Elastic quasi-isotropy and acousto-optics of γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals

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Abstract. We have studied experimentally acoustic-wave velocities and acousto-optic figure of merit (AOFM) for γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals. All the components of elastic-stiffness and elastic-compliance tensors have been determined. From the viewpoint of elasticity, the crystals behave as almost isotropic solid-state medium. Namely, the components of elastic stiffnesses and compliances satisfy the approximate relations $C_{11} \approx C_{33}$, $S_{11} \approx S_{33}$, $C_{44} \approx C_{66}$, $S_{44} \approx S_{66}$, $C_{12} = (C_{11} - C_{12})/2$ and $S_{66} = 2(S_{11} - S_{12})$. The obliquity angles and the angles of polarization non-orthogonality are rather small for all the acoustic modes. The maximal AOFM determined at the isotropic diffraction is equal to $M_2 = (38.6 \pm 0.7)\times 10^{-15}\,\text{s}^3/\text{kg}$. Since the acoustic-wave slownesses and the elasto-optic coefficients p_{ij} determined by us are not high (e.g., $|p_{11}| = 0.105 \pm 0.014$, $|p_{13}| = 0.072 \pm 0.010$ and $|p_{12}| = 0.09 \pm 0.03$), the main contribution into the AOFM arises from high refractive indices, which is typical for the spectral region close to optical absorption edge.

Keywords: γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals, acoustic-wave velocities, elastic stiffness, elastic compliance, elasto-optic coefficients, anisotropy.

UDC: 535.4, 534.2

1. Introduction

γ_1-$(Ga_xIn_{1-x})_2Se_3$ crystals are a wide-band semiconducting solid solution, which crystallizes in a hexagonal, wurtzite-like γ_1-phase of Ga_2Se_3–In_2Se_3 system. It is described by the point symmetry group 6 (the space groups $P6_1$ or $P6_5$) at 0.02 < x < 0.55 [1]. The optical pseudogap E_g^* increases with increasing Ga concentration from 2.160 eV for x = 0.1 to 2.344 eV for x = 0.4 [2]. The optical absorption edge of γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ at low absorption levels has been shown to be formed by indirect interband optical transitions [3]. With the temperature increase from 77 K to 300 K, the absorption edge is shifted towards longer wavelengths, and the linear dichroism manifests itself as a greater absorption of the extraordinary optical wave with respect to the ordinary one [3,4].

The basic optical properties of the γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals have been thoroughly studied. In particular, the dispersion of refractive indices has been scrutinized in Ref. [4]. We have $n_o = 2.90$ and $n_e = 2.95$ at the light wavelength $\lambda = 632.8$ nm. Exciton and impurity-related photoluminescence bands in the γ_1-$(Ga_xIn_{1-x})_2Se_3$ crystals have been revealed at low temperatures. Moreover, a number of temperature and compositional transformations have been studied [2].

Probably, the most prominent property of γ_1-$(Ga_xIn_{1-x})_2Se_3$ (x = 0.1, 0.2, 0.3 and 0.4) is its natural optical activity. In other words, these crystals have relatively high optical rotation. The rotatory power of γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ at λ = 632.8 nm is equal to 103 deg/mm, which is more than five times higher than the effect known for canonical quartz crystals [5].

It has been shown in Ref. [6] that the γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals represent a highly efficient acousto-optic (AO) material. In particular, their AO figure of merit reaches $445\times10^{-15}s^3/kg$ at the diffraction by longitudinal acoustic wave (AW). However, these results need further verification because of the absence of any information on the elasto-optic (EO) coefficients and unusually close values of the longitudinal (~ 1600 m/s) and transverse (~ 1200 m/s) AW velocities. Notice also that the piezo-optic effect in these crystals has been studied only partly [7] and no individual components of the piezo-optic tensor have been determined.

The present work begins comprehensive studies of the AO properties of γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals. Below we will report on their acoustic and EO properties.

