

Acoustic and acousto-optic properties of $\text{Ge}_a\text{Se}_b\text{Te}_c$ and $\text{Si}_a\text{Se}_b\text{Te}_c$ glasses in case diffraction on longitudinal and shear acoustic waves

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Received on June 10, 2021

Revised on July 01, 2021

Accepted on July 02, 2021

We investigated acoustic and acousto-optic properties of glasses based on germanium, selenium, silicon and tellurium. The results of measurement are presented for two group of glasses including germanium, selenium and tellurium (GeSeTe) and silicon and tellurium (SiTe). The major characteristics of glasses such as the velocity of longitudinal and shear acoustic waves as well as the values of figure of merit are presented. The values of figure of merit are obtained by diffraction on longitudinal and shear acoustic waves at the wavelength of optical radiation $\lambda = 3.39 \mu\text{m}$. We also present the results of measurements of acoustic attenuation values α for longitudinal and shear acoustic waves in tellurium glasses at ultrasound frequencies in the range $f = 75\text{--}100$ MHz.

Keywords: acousto-optics, acousto-optic figure of merit, germanium, selenium, silicon, tellurium, attenuation acoustic waves.

DOI: 10.21883/EOS.2022.13.53985.2417-21

1. Introduction

Currently the acousto-optic (AO) effect is widely used to control the parameters of electromagnetic radiation in optics, laser technology, spectroscopy, as well as telecommunication systems and devices for optical information processing. Acousto-optic devices are characterized by high speed, absence of mechanical moving elements, low electrical and optical losses [1,2]. At present AO devices have already been created and widely used to control ultraviolet ($\lambda = 0.01\text{--}0.4 \mu\text{m}$), visible ($\lambda = 0.4\text{--}0.74 \mu\text{m}$) and near infrared ($\lambda = 0.74\text{--}2.5 \mu\text{m}$) range of the electromagnetic spectrum [3,4]. Often the paratellurite crystals (TeO_2), lithium niobate (LiNbO_3), quartz ($\alpha\text{-SiO}_2$) and calcium molybdate (CaMoO_4) [1,2] are used in acousto-optics of the visible and near infrared ranges. However, with an increase in the wavelength of optical radiation and the transition to the medium ($\lambda = 2.5\text{--}7 \mu\text{m}$) and long-wavelength ($\lambda = 7\text{--}15 \mu\text{m}$) infrared ranges the question of efficient AO materials remains open [5]. The most common material is a germanium (Ge) crystal, however, this is a cubic, therefore, optically isotropic material, which limits its scope of application in the acousto-optics [1,2,5,6].

Currently many materials are considered in the literature as the basis for AO interaction in the medium and long wavelength ranges. They include a tellurium crystal [7,8], cubic crystals based on thallium halides (KRS) [9–11], as well as mercury-based compounds: calomel, bromide and mercuric iodide [12].

In this study the amorphous alloys or glasses based on germanium (Ge), selenium (Se), silicon (Si), and tellurium (Te) are investigated. The presented results continue and complete the series of studies [13,14]. The properties of

shear acoustic waves in the glasses under consideration are experimentally investigated, and the AO quality factors M_2 are determined for the optical radiation diffraction with a wavelength of $\lambda = 3.39 \mu\text{m}$ on a shear wave. The attenuation factors are measured for a longitudinal acoustic wave at a frequency of $f = 100$ MHz and for a shear acoustic wave at a frequency of $f = 75$ MHz. The results obtained show the potential of using tellurium glasses as the basis for AO devices controlling electromagnetic radiation in the medium and long wavelength range.

2. Description of the studied glasses and method of measuring their properties

In this study we studied several glass samples consisting of three chemical elements: germanium (Ge), selenium (Se), and tellurium (Te), as well as one glass sample consisting of silicon (Si), selenium (Se) and tellurium (Te). Following the classification adopted earlier, we will call glasses consisting of germanium, selenium and tellurium belonging to the GeSeTe system, and glasses made of silicon, selenium and tellurium — glasses from the SiSeTe [13,14] system. The general view of the glasses is shown in Fig. 1, on which each glass is signed taking into account its chemical composition: $\text{Ge}_{33}\text{Se}_{33}\text{Te}_{33}$, $\text{Ge}_{30}\text{Se}_{30}\text{Te}_{40}$, $\text{Ge}_{30}\text{Se}_{25}\text{Te}_{45}$, $\text{Ge}_{25}\text{Se}_{15}\text{Te}_{60}$ and $\text{Si}_{25}\text{Te}_{75}$. Hereinafter the chemical composition of the glasses is indicated in percent, and $\text{Ge}_{33}\text{Se}_{33}\text{Te}_{33}$ glass includes germanium, selenium, and tellurium in equal proportions.

