# Acoustic, elastic and acoustooptic properties of Tl<sub>3</sub>AsSe<sub>3</sub> crystals: acoustic isotropic point

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Abstract. We have experimentally studied acoustic wave (AW) velocities in Tl<sub>3</sub>AsSe<sub>3</sub> crystals. Basing on these results, the matrices of elastic stiffness and compliance coefficients are obtained and the cross sections of the AW velocity surfaces by the principal crystallographic planes are constructed. We have revealed a phenomenon termed as an acoustic isotropic point. We have found that, at the room temperature and the AW frequency equal to 10 MHz, the Tl<sub>3</sub>AsSe<sub>3</sub> crystals are close to the conditions of acoustic isotropy, which are analogous to the isotropic point known in crystal optics. Namely, the cross sections of the AW velocity surfaces under these conditions are almost circular and the difference between the velocities of transverse AWs with mutually orthogonal polarizations is very small. Then the specific relations among the elastic stiffness tensor components hold true, which are peculiar for the isotropic glass materials:  $C_{11} \approx C_{33}$ ,  $C_{12} \approx C_{13}$ ,  $C_{44} \approx C_{66} \approx (C_{11} - C_{12})/2$ , and  $C_{14} \approx 0$ . We have revealed that the Tl<sub>3</sub>AsSe<sub>3</sub> crystals exhibit very small obliquity of the acoustic energy flow with respect to the wave vector of the AW, as well as negligibly small angles of deviation of acoustic polarization from the purely transverse or longitudinal types. The acoustooptic figures of merit calculated for the interaction of waves propagating close to the principal crystallographic axes are as high as  $3068 \times 10^{-15}$  and  $3262 \times 10^{-15}$  s<sup>3</sup>/kg, respectively for the isotropic and anisotropic types of acoustooptic interactions.

**Keywords**: acoustic wave velocities, elastic stiffness, elastic compliance, acoustic anisotropy, acoustooptics

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## 1. Introduction

Tl<sub>3</sub>AsSe<sub>3</sub> crystals are ternary halcogenide semiconductors that belong to sulfo-salt compounds [1], with the band gap being equal to 0.98 eV [2]. The crystals refer to an acentric rhombohedral space symmetry group R3m (the point symmetry group 3m) and have the lattice parameters a = 9.870(2) and c = 7.094(3) Å [3]. Thallium arsenic selenide is optically transparent in the spectral range 1.23–18 µm [4]. The crystal represents one of the most efficient materials for the operation of optical radiation in the mid-IR spectral range [5]. For example, due to high nonlinear optical coefficients, Tl<sub>3</sub>AsSe<sub>3</sub> has been found to be promising for the second harmonic generation and parametrical oscillation at the carbone dioxide laser radiation wavelength,  $\lambda = 10.6$  µm [2, 6]. The material has also been considered useful for detecting X-ray and  $\gamma$ -ray radiations [7].

Perhaps, the most promising applications of thallium arsenic selenide refer to IR acoustooptic (AO) devices [8–10]. In particular, it has been employed as a working element of AO tunable filters [8, 9] in both collinear and non-collinear interaction geometries. At the wavelength of 3.39 µm, the AO figure of merit (AOFM) of these crystals is equal to 295 [4], as expressed in the relative units with respect to the AOFM value of the fused quartz ( $M_2 = 1.56 \times 10^{-15} \text{ s}^3/\text{kg}$  at  $\lambda = 0.6328 \,\mu\text{m}$ ). This value, which corresponds approximately to  $460 \times 10^{-15} \text{ s}^3/\text{kg}$  in the absolute units, has been obtained experimentally in Ref. [4] for the case of AO interactions with the longitudinal acoustic wave (AW) when the effective elastooptic coefficient (EEC)  $p_{ef}$  is equal to the tensor component  $p_{31}$ . Notice that the AW velocities for the Tl<sub>3</sub>AsSe<sub>3</sub> crystals have not been studied in a sufficient detail. As far as we know, a few components of the velocities have been determined. In particular, these are the velocities of the longitudinal waves that propagate along the directions [001], [010] and [210] (2100 m/s), as well as the velocities of the shear AWs propagating along [001] (1000 m/s) and along directions [010] and [210] (1050 m/s) [4].

