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 LiTaO$_3$ 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 $\vec{q}$

$$\vec{q} = \vec{k}' - \vec{k}. \quad (1)$$

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

$$T_{ij,j} = \rho \ddot{u}_i, \quad (2)$$
where $T_{ij}$ is the stress tensor, $\rho$ 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

$$T_{ij,j} = c^E_{ijkl}S_{kl,j} - ie_{nij}E_n,$$

and was derived from the formula

$$T_{ij} = c^E_{ijkl}S_{kl} - e_{nij}E_n,$$

where, both in (3) and (4), we can recognize the elasticity tensor $c^E_{ijkl}$ 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 $\chi_j$ are components of a unit-length-vector in the direction of the wave vector. This vector appears in both the displacement given by

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

and in the stress-induced electric field

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

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

$$D_m = e_{mkl}S_{kl} + \varepsilon_{mn}E_n,$$

where $\varepsilon_{mn}$ 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

$$e_{mkl}S_{kl,m} + \varepsilon_{mn}E_{n,m} = 0.$$

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

$$E_{n,j} = -\frac{e_{nkl}S_{kl,m}}{i\chi_m\varepsilon_{mn}} = -\frac{e_{mkl}\chi_k\chi_mu_l}{i\chi_m\varepsilon_{mn}},$$

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

$$S_{kl,j} = u_{k,lj} = -\chi_l\chi_ju_k$$

and

$$S_{kl,j} = u_{l,kj} = -\chi_k\chi_ju_l$$

derived from the definition of the strain tensor

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

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

$$-c^E_{ijkl}\chi_lu_k - \frac{e_{nij}e_{mkl}\chi_k\chi_m}{\varepsilon_{mn}\chi_m\chi_n} \cdot \chi_j\chi_lu_k = -\omega^2\rho u_k \delta_{ik}$$
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

\[
\left| \begin{array}{c}
\left( c_{ijkl}^E + \frac{e_{mkl}^n e_{nij}}{\varepsilon_{mn}^S \chi_{mn}} \right) \chi_j \chi_l - \omega^2 \rho \delta_{ik} \\
\end{array} \right| = 0.
\] (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 \( Q_{ik} = c_{ijkl}^E \chi_j \chi_l \) \[8, 9, 10\] can be recognized. Equation (14) can now be rewritten as follows

\[
|Q_{ik} - X \delta_{ik}| = 0.
\] (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,

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

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

\[
c_{ij}^E = \begin{bmatrix}
   c_{11}^E & c_{12}^E & c_{13}^E & c_{14}^E & 0 & 0 \\
   c_{12}^E & c_{22}^E & c_{23}^E & c_{24}^E & 0 & 0 \\
   c_{13}^E & c_{23}^E & c_{33}^E & 0 & 0 & 0 \\
   c_{14}^E & c_{24}^E & 0 & c_{44}^E & 0 & 0 \\
   0 & 0 & 0 & c_{55}^E & c_{56}^E & c_{56}^E \\
   0 & 0 & 0 & 0 & c_{56}^E & c_{66}^E
\end{bmatrix}.
\] (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 \( \pi/2 \).
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:

\[
\begin{align*}
Q_{22} & = 0.5 \cdot (c_{11}^E - c_{12}^E) + \frac{e_{16}e_{16}}{\varepsilon_{11}^S}, \\
Q_{33} & = c_{44}^E + \frac{e_{15}e_{15}}{\varepsilon_{11}^S}, \\
Q_{23} & = c_{14}^E + \frac{e_{15}e_{16}}{\varepsilon_{11}^S}, \quad (18) \\
Q_{31} & = 0, \quad Q_{12} = 0,
\end{align*}
\]

where $e_{ij}$ are the piezoelectric tensor elements written in the double-index formalism.

The eigenvalues of the $Q_{ij}$ matrix are equal to

\[
X_1 = c_{11}^E, \\
X_{2/3} = 0.5 \left[ 0.5(c_{11}^E - c_{12}^E) + c_{44}^E + \frac{e_{16}e_{16} + e_{15}e_{15}}{\varepsilon_{11}^S} \right]
\pm \left[ 0.5(c_{11}^E - c_{12}^E) + c_{44}^E + \frac{e_{16}e_{16} + e_{15}e_{15}}{\varepsilon_{11}^S} \right]^2 (19)
\]

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

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:

\[
f = \frac{n}{\lambda} \sqrt{\frac{2X}{\rho}} \quad (20)
\]
Fig. 2. Example of the Brillouin spectrum of the LiTaO$_3$ crystal: a) A configuration — full spectral range FSR equal to 75 GHz. Descriptions: $L$ – longitudinal wave, $T_1$ – quasi-transverse wave, $T_2$ – quasi-transverse wave, b) A configuration — full spectral range FSR equal to 37.5 GHz. Descriptions: $T_1$ – quasi-transverse wave, $T_2$ – 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.

\[ v = \sqrt{\frac{X}{\rho}}, \]  

where $n$ is the refractive index of the medium for an ordinary beam, $X$ is the eigenvalue of the characteristic matrix and $\rho$ 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.