For the specified list of samples, measurements of their acoustic and AO properties were carried out. Acoustic properties were measured using two methods — echo-pulses method and AO method. The AO quality M_2 was

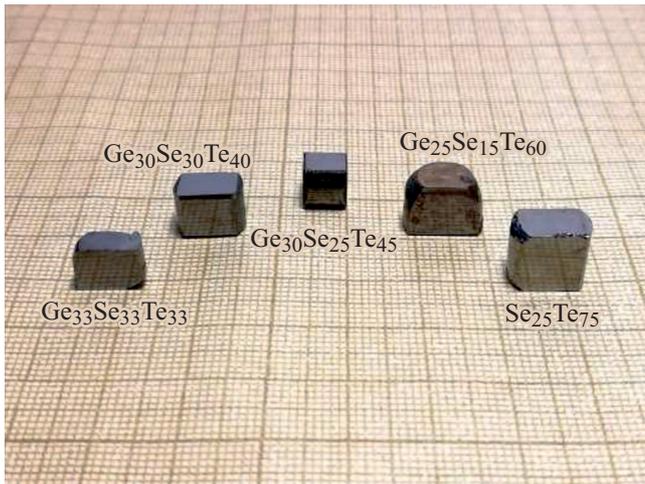


Figure 1. General view of the investigated glasses based on germanium, selenium, silicon and tellurium.

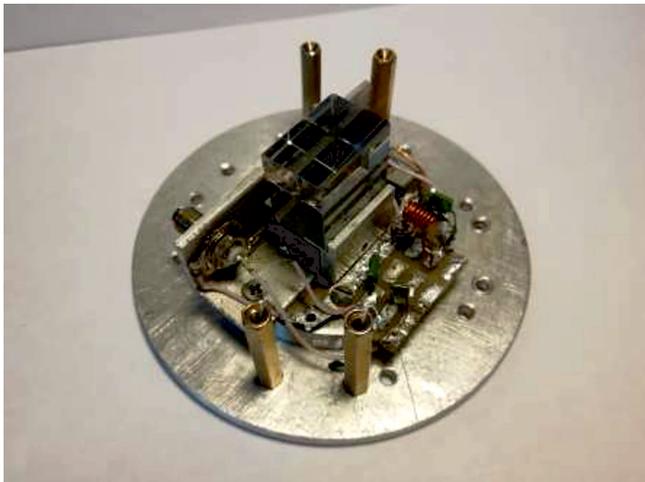


Figure 2. General view of the fused quartz buffer (SiO_2).

measured using the standard Dixon method [1,2]. Fused quartz SiO_2 was chosen as the material for the buffer, to which two piezoelectric transducers were attached. The first of them excited a longitudinal acoustic wave in the frequency range $f = 75\text{--}110$ MHz, the second — shear acoustic wave in the frequency range $f = 55\text{--}75$ MHz. A photograph of the buffer used is shown in Fig. 2.

In the course of an experimental study, the acoustical attenuation coefficients of longitudinal α_L and shear α_S acoustic waves propagating in tellurium glasses were measured. These measurements were carried out using the AO method, and their results are discussed in detail below.

All measurements were carried out on the unit, its scheme is given in Fig. 3. As the source of optical radiation He–Ne-laser LGN-118, which produced radiation on three wavelengths: $\lambda = 0.63, 1.15, 3.39\ \mu\text{m}$ was used. Two laser wavelengths were used in the study: $0.63\ \mu\text{m}$ for working with buffer and $3.39\ \mu\text{m}$ for working with tellurium glasses.