As testified by the available experimental data, the anisotropy of AW velocities is rather weak. At the same time, it follows from Refs. [11, 12] that the anisotropy of EECs is more pronounced. Moreover, the EECs determined at 3.39  $\mu$ m are high enough. The refractive indices for Tl<sub>3</sub>AsSe<sub>3</sub> crystals are also high, being equal to  $n_o = 3.339$  and  $n_e = 3.152$  at 3.39  $\mu$ m [4, 13], where the indices *o* and *e* refer respectively to the ordinary and extraordinary optical waves. This implies that the anisotropy of EECs can provide some other geometries of AO interactions that yield the AOFM values higher than those mentioned above. As an example, the authors of Ref. [12] have found that the highest AOFM for the Tl<sub>3</sub>AsSe<sub>3</sub> crystals is equal to 2886 relative to the AOFM known for the fused silica. Thus, a further comprehensive analysis of different possible AO interaction geometries for thallium arsenic selenide seems to be important. To perform this analysis and determine the actual anisotropy of AW velocities, one needs to know all of the nonzero components of the elastic stiffness tensor and the elastooptic tensor. The present work represents a first step in this direction. It reports on experimental determination of the AW velocities for the Tl<sub>3</sub>AsSe<sub>3</sub> crystals and analyses of the acoustic and elastic anisotropy of this material.

#### 2. Experimental methods

Tl<sub>3</sub>AsSe<sub>3</sub> compound was synthesized from high-pure elemental thallium (99.99 wt. % Tl), arsenic (99.9998 wt. % As), and selenium (99.999 wt. % Se), which were additionally purified using a vacuum distillation. The synthesis was performed in quartz ampoules of special shape evacuated down to the pressure 0.13 Pa. The following regime was used: heating at the rate 40–50 K/h up to 637 K while rotating the ampoule for saturation, ageing at this temperature during 72 h, and then cooling down to the room temperature at the rate 50 K/h. The substance thus obtained was investigated using a differential thermal analysis and X-ray powder diffraction techniques (an X-ray diffractometer DRON-4; Cu K<sub>a</sub> radiation). A single endothermic maximum was observed at 587±2 K on the heating DTA curves for our samples, which corresponded to the melting temperature. Our experimental X-ray diffraction powder patterns agreed well with those calculated issuing from the structural data reported in the literature.

Tl<sub>3</sub>AsSe<sub>3</sub> single crystals were grown from a stoichiometric composition using a Bridgman method. The temperatures in the melting and annealing zones were equal respectively to 637 and

483 K. The solidification rate was equal to  $\sim 0.1-0.25$  mm/h and the temperature gradient at the solid–liquid interface to 4–6 K/mm. The crystals obtained by us (see Fig. 1) were annealed in the growth zone at 483 K for 168 h and then cooled down to the room temperature at the rate 4 K/h.



Fig. 1. Examples of our Tl<sub>3</sub>AsSe<sub>3</sub> crystalline samples grown with the Bridgman method.

We prepared a number of samples for the AW velocity measurements. The samples had parallelepiped shapes, with the dimensions ~  $6 \times 7 \times 9 \text{ mm}^3$  and the surfaces being perpendicular to the directions <100> and <110>. The AW velocities were measured with a standard pulse-echo overlap technique [14]. We excited the AWs in the samples using LiNbO<sub>3</sub> transducers (the resonance frequency f = 10 MHz, the bandwidth  $\Delta f = 0.1 \text{ MHz}$ , and the acoustic power  $P_a = 1-2 \text{ W}$ ). The acoustic velocities were denoted relative to the crystallographic axes, using the notations a = 1, b = 2 and c = 3.

