHTER, Negative elastic constants in intermediate valent Sm_xLa_{1-x}S, Solid St. Comm., 96, 497–501 (1995).
- [5] R. DANNER, R.P. HUEBENER, C.L. CHUN, M. GRIMSDITCH and I.K. SCHULLER, Surface acoustic waves in Ni/V superlattices, Phys. Rev., B33, 6, 3696–3701 (1986).
- [6] M. HUES, R. BHADRA, M. GRIMSDITCH, E. FULLERTON and I.K. SCHULLER, Effect of high-energy ion irradiation on the elastic moduli of Ag/Co superlattices, Phys. Rev., B39, 17, 12966–12968 (1989).
- [7] J. XU and R. STROUD, Acousto-optic devices: principles, design and applications, John Wiley & Sons Inc., 1992.
- [8] A.W. WARNER, M. ONOE and G.A. COQUIN, Determination of elastic and piezoelectric constants for crystals in class (3m), J. Acoust. Soc. Am., 42, 6, 1223–1231 (1966).
- [9] W. SOLUCH, Introduction to piezoelectronics, Wydawnictwa Komunikacji i Łączności, Warszawa 1980.
- [10] T. BLACHOWICZ and Z. KLESZCZEWSKI, Observation of hypersonic acoustic waves in a LiTaO₃ crystal, Archives of Acoustics, 22, 3, 351–357 (1997).
- T. BLACHOWICZ, Wykorzystanie światła laserowego do badania sprężstych własności ciał stałych w obszarze hiperdźwiękowym, Wydawnictwo Politechniki Śląskiej, Gliwice 1999.
- [12] T. BLACHOWICZ, R. BUKOWSKI and Z. KLESZCZEWSKI, Fabry-Perot interferometer in Brillouin scattering experiments, Rev. Sci. Instrum., 67, 12, 4057–4060 (1996).
- [13] R.T. SMITH and F.S. WELSH, Temperature dependence of the elastic, piezoelectric and dielectric constants of lithum tantalate and lithium niobate, J. App. Phys., 42, 6, 2219–2230 (1971).
- [14] T. BLACHOWICZ and Z. KLESZCZEWSKI, Elastic constants of the lithium tantalate crystal in the hypersonic range, J. Acoust. Soc. Am., 104, 6, 3356–3357 (1998).