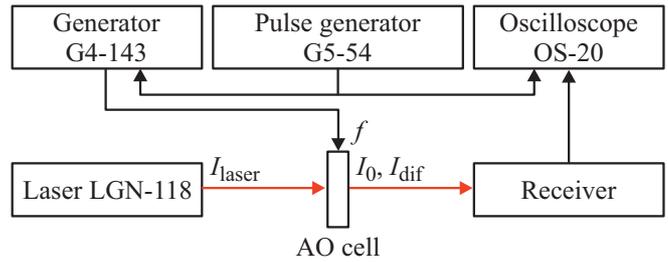


Figure 3. Experimental unit scheme.

Table 1. The velocity of acoustic waves in alloys of GeSeTe and SiSeTe systems

Chemical composition	Velocity of longitudinal acoustic waves [14,15] v_L , m/s	Velocity of shear acoustic waves v_S , m/s
$Si_{25}Te_{75}$	1920 ± 50	1160 ± 20
$Ge_{33}Se_{33}Te_{33}$	2230 ± 20	1260 ± 20
$Ge_{30}Se_{30}Te_{40}$	2200 ± 20	1260 ± 30
$Ge_{30}Se_{25}Te_{45}$	2200 ± 30	1230 ± 20
$Ge_{30}Se_{25}Te_{60}$	2050 ± 10	1210 ± 20

Laser radiation I_{laser} was incident on the AO cell, which was fixed on a turntable. Transmitted I_0 and diffracted I_{dif} radiation were recorded by two detectors: for visible radiation, a photodiode-based detector was used, for IR radiation — a detector based on gold-doped germanium (e: Au), which was cooled with liquid nitrogen. Longitudinal and shear acoustic waves were created in the buffer using piezoelectric transducers, to which a signal with a frequency of f was sent from a HF signal generator G4-143. External synchronization for the generator and oscillograph was provided by a pulse generator G5-54.

3. Measuring the speed of shear acoustic waves in tellurium glasses

The buffer based on fused quartz SiO_2 , described in the previous section was used to determine the phase velocity of shear acoustic waves in tellurium glasses. The results of these measurements are shown in Table 1. Also, for comparison and completeness of the description of the physical properties of the alloys under consideration, Table 1 lists the values of the velocities of longitudinal acoustic waves already known from the literature [14,15]. It should be noted, that the presented values of shear acoustic wave velocities were measured by the echo-pulse method in all glasses. In this study the measurements of velocities by the AO method were performed, however, it was not possible to register AO diffraction on shear acoustic waves in all samples (more details in the next section).

As follows from the results presented in Table 1, the studied glasses have sufficiently low ultrasound velocities. Thus, the values of the velocities of longitudinal acoustic waves presented in the second column of Table 1 slightly exceed the value of 2000 m/s. The highest value of the velocity of longitudinal waves was obtained for glass with the chemical composition $\text{Ge}_{33}\text{Se}_{33}\text{Te}_{33}$ and turned out to be equal to $v_L = (2230 \pm 20)$ m/s, and the smallest value — for glass $\text{Si}_{25}\text{Te}_{75}$ $v_L = (1920 \pm 50)$ m/s [14].

The experimentally measured speed of shear waves in tellurium glasses is presented in Table 1 in the third column. For example, for glass with the chemical composition $\text{Ge}_{33}\text{Se}_{33}\text{Te}_{33}$ it turned out to be equal to $v_S = (1260 \pm 20)$ m/s. The obtained value is the maximum in those samples, which were studied in this work. The minimum value was obtained for $\text{Si}_{25}\text{Te}_{75}$ glass and turned out to be $v_S = (1160 \pm 20)$ m/s. Such low velocities of shear acoustic waves allow to experimentally study the AO interaction in tellurium glasses at the optical radiation wavelength $\lambda = 3.39 \mu\text{m}$ using shear acoustic waves.

4. Measurement of AO quality in tellurium glasses

In the work, the AO characteristics of glasses were experimentally determined, in particular, the AO quality factor M_2 . In the literature there is information about AO quality values in tellurium-based alloys during diffraction by a longitudinal acoustic wave [13–15]. For ease of perception, these AO quality values are presented in Table 2 — the data in the second and third columns are taken from the study [14], the data in the fourth and fifth columns are taken from the study [15]. It should be noted, that in the literature the AO quality factor for diffraction by a longitudinal acoustic wave is given for two optical radiation polarization states (wavelength $\lambda = 3.39 \mu\text{m}$): the polarization vector is parallel ($M_{2\parallel}$) and orthogonal ($M_{2\perp}$) to the ultrasonic wave vector [14,15].