2. Experimental methods

A synthesis of a γ_1-$(In_{0.7}Ga_{0.3})_2Se_3$ solid solution was performed from simple substances such as indium (99.999%), gallium (99.999%) and selenium (99.9999%), which were taken according to stoichiometric ratios. Quartz ampoules evacuated down to 0.13 Pa were used. A regime adopted for the synthesis of γ_1-$(In_{0.7}Ga_{0.3})_2Se_3$ included a step heating up to 873 K at the rate 100 K/h (including annealing during 24 h), a further temperature increase up to 1340 K (~ 50 K above the melting point of Ga_2Se_3) at the rate 50 K/h, and annealing at this temperature for 24 h. A further cooling was carried out in an "off" regime of a furnace.

To establish the optimal temperature regime for growing single crystals of the γ_1-$(In_{0.7}Ga_{0.3})_2Se_3$ solid solution, we studied a phase diagram, using a differential thermal analysis (a Pt/PtRh thermocouple and a heating/cooling rate 700 K/h). As a consequence, we established a presence of two endothermic effects at the temperatures 1114±2 K and 1126±2 K, which corresponded to solidus and liquidus. It is noteworthy that, in the cooling mode, there was a single exothermic effect only, with super-cooling (1013±2 K). It corresponds to the process of crystallization of γ_1-$(In_{0.7}Ga_{0.3})_2Se_3$.

Taking into account the nature of melting and crystallization, we grew the single crystals of γ_1-$(In_{0.7}Ga_{0.3})_2Se_3$ solid solution, using a melt solution and a method of vertical floating-zone melting. The process was carried out in a two-zone tubular resistance furnace with the melt-zone temperature 1163 K and the annealing zone 833 K. A quartz container of special configuration was employed for this aim. In order to homogenize the melt, the ampoule was kept in the melt zone for 24 h. The growth of single crystals involved a formation of nucleus in a lower conical part of the container with a method of collective recrystallization (24 h) and a crystal growth performed at a seed formed in this way. The optimal velocity of crystallization front was equal to 0.4–0.5 mm/h, the annealing temperature 833 K (120 h), and the rate of cooling down to the room temperature amounted to 5 K/h. According to this method, dark-red single γ_1-$(In_{0.7}Ga_{0.3})_2Se_3$ crystals were obtained, with the length 30–40 mm and the diameter 20 mm (see Fig. 1).

The stoichiometry was verified with the aid of electron scanning microscope REMMA-102-02. It was found that the composition of our crystals was $(In_{0.75}Ga_{0.29})_2Se_{2.93}$, with the accuracy not less than 5%. The orientation of crystals with respect to the principle crystallographic directions was carried out using a standard X-ray technique. The setting of right-handed coordinate system *XYZ* was as follows: $Z \parallel [0001]$, $X \parallel [2\bar{1}\bar{1}0]$ and $Y \parallel [01\bar{1}0]$. The positive direction of the *Z* axis was chosen basing on piezoelectric response verified experimentally under the condition of positive value of piezoelectric coefficient ($d_{33} > 0$) [8]. Three samples in the shape of parallelepipeds were cut from a boule. Their faces were (1) perpendicular to the *X*, *Y* and *Z* axes, (2) perpendicular to the *X* axis and the diagonal of the *Y* and *Z* axes, and (3) perpendicular to the *Y* axis and the diagonal of the *X* and *Z* axes. The average dimensions of the samples were equal to 5.0×5.0×5.0 mm^3.

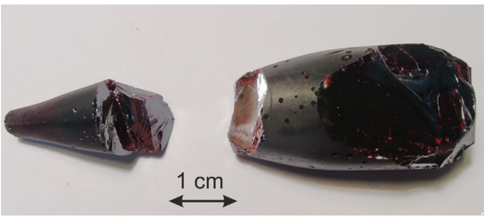

Fig. 1. A boule of as-grown γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals.

The AW velocities were studied by a pulse-echo overlap method at the temperature $T = 293$ K [9]. The AWs were excited with piezoelectric transducers made of $LiNbO_3$ plates of different crystallographic orientations (with the resonance frequency $f = 8$–10 MHz, the bandwidth $\Delta f = 0.1\,\text{MHz}$ and the acoustic power $P_a = 1$–2 W). Zero-induction elastic stiffness coefficients were calculated from a Christoffel equation.