### 3. Results and discussion

There are six independent nonzero elastic stiffness coefficients  $C_{klmn} = C_{\lambda\mu}$  for the trigonal crystals belonging to the point symmetry group 3m:  $C_{11}$ ,  $C_{12}$ ,  $C_{13}$ ,  $C_{33}$ ,  $C_{44}$  and  $C_{14}$  (with  $\lambda$ ,  $\mu = 1-6$ , and the notations 1 = 11, 2 = 22, 3 = 33, 4 = 23, 5 = 13 and 6 = 12). These coefficients have been calculated following from the known AW velocities and the theoretical relations

$$C_{11} = \rho v_{11}^2, \quad C_{33} = \rho v_{33}^2, \quad C_{44} = \rho v_{23}^2, \quad C_{66} = \rho v_{12}^2,$$

$$C_{12} = C_{11} - 2C_{66}, \quad C_{14} = \rho v_{14}^2 - 0.5(C_{44} + C_{66}),$$

$$C_{13} = C_{14} - C_{44} + \sqrt{(C_{33} - 2\rho v_{4\overline{4}}^2 + C_{44})(C_{11} - 2\rho v_{4\overline{4}}^2 + C_{44})}.$$
(1)

Here the crystal density  $\rho$  is taken to be 7827 kg/m<sup>3</sup>, which represents the average of the measured (7834±20 kg/m<sup>3</sup>) and calculated (7820 kg/m<sup>3</sup> [2]) density values, and  $v_{ij}$  are the AW velocities, with *i* and *j* being respectively the propagation and polarization directions. Using the known elastic stiffness coefficients and the Christoffel equation, one can construct the cross sections of the AW velocity surfaces by the principal crystallographic planes (see, e.g., Ref. [15]). The elastic compliance coefficients  $S_{\mu\nu}$  were calculated from the known elastic stiffness coefficients with the aid of the relation

$$C_{\lambda\mu}S_{\mu\nu} = \delta_{\lambda\nu} , \qquad (2)$$

where  $\delta_{\lambda\nu}$  implies the Kronecker delta.

The obliquity angle between the acoustic group velocity direction and the AW vector was calculated as [16]

$$\Delta_i = \arctan \frac{1}{\nu(\phi_i)} \frac{\partial \nu(\phi_i)}{\partial \phi_i} + \phi_i \,. \tag{4}$$

Here  $v(\phi_i)$  denotes a function of the acoustic velocity that depends upon the angle  $\phi_i$  between the wave vector and the corresponding axis of the crystallographic coordinate system, with the subscript *i* referring to the axis perpendicular to the geometric plane under consideration.

The angle of deviation of the acoustic polarization from a purely longitudinal type was calculated basing on the Christoffel equation [17]:

$$\zeta_k = \phi_k - \frac{1}{2} \arctan \frac{2M_{ij}}{M_{ii} - M_{jj}}, \qquad (5)$$

where  $M_{ij}$  are components of the Christoffel tensor and  $\phi_k$  is the angle between the AW vector and the crystallographic axis *i* in the crystallographic plane *ij*, whereas the axis *k* is perpendicular to the mentioned plane. The corresponding non-orthogonality of the quasi-transverse waves may, in principle, be calculated with the same formulae. The only difference is that the term 90° should be added to the right-hand side of Eq. (5).

The AW velocities obtained experimentally are collected in Table 1. It is seen that the velocities of the quasi-longitudinal waves are almost the same, lying in the range 2098–2221 m/s. The same is true of the quasi-transverse AW velocities (978–1073 m/s). Hence, the AW anisotropy in Tl<sub>3</sub>AsSe<sub>3</sub> is indeed weak.

Propagation direction	Polarization direction	AW velocity, m/s
[100]	[100]	2108±28
[010]	[010]	2098±28
[001]	[001]	2221±147
[011]	[011]	2143±30
[101]	[101]	2181±30
[110]	[110]	2081±47
[100]	[010]	1073±33
[100]	[001]	1009±12
[010]	[001]	1071±52
[011]	[100]	1017±9
[011]	$[01\overline{1}]$	978 ±16

Table 1. AW velocities measured for the Tl<sub>3</sub>AsSe<sub>3</sub> crystals.