It follows from the values presented in the Table 2, that AO quality during diffraction by longitudinal acoustic waves strongly depends on the optical radiation polarization [14]. From this it can be assumed, that the numerical values of the photoelastic constants p_{11} and p_{12} of glasses differ from each other. It is impossible to directly calculate the values of photoelastic constants based on the AO quality M_2 values presented in the study [14], since the numerical values of the refraction indices n at the investigated wavelength ($\lambda = 3.39 \mu\text{m}$) are unknown. However, it is possible to estimate the AO quality M_{2S} for shear wave diffraction using the following expression:

$$M_{2S} = \frac{((p_{11} - p_{12})/2)^2 n^6}{\rho v_S^3} = (\sqrt{M_{2\parallel}} - \sqrt{M_{2\perp}})^2 \frac{v_L^3}{4v_S^3}. \quad (1)$$

The results of these calculations are presented in the Table 3 in the second column for all glasses under study (M_{2S}^*). The third column of the Table 3 presents the obtained experimental data on AO quality value (M_{2S}), which were obtained by the Dixon method with respect to the buffer on fused quartz SiO_2 . The obtained experimental data were processed according to a formula, that takes into account both different wavelengths used for working with a buffer ($\lambda_b = 0.63 \mu\text{m}$) and with tellurium glasses ($\lambda_x = 3.39 \mu\text{m}$) and different regions of AO interaction in the buffer (l_b) and in tellurium glasses (l_x):

$$\frac{M_{2x}}{M_{2B}} = \sqrt{\frac{I_4 I_5}{I_1 I_3}} \frac{I_o^b}{I_o^x} \left(\frac{\lambda_x \cos \theta_x}{\lambda_b \cos \theta_b} \right)^2 \frac{l_b}{l_x}, \quad (2)$$

where M_{2x} and M_{2B} — AO quality factors of the sample and buffer under study, I_1 and I_3 — intensities of diffracted radiation on direct and reflected acoustic waves during diffraction in a buffer, I_4 and I_5 — intensities of diffracted radiation on direct and reflected acoustic waves during diffraction in the sample under study, I_o^b — intensity of radiation transmitted through the buffer at wavelength λ_b , I_o^x — intensity of the radiation transmitted through the sample under study, at wavelength λ_x , θ_b and θ_x — the Bragg angles for diffraction in the buffer and in the sample under study respectively.

It can be seen from the results presented in the Table 3, that in the glasses of the GeSeTe and SiSeTe systems, a rather effective AO interaction is observed on shear acoustic waves. The maximum AO quality factor reaching $(90 \pm 10) \cdot 10^{-15} \text{ s}^3/\text{kg}$ was obtained for $\text{Ge}_{25}\text{Se}_{15}\text{Te}_{60}$. It is known from the literature, that the glasses under consideration have a fairly large range of optical transparency $\lambda = 1.5\text{--}20 \mu\text{m}$ [13–15], which makes the glasses under consideration promising for use in acousto-optics.

It should also be noted, that the calculated estimated values of the AO quality M_{2S}^* (Table 3, second column) are in good agreement with the experimentally measured values M_{2S} (Table 3, third column). This coincidence confirms the correctness of the obtained results on the AO quality value M_2 at diffraction by both longitudinal and shear acoustic waves in tellurium glasses [14]. It should be noted, that based on the estimated values M_{2S}^* in $\text{Ge}_{33}\text{Se}_{33}\text{Te}_{33}$ the AO quality value at diffraction by a shear acoustic wave does not exceed several units: $M_{2S}^* = 2 \cdot 10^{-15} \text{ s}^3/\text{kg}$. Due to the low AO quality value, an experimental study on a shear acoustic wave with this glass was not carried out. Also, in the course of the work, it was not possible to measure the AO quality in $\text{Ge}_{30}\text{Se}_{25}\text{Te}_{45}$ glass, which is due to technical difficulties in the search and registration of reflected pulses in the studied glass and buffer. Thus, out of the five AO glasses studied in this work, shear-wave diffraction was observed only in three of them. This explains the fact, that in the previous section, when describing the velocities of shear acoustic waves in glasses, the values obtained by the echo-pulses method, rather than AO method, were given.