Since our crystals belong to the hexagonal symmetry group, their elastic-stiffness tensor includes five nonzero independent elastic components. The AW velocities v_{ij} that include the contributions of piezoelectric effect (with the index *i* representing the direction of AW propagation and *j* the direction of its polarization) and the elastic-stiffness coefficients are coupled by the relations $C_{11} = C_{22} = \rho v_{11}^2 = \rho v_{22}^2$, $C_{44} = C_{55} = \rho v_{32}^2 = \rho v_{31}^2$, $C_{66} = \rho v_{12}^2$, $C_{12} = C_{11} - 2C_{66}$, $C_{13} = \frac{1}{2}\sqrt{(2\rho v_{4\bar{4}}^2 - 2\rho v_{44}^2)^2 - (C_{33} - C_{11})^2} - C_{44}$, $C_{66} = \rho v_{12}^2$ and $C_{66} = (C_{11} - C_{12})/2$, where ρ ($\rho = 5440\ \text{kg}/\text{m}^3$ [6]) is the crystal density.

The elastic compliances S_{jk} were determined from the matrix C_{ij} of elastic constants, using a well-known relation $C_{ij}S_{jk} = \delta_{ik}$, with δ_{ik} being the Kronecker delta.

The obliquity angle for the AWs can be calculated according to the relation (see Ref. [10])

$$\phi_{X,Y} - \Delta_{X,Y} = \arctan \frac{1}{v(\phi_{X,Y})} \frac{\partial v(\phi_{X,Y})}{\partial \phi_{X,Y}}, \tag{1}$$

where $v(\phi_{X,Y})$ denotes a function of acoustic velocities, which depends upon the angle $\phi_{X,Y}$ between the AW vector and the *X* (or *Y*) axis of the Cartesian coordinate system *XYZ*, while $\Delta_{X,Y}$ is the angle between the Poynting vector and the *X* (or *Y*) axis. Here *X* and *Y* are the axes perpendicular to the geometric plane under consideration. The angle of deviation of the acoustic polarization from a purely longitudinal type for the quasi-longitudinal (QL) wave can be found from the Christoffel equation [11]:

$$\zeta_{X,Y} = \phi_{X,Y} - \frac{1}{2}\arctan\left[\frac{\sin(2\phi_{X,Y})(C_{13} + C_{44})}{\cos^2(\phi_{X,Y})(C_{11} - C_{44}) + \sin^2(\phi_{X,Y})(C_{44} - C_{33})}\right], \tag{2}$$

which refers to the *YZ* and *XZ* planes. In Eq. (2), $\phi_{X,Y}$ is the angle between the AW vector and the *X* (or *Y*) axis. The non-orthogonality of the quasi-transverse (QT) waves can, in principle, be calculated using the same formula, with the only difference that the result given by formula (2) must be corrected by 90 deg.

The EO coefficients were measured with a Dixon–Cohen method at the wavelength of optical radiation $\lambda = 632.8$ nm. The AWs in the AO cell made from a standard material (e.g., fused quartz) were excited with a piezoelectric transducer fabricated from $LiNbO_3$ crystals. A longitudinal AW with the frequency f_A = 50 MHz excited in this manner was modulated by square pulses with the duration 0.5 μs and the repetition rate 300 μs. A sample was glued onto the AO cell. The experimental setup and the method itself have been described in detail in our recent work [12].

3. Results and discussion

The AW velocities determined by us are as follows: $v_{11} = v_{22} = 3233\pm17$, $v_{33} = 3150\pm13$, $v_{44} = 3098\pm22$, $v_{12} = v_{21} = 1713\pm3$, $v_{31} = 1756\pm13$, $v_{41} = 1718\pm11$, and $v_{4\bar{4}} = 1775\pm8$ m/s. Here the mean-square deviation is determined at the confidence level 95%. The elastic-stiffness and compliance coefficients calculated on this basis are presented in Table 1.