The elastic stiffness tensor components calculated from Eq. (1) are as follows:  $C_{11} = 34.6 \pm 0.9$ ,  $C_{33} = 38.6 \pm 5.1$ ,  $C_{44} = 8.0 \pm 0.2$ ,  $C_{66} = 9.0 \pm 0.6$ ,  $C_{12} = 16.6 \pm 1.2$ ,  $C_{13} = 21.6 \pm 2.5$  and  $C_{14} = -0.40 \pm 0.33$  GPa. As a result, we have the approximate relationships  $C_{11} \approx C_{33}$ ,  $C_{12} \approx C_{13}$ ,  $C_{44} \approx C_{66} \approx (C_{11} - C_{12})/2$  and  $C_{14} \approx 0$ . The components of the elastic compliance tensor reveal the same symmetry properties as the elastic stiffness components. They are as follows:  $S_{11} = (46.3 \pm 4.7)$ ,  $S_{33} = (49.0 \pm 16.1)$ ,  $S_{12} = -(9.3 \pm 5.2)$ ,  $S_{13} = -(20.7 \pm 8.6)$ ,  $S_{14} = (2.8 \pm 2.3)$ ,  $S_{44} = (125.8 \pm 3.1)$ ,  $S_{66} = (111.2 \pm 6.7)$  (in the units of  $10^{-12}$  m<sup>2</sup>/N). Notice that the mentioned relationships among the elastic stiffnesses and the elastic compliances are peculiar for the isotropic media. In such a case the cross sections of the AW velocities by the principal crystallographic planes should be circular and the velocities of the two transverse acoustic eigenmodes should be the same. As see from Fig. 2, this is almost our case. Moreover, the appropriate cross sections obtained basing on the Christoffel equation are in fact almost circular and the difference between the velocities of the transverse AWs with the orthogonal polarizations is very small. As a result, the Tl<sub>3</sub>AsSe<sub>3</sub> crystals are similar to the glass media from the viewpoint of acoustic anisotropy.





**Fig. 2.** Cross sections of AW velocities by the principal crystallographic planes, as calculated for the  $Tl_3AsSe_3$  crystals: (a) *ac* plane, (b) *bc* plane and (c) *ab* plane. *e* denotes the displacement vector, while and QL and QT are quasi-longitudinal and quasi-transverse waves.

The equality of the velocities of the transverse acoustic eigenwaves, together with almost circular shapes of the cross sections of the AW velocity surfaces for all of the eigenwaves, imply that the Tl<sub>3</sub>AsSe<sub>3</sub> crystals are under the conditions of acoustic isotropy at the room temperature and the AW frequency of 10 MHz. A corresponding acoustic isotropic point represents an analogue of the isotropic point well known in the crystal optics. Let us remind that, in the latter case, the isotropic point can appear in the crystals of symmetries lower than the cubic one. Then their principal refractive indices become equal to each other under certain conditions given by the wavelength of optical radiation, the temperature, etc. (see, e.g., Ref. [15]). Together with the optical activity effect, the existence of the 'isotropic point' enables one to develop various optical devices, e.g. optical filters [18–20]. We are to note in this respect that a fourth-rank axial tensor with the internal symmetry  $\varepsilon V[V^3]$  describing the acoustic activity is not equal to zero for the point group 3m [15]. This is why the acoustic activity is allowed by the symmetry of Tl<sub>3</sub>AsSe<sub>3</sub> and, in case when the velocities of the orthogonal linearly polarized transverse AWs are equal to each other, the eigenwaves can become circularly polarized and propagate with different velocities resulting in the rotation of AW polarization.

As expected for the case of almost spherical surfaces of the AW velocities, the obliquity of the acoustic group velocity vector with respect to the phase velocity vector is small enough. It does not exceed ~ 4.6 deg, with the accuracy ~ 6% (see Fig. 3). The same is true of the non-orthogonality of the transverse AWs and the deviation of the displacement vector from the phase velocity vector for the longitudinal AWs. All of these deviation angles are approximately zero.





**Fig. 3.** Dependences of obliquity angle between the acoustic group velocity direction and the AW vector upon the angle between the wave vector direction and the corresponding crystallographic axes, as calculated for the  $TI_3AsSe_3$  crystals: (a) *ac*, (b) *bc* and (c) *ab* planes.

The AOFM can be calculated with the well-known formula [17]

$$M_2 = \frac{n_i^3 n_d^3 p_{ef}^2}{\rho v_{ij}^3} , \qquad (6)$$

where  $n_i$  and  $n_d$  denote the refractive indices of the incident and diffracted optical waves and  $p_{ef}$  is the EEC. The relations for the EEC for the crystals of the symmetry group 3m are quite complicated (see Refs. [21, 22]). Moreover, the signs of the EECs determined with the Dixon–Kohen method [11, 12] remain unknown. As a consequence, we can estimate the AOFM only for some particular AO interaction geometries, namely for those geometries under which the signs of EECs are not essential in the calculations.