Table 2. AO quality factor for diffraction by a longitudinal acoustic wave for glasses of the SiSeTe and GeSeTe systems

Chemical composition	$M_{2\parallel}, 10^{15} \text{ s}^3/\text{kg}$, [14]	$M_{2\perp}, 10^{15} \text{ s}^3/\text{kg}$, [14]	$M_{2\parallel}, 10^{15} \text{ s}^3/\text{kg}$, [15]	$M_{2\perp}, 10^{15} \text{ s}^3/\text{kg}$, [15]
Si ₂₅ Te ₇₅	1200 ± 200	800 ± 100	—	—
Ge ₃₃ Se ₃₃ Te ₃₃	380 ± 80	300 ± 40	—	—
Ge ₃₀ Se ₃₀ Te ₄₀	420 ± 80	240 ± 40	500	420
Ge ₃₀ Se ₂₅ Te ₄₅	440 ± 80	240 ± 40	680	580
Ge ₂₅ Se ₁₅ Te ₆₀	1300 ± 250	700 ± 100	1320	1100

Table 3. AO quality factor for diffraction by a shear acoustic wave for glasses of the SiSeTe and GeSeTe systems

Chemical composition	$M_{2S}^*, 10^{15} \text{ s}^3/\text{kg}$	$M_{2S}, 10^{15} \text{ s}^3/\text{kg}$
Si ₂₅ Te ₇₅	45	65 ± 10
Ge ₃₃ Se ₃₃ Te ₃₃	2	—
Ge ₃₀ Se ₃₀ Te ₄₀	35	23 ± 2
Ge ₃₀ Se ₂₅ Te ₄₅	45	—
Ge ₂₅ Se ₁₅ Te ₆₀	110	90 ± 10

5. Measurement of attenuation of longitudinal and shear acoustic waves in tellurium glasses

In the course of an experimental study of AO interaction in tellurium glasses, measurements were made to estimate the attenuation coefficient of longitudinal α_L and shear α_S ultrasonic waves. The measurements were performed using the AO method, the essence of which is to measure the intensity of diffracted light depending on the distance to the ultrasound exciter [16–18]. A buffer was used as an ultrasound exciter, the parameters of which were described above in the Section 2. The acoustic waves attenuation coefficients were measured at fixed frequencies of ultrasound, which amounted to $f = 100$ MHz for a longitudinal acoustic wave, and $f = 70$ MHz for a shear wave. It is important to note, that all measurements were carried out at low acoustic power P_a , therefore, the intensity of diffracted light is proportional to the applied acoustic power: $I_d \sim P_a$ [1,2,16–18].

It is known from theory, that the dependence of the diffracted radiation intensity on the distance can be approximated by the Bouguer law [16–18]:

$$I_d = I_{d\max} \exp(-\alpha L) \quad (3)$$

or in the case of low attenuation by a linear dependence

$$I_d = I_{d\max} \exp(-\alpha L) \approx I_{d\max}(1 - \alpha L), \quad (4)$$

where $I_{d\max}$ — intensity of diffracted radiation near the ultrasound exciter, I_d — intensity of diffracted radiation at a distance L from the exciter, α — attenuation coefficient of ultrasound [cm^{-1}].

The measurement results are shown in Fig. 4, *a–e* for a longitudinal acoustic wave and in Fig. 5, *a–d* for a shear acoustic wave. On all dependences the experimental data are represented by dots, and their interpolation — by solid lines. Also on the graphs there is a vertical dashed line, which displays the boundaries of the studied glass. A sharp decrease in the diffracted light intensity near the dashed line is not associated with the ultrasonic wave attenuation, but with a decrease in the AO interaction region.

It can be seen from the dependences presented, that the investigated tellurium glasses have a rather low acoustic attenuation coefficient for longitudinal acoustic waves (α_L). Therefore, formula (4) was used in processing the experimental data, and the approximation parameters are shown on each of the graphs. The final results are presented in Table 4 in the second and third columns for longitudinal acoustic waves.

The results of similar measurements for shear waves are presented in Fig. 5, *a–d*, and the final results — in Table 4 in the fourth and fifth columns. Note, that the measurements for shear waves were approximated by formula (3).