<table>
<thead>
<tr>
<th>Descriptions</th>
<th>Longitudinal wave (X_1)</th>
<th>Quasi-transverse wave (X_2)</th>
<th>Quasi-transverse wave (X_3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Calculated eigenvectors</td>
<td>[1, 0, 0]</td>
<td>[0, −0.6043, 0.7967]</td>
<td>[0, 0.7967, 0.6043]</td>
</tr>
<tr>
<td>Calculated eigenvalues (10^{10} Pa)</td>
<td>23.30</td>
<td>12.97</td>
<td>8.30</td>
</tr>
<tr>
<td>Calculated velocities (m/s)</td>
<td>5592</td>
<td>4172</td>
<td>3338</td>
</tr>
<tr>
<td>Calculated frequencies (GHz)</td>
<td>33.97</td>
<td>25.35</td>
<td>20.28</td>
</tr>
<tr>
<td>Measured velocities (m/s)</td>
<td>5554 ± 26</td>
<td>4214 ± 28</td>
<td>3352 ± 25</td>
</tr>
<tr>
<td>Measured frequencies (GHz)</td>
<td>33.74 ± 0.16</td>
<td>25.60 ± 0.17</td>
<td>20.36 ± 0.15</td>
</tr>
<tr>
<td>Measured eigenvalues (10^{10} Pa)</td>
<td>22.98 ± 0.21</td>
<td>13.23 ± 0.18</td>
<td>8.37 ± 0.13</td>
</tr>
</tbody>
</table>

2.2. The B configuration – determination of the elastic constants \(c_{E33}^F\) and \(c_{E44}^F\)

The \(Q_{33}\) element of the characteristic matrix provides information about the elastic constant \(c_{E33}^F\). The \(c_{E44}^F\) 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:

\[
Q_{11} = c_{E44}^F, \quad Q_{22} = c_{E44}^F, \quad Q_{33} = c_{E33}^F + \frac{e_{33}^3 c_{33}^3}{\varepsilon_{33}^3},
\]

\[
Q_{23} = 0, \quad Q_{31} = 0, \quad Q_{12} = 0,
\]

\[
X_1 = c_{E44}^F, \quad X_2 = c_{E44}^F, \quad X_3 = c_{E33}^F + \frac{e_{33}^3 c_{33}^3}{\varepsilon_{33}^3}.
\]

It is obvious that the equation \(c_{E33}^F = X_3 - e_{33}^3 / \varepsilon_{33}^3\) is useful for the determination of the elastic constants \(c_{E33}^F\). 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
equal to $20.94 \pm 0.16$ 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.

<table>
<thead>
<tr>
<th>Descriptions</th>
<th>Longitudinal wave $(X_3)$</th>
<th>Transverse wave $(X_1)$</th>
<th>Transverse wave $(X_2)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Calculated eigenvectors</td>
<td>$[0, 0, 1]$</td>
<td>$[1, 0, 0]$</td>
<td>$[0, 1, 0]$</td>
</tr>
<tr>
<td>Calculated eigenvalues (10$^{10}$ Pa)</td>
<td>6180</td>
<td>3552</td>
<td>3552</td>
</tr>
<tr>
<td>Calculated velocities (m/s)</td>
<td>37.54</td>
<td>21.58</td>
<td>21.58</td>
</tr>
<tr>
<td>Calculated frequencies (GHz)</td>
<td>28.45</td>
<td>9.40</td>
<td>9.40</td>
</tr>
<tr>
<td>Measured velocities (m/s)</td>
<td>5997 ± 31</td>
<td>3447 ± 26</td>
<td>3447 ± 26</td>
</tr>
<tr>
<td>Measured frequencies (GHz)</td>
<td>36.43 ± 0.19</td>
<td>20.94 ± 0.16</td>
<td>20.94 ± 0.16</td>
</tr>
<tr>
<td>Measured eigenvalues (10$^{10}$ Pa)</td>
<td>26.79 ± 0.27</td>
<td>8.85 ± 0.14</td>
<td>8.85 ± 0.14</td>
</tr>
</tbody>
</table>

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:

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

$$X_1 = \frac{1}{2} (c_{11}^E - c_{12}^E), \quad X_2 = \frac{1}{2} \left[ c_{11}^E + c_{44}^E + \frac{e_{22}^E c_{22}^E + e_{15}^E c_{15}^E}{\varepsilon_{11}^S} \right] - \left( \frac{c_{11}^E + c_{44}^E + \frac{e_{22}^E c_{22}^E + e_{15}^E c_{15}^E}{\varepsilon_{11}^S}}{\varepsilon_{11}^S} \right)^2 \left[ -c_{14}^E + \frac{e_{22}^E c_{22}^E + e_{15}^E c_{15}^E}{\varepsilon_{11}^S} \right]^{1/2},$$  

$$X_3 = \frac{1}{2} \left[ c_{11}^E + c_{44}^E + \frac{e_{22}^E c_{22}^E + e_{15}^E c_{15}^E}{\varepsilon_{11}^S} \right] + \left( \frac{c_{11}^E + c_{44}^E + \frac{e_{22}^E c_{22}^E + e_{15}^E c_{15}^E}{\varepsilon_{11}^S}}{\varepsilon_{11}^S} \right)^2 \left[ -c_{14}^E + \frac{e_{22}^E c_{22}^E + e_{15}^E c_{15}^E}{\varepsilon_{11}^S} \right]^{1/2}.\)
The acoustic wave frequency observed in the experiment and responsible for the measurement of the eigenvalue $X_1$, was equal to $21.45 \pm 0.19$ 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.

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

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

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

(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 $c_{13}^E$ 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:

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

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

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

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

(27)
\( X_1 = Q_{11}, \)
\( X_2 = \frac{1}{2} \left[ Q_{22} + Q_{33} + \sqrt{Q_{22}^2 + 4Q_{23}^2 - 2Q_{22}Q_{33} + Q_{33}^2} \right], \)
\( X_3 = \frac{1}{2} \left[ Q_{22} + Q_{33} - \sqrt{Q_{22}^2 + 4Q_{23}^2 - 2Q_{22}Q_{33} + Q_{33}^2} \right]. \)

(28)

The elastic constant \( c_{14}^E \) was calculated from the following formula
\[ c_{14}^E = X_1 - \frac{1}{2} \left( c_{66}^E + c_{44}^E \right), \]
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.** 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 D configuration. Eigenvectors \( \vec{\gamma} \) and eigenvalues \( (X_1, X_2, X_3) \) calculated from the ultrasonic elastic constants.

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

**Table 5.** Summary of the measurements of the elastic constants of the rhomboedral LiTaO\(_3\) crystal.

Comparison of the measured (hypersonic) and ultrasonic values of the elastic constants.

<table>
<thead>
<tr>
<th>Elastic constant</th>
<th>Experimental configuration</th>
<th>Measured elastic constant ( c_{ij}^E ) ( (10^{10} \text{ Pa}) )</th>
<th>Ultrasonically measured elastic constant ( c_{ij}^E ) ( (10^{10} \text{ Pa}) ) [13]</th>
<th>Ultrasonically measured elastic constant ( c_{ij}^E ) ( (10^{10} \text{ Pa}) ) [8]</th>
</tr>
</thead>
<tbody>
<tr>
<td>( c_{11}^E )</td>
<td>A</td>
<td>22.98 ± 0.21</td>
<td>22.98</td>
<td>23.3</td>
</tr>
<tr>
<td>( c_{33}^E )</td>
<td>B</td>
<td>26.48 ± 0.27(^a)</td>
<td>27.98</td>
<td>27.5</td>
</tr>
<tr>
<td>( c_{13}^E )</td>
<td>B</td>
<td>8.85 ± 0.14</td>
<td>9.68</td>
<td>9.4</td>
</tr>
<tr>
<td>( c_{66}^E )</td>
<td>C</td>
<td>9.28 ± 0.16</td>
<td>9.29</td>
<td>9.3</td>
</tr>
<tr>
<td>( c_{12}^E )</td>
<td>A, C</td>
<td>4.42 ± 0.53</td>
<td>4.40</td>
<td>4.7</td>
</tr>
<tr>
<td>( c_{14}^E )</td>
<td>D, B, C</td>
<td>0.45 ± 0.29</td>
<td>−1.04</td>
<td>−1.1</td>
</tr>
<tr>
<td>( c_{13}^E )</td>
<td>D, A, B</td>
<td>5.36 ± 0.47(^a)</td>
<td>5.05 ± 0.44(^b)</td>
<td>8.12</td>
</tr>
</tbody>
</table>

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

\(^b\) – The piezoelectric constants \( e_{ij} \) and permittivities \( \varepsilon_{ij}^S \) were taken from Reference [8].
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 LiTaO$_3$ 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