As seen from Table 2, the highest AOFM value that can be reached at the isotropic diffraction is equal to  $3068 \times 10^{-15} \text{ s}^3/\text{kg}$ . According to Ref. [21], this diffraction corresponds to the so-called type II of AO interactions. This is the interaction of the longitudinal AW propagating along the *b* axis with the optical wave propagating close to the *c* axis and having polarization almost parallel to the *b* axis. Almost the same AOFM value  $(3024 \times 10^{-15} \text{ s}^3/\text{kg})$  is peculiar for the isotropic diffraction under the conditions when the longitudinal AW propagating along the *a* axis interacts with the optical wave that propagates close to the *c* axis and has the polarization parallel to the *a* axis (the type II of AO interactions [21]). It is worthwhile that our data for the AOFM is similar to those obtained in Ref. [12] for the same type of AO interactions.

The highest AOFM for the case of anisotropic diffraction  $(3262 \times 10^{-15} \text{ s}^3/\text{kg})$  has been found under the conditions that correspond to the type IX of AO interactions (the optical incident wave that propagates along the *a* axis and is polarized parallel to the *c* axis interacts with the transverse AW that propagates along the *c* axis and has polarization parallel to the *b* axis). On the other hand, we have not confirmed a high AOFM value reported in Ref. [12] for the case of anisotropic AO diffraction.

Type of AO	AW		v <sub>ij</sub> , m/s	Optical wave		Refractive	EEC module	AOFM,
interaction [21, 22]	Propagation direction	Polarization direction	(average value)	Propagation direction	Polarization direction	index	$p_{ef}$ [11]	10 <sup>-13</sup> s <sup>3</sup> /kg
Isotropic AO diffra	ction							
I	[100]	[100]	2108	almost parallel to [001]	[010]	$n_o \rightarrow n_o$	$p_{21} = 0.22$	915
I	[001]	[001]	2221	almost parallel to [100]	[010]	$n_o \rightarrow n_o$	$p_{23} = 0.24$	931
I	[010]	[010]	2098	almost parallel to [001]	[100]	$n_o \rightarrow n_o$	$p_{12} = 0.22$	928
Π	[100]	[100]	2108	almost parallel to [001]	almost parallel to [100]	$n_o \rightarrow n_o$	$p_{11} = 0.4$	3024
Π	[001]	[001]	2221	almost parallel to [100]	almost parallel to [001]	$n_e \rightarrow n_e$	$p_{33} = 0.22$	554
II	[010]	[010]	2098	almost parallel to [001]	almost parallel to [010]	$n_o \rightarrow n_o$	$p_{22} = 0.4$	3068
Π	[100]	[100]	2108	almost parallel to [010]	[001]	$n_e \rightarrow n_e$	$p_{31} = 0.2$	535
Ш	[010]	[001]	1071	[001]	[100]	$n_o \rightarrow n_o$	$p_{14} = 0.04$	231
Π	[010]	[001]	1071	almost parallel to [001]	almost parallel to [010]	$n_o \rightarrow n_o$	$p_{14} = 0.04$	231
IV	[001]	[010]	1009	almost parallel to [100]	[010]	$n_o \rightarrow n_o$	$p_{14} = 0.04$	276
Anisotropic AO diff	raction							
ΠΛ	[100]	[100]	2108	[100]	$[001] \rightarrow [010]$	$n_e \rightarrow n_o$	$p_{41} = 0.018$	5.2
VIII	[100]	[001]	1009	[001]	$[010] \rightarrow [100]$	$n_o \rightarrow n_e$	$p_{14} = 0.04$	276
IX	[100]	[010]	1081	[001]	$[010] \rightarrow [100]$	$n_o \rightarrow n_e$	$p_{66} = 0.09$	1135
IX	[001]	[010]	1009	[100]	$[001] \rightarrow [010]$	$n_e \rightarrow n_o$	$p_{44} = 0.15$	3262

Table 2. AOFMs of Tl<sub>3</sub>AsSe<sub>3</sub> crystal calculated for different AO interaction geometries ( $\lambda = 3.39 \text{ }\mu\text{m}$ ).