In the literature there are values of the attenuation coefficient for a longitudinal acoustic wave depending on the chemical composition of the glasses of the GeSeTe [11] system. They are listed in the fourth column in the Table 4 [11]. It can be seen from the obtained results, that glasses have relatively low attenuation for longitudinal acoustic waves at a frequency $f = 100$ MHz. The maximum attenuation coefficient was obtained for Si₂₅Te₇₅ glass and amounted to $\alpha_L = 3.2$ dB/cm. However, for shear waves, the attenuation coefficient is already quite large, about 10 dB/cm on average, that should taken into account when developing AO devices based on tellurium glasses. It is important to note, that the large error in the experimental results for Ge₃₀Se₂₅Te₄₅ glass is most likely due to the inhomogeneity of its glass volume properties. It also follows from the dependences shown in Figs 4, 5, that the measurements were carried out at fairly short propagation lengths of acoustic waves, that directly follows from the small dimensions of the samples themselves (~ 5 mm). Therefore, to obtain more accurate results, it is necessary to carry out a series of similar measurements on tellurium glass samples with large geometric dimensions.

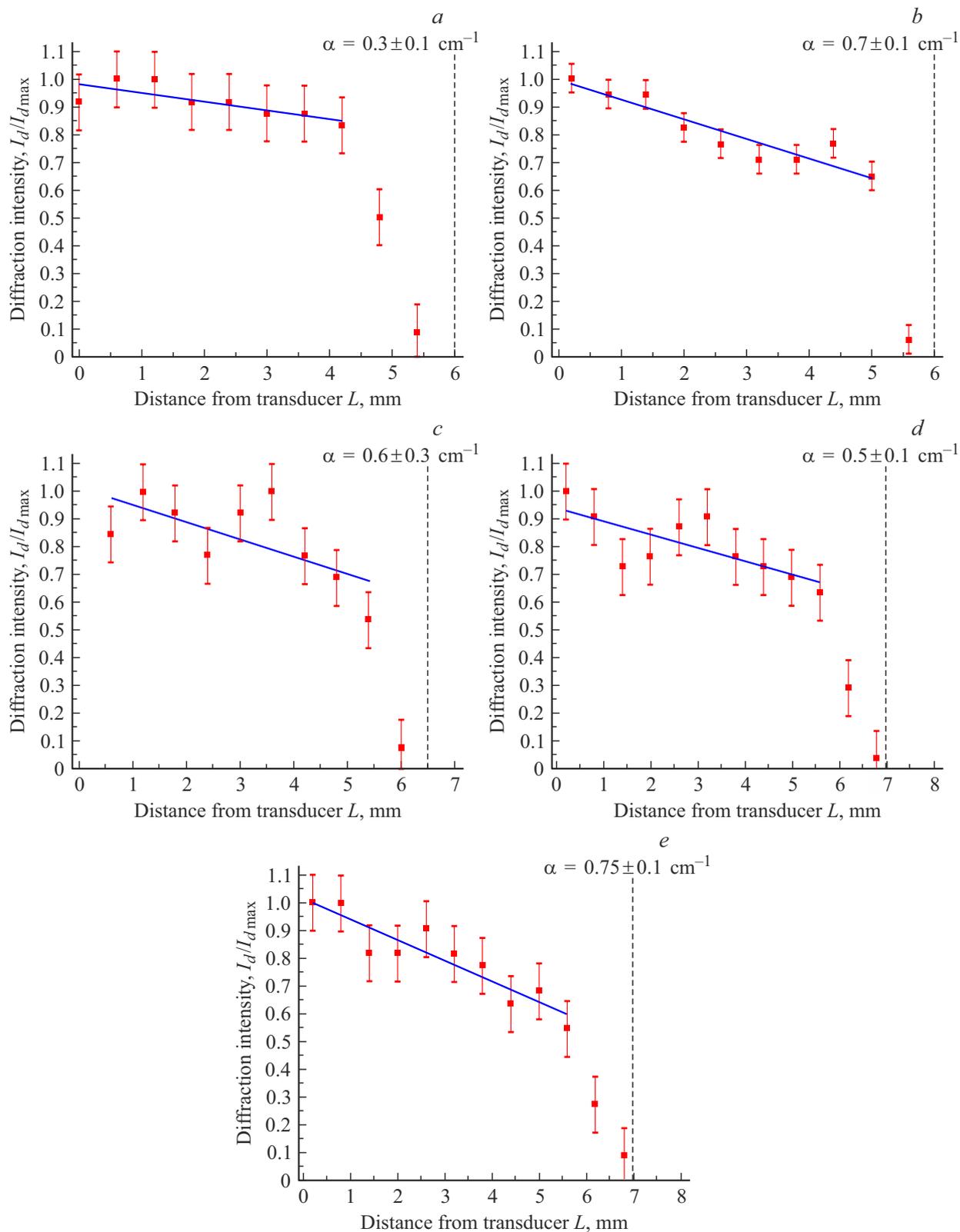


Figure 4. Dependences of the diffraction intensity on the distance to the ultrasound exciter during AO interaction on a longitudinal acoustic wave in tellurium glasses with different chemical compositions: (a) GeSeTe, (b) Ge₃₀Se₃₀Te₄₀, (c) Ge₃₀Se₂₅Te₄₅, (d) Ge₂₅Se₁₅Te₆₀, (e) Si₂₅Te₇₅.