Table 1. Elastic-stiffness and elastic-compliance coefficients calculated for the γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals.

ij	11	33	12	13	44	66
C_{ij}, GPa	56.9±0.6	54.0±0.5	24.9±0.6	18.3±0.8	16.8±0.3	16.0±0.1
S_{ij}, 10^{-12} m^2/N	22.9±0.5	21.8±0.4	–8.5±0.5	–4.9±0.3	59.6±0.9	62.6±0.2

It is easily seen that both the elastic stiffnesses and the elastic compliances satisfy the conditions

$$C_{11} \approx C_{33},\ S_{11} \approx S_{33},\ C_{44} \approx C_{66}, S_{44} \approx S_{66},\ C_{12} = \left(C_{11} - C_{12}\right)/2,\ S_{66} = 2\left(S_{11} - S_{12}\right). \quad (3)$$

They are peculiar for the isotropic solid-state media. Using the elastic-stiffness coefficients and the Christoffel equation, one can obtain the dependences of AW velocities on the direction of AW propagation (see Fig. 2).

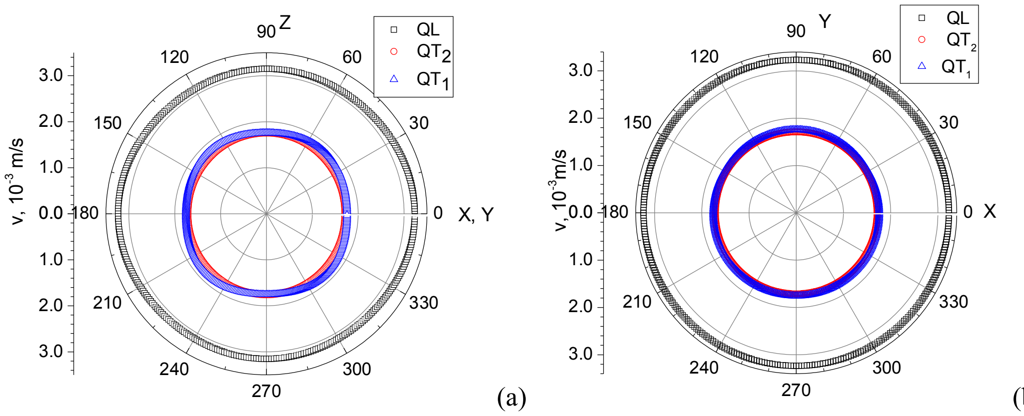

Fig. 2. Dependences of AW velocities found for the principal planes *XZ* (or *YZ*) (a) and *XY* (b) on the direction of AW propagation in γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals: black squares correspond to the QL AW, blue triangles to the AW QT_1 and red circles to the AW QT_2.

As seen from Fig. 2a, b, the AWs QT_1 and QT_2 propagate with the same velocities along the *Z* axis, so that this direction represents an acoustic axis. For the propagation directions parallel to the *X* (or *Y*) and *Z* axes in the *XZ* and *YZ* planes, the velocities of both the QL AW and the AWs QT_1 or QT_2 are almost the same. They are represented by quasi-circles. The maximal difference among the velocities of the QL AW is equal to (83±30) m/s. It corresponds to the propagation direction parallel to the *X* (or *Y*) and *Z* axes. A somewhat larger difference, (114±19) m/s, is typical for the QT AWs in the case of their propagation at the angle of 42 deg with respect to the *X* (or *Y*) axis. In the *XY* plane, the velocities of the AWs QT_1 and QT_2 are nearly equal to each other, and the corresponding difference is as small as (43±16) m/s. Besides, the velocities of all the three acoustic modes do not depend on the propagation direction in the *XY* plane. This result agrees well with the known fact that the hexagonal crystals represent transversely isotropic media, which means that the velocities of the QL AW and the AWs QT_1 and QT_2 do not depend on the direction of their propagation within the *XY* plane [13].

There can be two possible manifestations of the acoustic axes in these crystals [14]: (1) a cone of the acoustic axes around the *Z* axis is purely imaginary and the only acoustic axis is *Z*, or (2) a cone of the acoustic axes is a real circular cone aligned with the *Z* axis, while the acoustic axes are the *Z* direction and all the directions lying on this real cone. In the case of γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals, we deal with the case (1), i.e. a single acoustic axis is parallel to the *Z* axis. However, in the approximate case determined by the formulae (3), every direction of AW propagation represents the acoustic axis, with very small differences of the velocities of QT AWs.