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#### 4. Conclusions

In the present work we have studied experimentally the AW velocities for the Tl<sub>3</sub>AsSe<sub>3</sub> crystals. Complete sets of the elastic stiffness and compliance tensor components have been determined and cross sections of the AW velocity surfaces by the principal crystallographic planes have been constructed. We have revealed a phenomenon of approximate acoustic isotropy existing in the Tl<sub>3</sub>AsSe<sub>3</sub> crystals. Namely, the conditions very close to the acoustic isotropic point are realized at the room temperature and the AW frequency equal to 10 MHz. It is analogous to the isotropic point phenomenon known in the crystal optics. In fact, the cross sections of the AW velocities turn out to be almost circular, whereas the differences in the velocities of the transverse AW having orthogonal polarizations become very small.

Under the above conditions, we have found the following relations among the elastic stiffness coefficients:  $C_{11} \approx C_{33}$ ,  $C_{12} \approx C_{13}$ ,  $C_{44} \approx C_{66} \approx (C_{11} - C_{12})/2$  and  $C_{14} \approx 0$ . In their exact forms, these formulae are peculiar for the isotropic media. The components of the elastic compliance tensor manifest the same symmetry properties. Hence, issuing from the viewpoint of the acoustic anisotropy, the Tl<sub>3</sub>AsSe<sub>3</sub> crystals are similar to the glass media.

We have also proved that Tl<sub>3</sub>AsSe<sub>3</sub> exhibits very small obliquity of the acoustic energy flow with respect to the AW vector. The same is true of the angle of non-orthogonality and the deviation angle of AW polarization from the purely longitudinal type. All of these angles turn out to be negligibly small.

Finally, we have calculated the AOFM values for some AO interactions of the waves propagating close to the principal crystallographic axes. In particular, the highest AOFMs are equal to  $3068 \times 10^{-15}$  and  $3262 \times 10^{-15}$  s<sup>3</sup>/kg. They refer respectively to the isotropic and anisotropic interaction types under the conditions when the AWs and the optical waves propagate close to the principal crystallographic axes.

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Анотація. У роботі експериментально визначено швидкості поширення акустичних хвиль у кристалах Tl<sub>3</sub>AsSe<sub>3</sub>. На основі цих результатів одержано матриці коефіцієнтів жорсткості і податливості, а також побудовано перетини поверхонь швидкостей акустичних хвиль головними кристалографічними площинами. Нами було виявлене явище, так званої, акустичної ізотропної точки. Було встановлено, що кристали Tl<sub>3</sub>AsSe<sub>3</sub> при кімнатній температурі і частоті акустичних хвиль 10 МГц перебувають при умовах близьких до акустичної ізотропної точки – ефекту, який є аналогічним до ефекту ізотропної точки в кристалооптииі. Перетини поверхонь швидкостей акустичних хвиль в кристалах Tl<sub>3</sub>AsSe<sub>3</sub> є майже коловими, а різниця між швидкостями поширення поперечних хвиль з ортогональною поляризацією є дуже малою. При цьому співвідношення між компонентами тензора пружних модулів є наступними:  $C_{11} \approx C_{33}$ ,  $C_{12} \approx C_{13}$ ,  $C_{44} \approx C_{66} \approx (C_{11} - C_{12})/2$  і  $C_{14} \approx 0$ . Виявлено, що кристали Tl<sub>3</sub>AsSe<sub>3</sub> проявляють досить малий знос потоку енергії акустичних хвиль відносно хвильового вектора і нехтувано малий кут відхилення стану поляризації від чисто поперечного і поздовжнього типу. Розраховані значення коефіцієнтів акустооптичної якості для взаємодії хвиль, які поширюються у напрямках близьких до головних кристалографічних осей є досить великими і досягають значень 3068×10<sup>-15</sup>с<sup>3</sup>/кг and  $3262 \times 10^{-15} c^3 / \kappa c \, \partial_{\pi} \pi$  ізотропної і анізотропної взаємодії, відповідно.