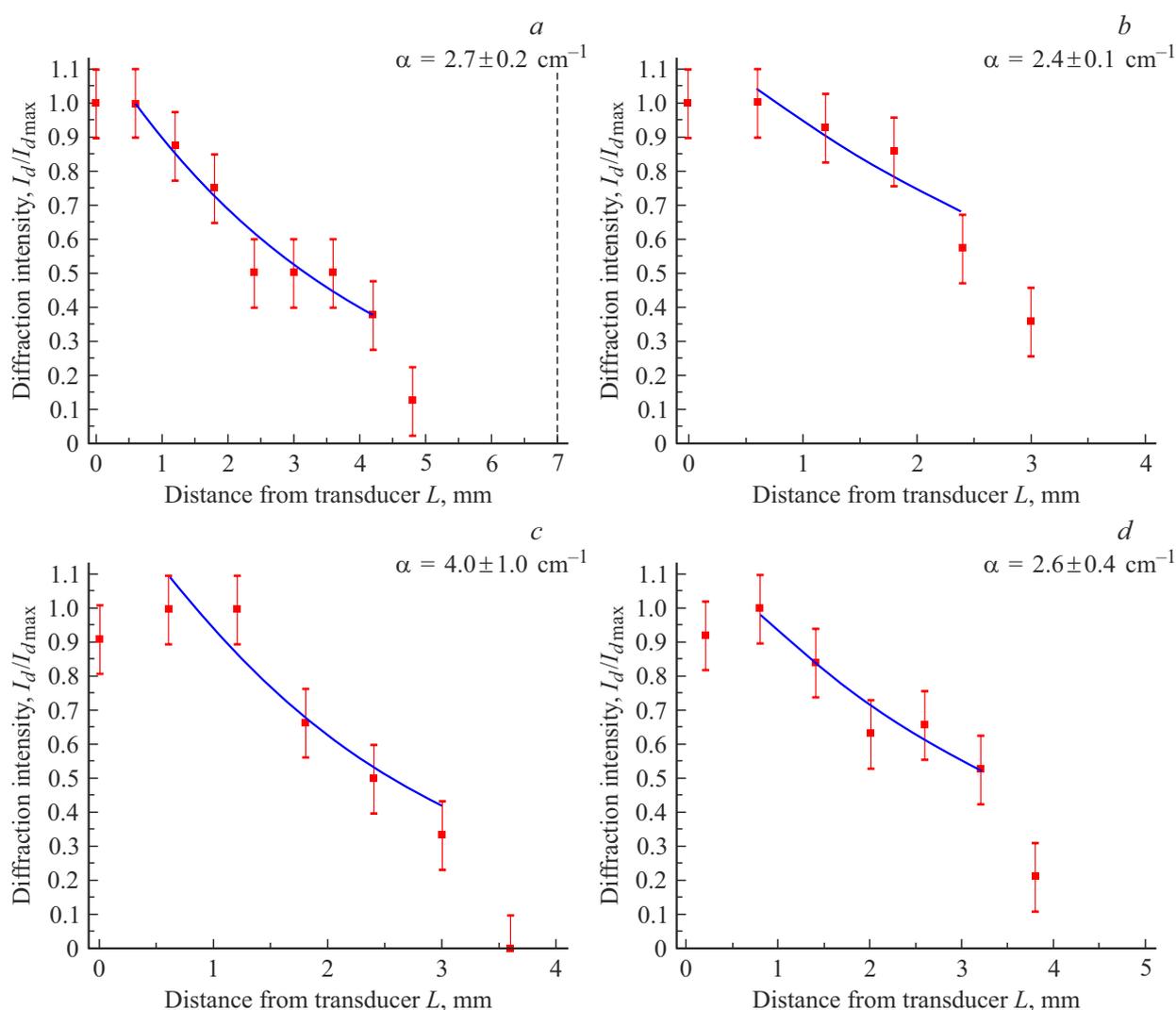


Figure 5. Dependences of the diffraction intensity on the distance to the ultrasound exciter during AO interaction on a shear acoustic wave in tellurium glasses with different chemical compositions: (a) $Si_{25}Te_{75}$, (b) $Ge_{30}Se_{30}Te_{40}$, (c) $Ge_{30}Se_{25}Te_{45}$, (d) $Ge_{25}Se_{15}Te_{60}$.

Table 4. Attenuation coefficients of longitudinal and shear acoustic waves for the glasses of SiSeTe and GeSeTe systems

Chemical Composition	Attenuation of longitudinal wave α_L , cm^{-1}	Attenuation of longitudinal wave α_L , dB/cm	Attenuation of longitudinal wave α_L , dB/cm [11]	Attenuation Shear wave α_S , cm^{-1}	Attenuation Shear wave α_S , dB/cm
$Si_{25}Te_{75}$	0.75 ± 0.1	3.2 ± 0.4	7	2.7 ± 0.2	13 ± 1
$Ge_{33}Se_{33}Te_{33}$	0.3 ± 0.1	1.3 ± 0.4	—	—	—
$Ge_{30}Se_{30}Te_{40}$	0.7 ± 0.1	3.0 ± 0.4	—	2.4 ± 0.2	10.5 ± 0.5
$Ge_{30}Se_{25}Te_{45}$	0.6 ± 0.3	2.6 ± 1.3	—	4.0 ± 1.0	17 ± 4
$Ge_{25}Se_{15}Te_{60}$	0.5 ± 0.1	2.2 ± 0.4	2	2.6 ± 0.4	11 ± 2

6. Results and conclusions

The results of an experimental study of glasses based on germanium, selenium, silicon and tellurium are presented. The velocities of shear acoustic waves in glasses of the GeSeTe and SiSeTe systems are determined for the first

time. The highest velocity was obtained for glass with the chemical composition $Ge_{33}Se_{33}Te_{33}$ and turned out to be 1260 ± 20 m/s. Such low absolute values of shear wave velocities make glasses promising materials for acoustics and acousto-optics, for example, for constructing acoustic delay lines on their basis.