The obliquity angle between the acoustic group-velocity direction and the AW vector is quite small (see Fig. 3). It reaches its maximal values, 5.8 deg at $\phi_{X,Y}$ = 23 deg and –5.8 deg at $\phi_{X,Y}$ = 68 deg, in case of the AW QT_1 and the *XZ* (or *YZ*) plane. The maximal obliquity values for the QL AW in these planes are equal to –3.0 deg at $\phi_{X,Y}$ = 28 deg and 3.0 deg at $\phi_{X,Y}$ = 152 deg. The maximal obliquity for the AW QT_2 is equal to +1.4 deg and –1.4 deg respectively at $\phi_{X,Y}$ = 45 deg and 135 deg. Finally, the obliquity is equal to zero in the *XY* plane.

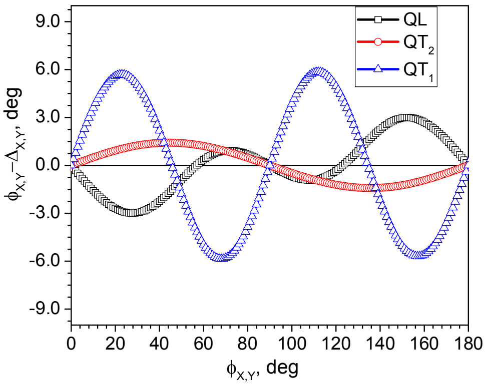

Fig. 3. Dependence of obliquity angle on the direction of AW propagation within *XZ* (or *YZ*) plane, as calculated for γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals: black squares correspond to the QL AW, blue triangles to the AW QT_1 and red circles to the AW QT_2.

The angle of deviation from the purely longitudinal polarization state is small enough for the QL AW propagating in the *XZ* (or *YZ*) planes. It reaches its maximums, +2.2 deg and –2.2 deg at $\phi_{X,Y}$ = 27 and 153 deg, respectively (see Fig. 4). The polarization deviation from the purely transverse state for the AW QT_1 is the same as for the QL AW. The angle ζ_Z is equal to zero in the *XY* plane. Hence, the AWs propagating in γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ along any direction can be accepted in some approximation as the waves with purely longitudinal (or transverse) polarization.

The efficiency of AO diffraction occurring in the γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals have been studied by the Dixon–Cohen method in different geometries. The propagation directions of light and AWs were parallel to the principal axes. The other geometry corresponded to the propagation of AWs along the bisector of –*Y* and *Z* axes and the light propagation along the bisector of *Y* and *Z* axes (with the polarization being parallel to the *X* axis). In all these geometries, we dealt with the isotropic AO diffraction of the ordinarily polarized optical radiation. It has been found that $|p_{11}| = 0.105 \pm 0.014$ and $|p_{13}| = 0.072 \pm 0.010$ for the EO coefficients. The appropriate AO figures of merit are equal to $M_2 = (38.6 \pm 0.7) \times 10^{-15} \mathrm{s}^3/\mathrm{kg}$ and $M_2 = (18.1 \pm 0.3) \times 10^{-15} \mathrm{s}^3/\mathrm{kg}$. When the AW propagates along the bisector of $-Y$ and Z axes and the light propagates along the bisector of Y and Z axes, the effective EO coefficient determined by us amounts to $p_{eff} = 0.06 \pm 0.01$. The corresponding AO figure of merit is then equal to $M_2 = (12.6 \pm 0.3) \times 10^{-15} \mathrm{s}^3/\mathrm{kg}$.

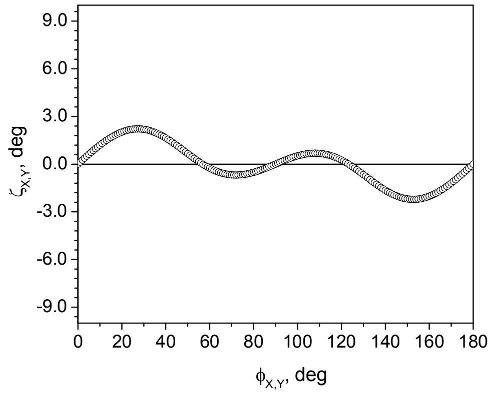

Fig. 4. Dependence of the angle of deviation from purely longitudinal polarization state, as calculated for the QL AW propagating inside the *XZ* (or *YZ*) plane of γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals.