Moreover, for these glasses the AO quality coefficient was measured during diffraction by a shear acoustic wave. The maximum AO quality factor was obtained for $\text{Ge}_{25}\text{Se}_{15}\text{Te}_{60}$ glass and turned out to be equal to $(90 \pm 10) \cdot 10^{-15} \text{ s}^3/\text{kg}$. This shows the promise of using tellurium glasses in acousto-optics of the near, middle, and long-wavelength IR ranges, for example, as an alternative to germanium (Ge) single crystals. Besides, the results obtained confirm a fairly strong difference in the numerical values of the photoelastic constants p_{11} and p_{12} in the alloys under consideration.

The results of measurements of the attenuation of longitudinal and shear acoustic waves are also presented. It has been experimentally obtained, that the attenuation coefficient of longitudinal waves with a frequency $f = 100 \text{ MHz}$ is rather small and averages 3 dB/cm . At the same time, it was established for the first time, that the attenuation coefficient for shear acoustic waves at the ultrasonic frequency $f = 70 \text{ MHz}$ varies by about 10 dB/cm . The obtained attenuation values of ultrasonic waves must be taken into account when designing AO devices for the mid- and long-wavelength IR ranges based on tellurium glasses.

Funding

The research was supported by a financial grant provided by the Russian Science Foundation (RSF) (№ 19-12-00072).

Conflict of interest

The author declares that he has no conflict of interest.

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