The effective EO coefficient calculated with the coefficients p_{11} and p_{13} determined experimentally and the formula $p_{eff} = \frac{1}{2}(p_{11} + p_{13})$ is equal to 0.089 ± 0.024. It is close to the value measured directly. The coefficient $p_{12} = p_{21}$ has been measured under the condition of light propagation along the optic axis, which possesses a large optical rotation. Since the polarization of diffracted light has not been controlled, it is not clear whether the both (right-handed and left-handed) circularly polarized waves take part in the AO interaction or only one of these waves is involved in this interaction. Nonetheless, we have found $|p_{12}| = 0.09 \pm 0.03$ under the conditions mentioned above.

Notice that the main problem of our studies of the EO coefficients in γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ with the Dixon–Cohen method is relatively high optical absorption and linear dichroism that occur at the light wavelength 632.8 nm. This is the reason why we have not detected the AO diffraction in which the extraordinary light wave takes part. As a result, one can determine the other components of the EO tensor only in the region of longer light wavelengths.

4. Conclusions

As a result of our experimental studies of the AW velocities and the EO coefficients in the γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals, all the tensor components of the elastic stiffnesses and elastic compliances have been determined. It has been found that the components of these tensors satisfy the approximate relationships $C_{11} \approx C_{33}$, $S_{11} \approx S_{33}$, $C_{44} \approx C_{66}$, $S_{44} \approx S_{66}$, $C_{12} = (C_{11} - C_{12})/2$ and $S_{66} = 2(S_{11} - S_{12})$, which are peculiar for the isotropic solid-state media. The obliquity angles for all the acoustic modes are quite small. They reach their maximums (5.8 deg at $\phi_{X,Y} = 23$ deg and –5.8 deg at $\phi_{X,Y} = 68$ deg) for the AW QT_1 propagating in the *XZ* (or *YZ*) plane. Both the angle of non-orthogonality for the AW QT_1 and the deviation angle from the purely longitudinal polarization state are small for the QL AW. This implies that these AWs represent pure polarization states in some approximation.

The AO figure of merit for the γ_1-$(Ga_{0.3}In_{0.7})_2Se_3$ crystals has been measured in different geometries of isotropic diffraction. The maximal AO figure of merit determined by us is equal to $M_2 = (38.6 \pm 0.7) \times 10^{-15} s^3/kg$, which corresponds to the effective EO coefficient $|p_{11}| = 0.105 \pm 0.014$. Let us notice that, since the AW slownesses and the EO coefficients determined by us are not too large (e.g., we have $|p_{11}| = 0.105 \pm 0.014$, $|p_{13}| = 0.072 \pm 0.010$ and $|p_{12}| = 0.09 \pm 0.03$), the main contribution into the AO figure of merit is concerned with high refractive indices observed in the spectral region close to the optical absorption edge.

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Анотація. *У цій роботі експериментально досліджено швидкості акустичних хвиль і коефіцієнт акустооптичної якості (КАОЯ) для кристалів* γ_1*-*$(Ga_{0,3}In_{0,7})_2Se_3$*. Визначено всі складові тензорів пружної жорсткості та пружної податливості. З точки зору пружності, кристали поводяться як майже ізотропне твердотільне середовище. А саме, складові пружних жорсткості та податливості задовольняють наближеним співвідношенням* $C_{11} \approx C_{33}$, $S_{11} \approx S_{33}$, $C_{44} \approx C_{66}$, $S_{44} \approx S_{66}$, $C_{12} = (C_{11} - C_{12})/2$ *і* $S_{66} = 2(S_{11} - S_{12})$. *Кути зносу акустичної енергії і неортогональності поляризації досить малі для всіх акустичних мод. Максимальний КАОЯ, визначений при ізотропній дифракції, дорівнює* $M_2 = (38,6 \pm 0,7) \times 10^{-15}\,\text{с}^3/\text{кг}$. *Оскільки визначені нами сповільнення акустичних хвиль і пружно-оптичні коефіцієнти* p_{ij} *незначні (наприклад,* $|p_{11}| = 0,105 \pm 0,014$, $|p_{13}| = 0,072 \pm 0,010$ *і* $|p_{12}| = 0,09 \pm 0,03$*), то основний внесок до КАОЯ походить від високих показників заломлення, що характерно для спектрального діапазону, близького до оптичного краю поглинання.*