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Photoelastic and acousto-optic effects in $65GeS_2$-$25Ga_2S_3$-10CsCl glass

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ABSTRACT

The peculiarities of photoelastic and acousto-optic effects are first tested in chalcohalide $65GeS_2$-$25Ga_2S_3$-10CsCl glass exploring the interferometry method. The determined piezo-optic coefficients occur to be (in Br) $\pi_{11} = 2.8 \pm 0.6$, $\pi_{12} = 6.0 \pm 0.9$ and $\pi_{44} = -3.2 \pm 1.1$. In respect to maximal acousto-optic figure of merit $M_2 = 55.6 \cdot 10^{-15}$ s^3/kg proper for longitudinal acoustic wave propagation and polarization along X_2 ($i = 2$) direction, this glass seems more perspective for photoelastic and acousto-optic modulation in visible and near IR ranges, than many alternative candidates, such as crystalline quartz, $LiNbO_3$, $CaWO_4$, $PbMoO_4$, β-BaB_2O_4, etc. For the first time it has been shown that piezo-optic effect of isotropic solids is anisotropic, and the degree of anisotropy is the difference of π_{11}–π_{12} coefficients.

1. Introduction

Among a large group of functional media exploring IR spectral region, the special glasses based on chalcogenides, i.e. non-oxide compounds of chalcogens S, Se and/or Te prepared by quenching from a melt, which are often referred to as *chalcogenide glasses*, are in a sphere of tight interests for a great number of scientists in view of their promising implementation in functional optics (photonics, optoelectronics, telecommunication and information technologies, sensing electronics, etc.) [1–6].

Near two decades ago, it was shown that principal IR functionality of these glasses covering both telecommunication windows (3–5 and 8–12 μm) up to space telecommunication domain (20–25 μm) could be conveniently shared with high transmittance of halides in a visible range [7–9]. New class of disordered materials viz. mixed chalcohalide glasses (ChHGs) and glass-ceramics was emerged, thus comprising advantages of both chalcogenide and halide materials platforms [9,10].

This class of materials can be well exemplified, in part, by glassy GeS_2-Ga_2S_3-CsCl system with one of most typical representative of $65GeS_2$-$25Ga_2S_3$-10CsCl chemical composition [11–15]. Main optical properties of this ChHG (short-wave optical transmission edge, long-wave optical transmittance cut-off, wavelength dispersion of refractive index, chromatic dispersion, etc.) were studied in details elsewhere [11–14]. Good transmittance in a visible spectral range in these ChHGs is worth to be highlighted, along with their ability to be protected against atmospheric aggression by anti-reflecting ZnS coating, and excellent technological applicability in different macrooptic performances due to highly reproducible molding route [13,14].

Such promising realization of chalcohalide platform serves as a basis for wider practical using of these glassy-like media, especially in controllable light-guiding acousto-optic systems [16–23]. In this respect, the changes in optical properties of the ChHGs (refractive index, birefringence) under mechanical strain (piezo- and elasto-optic effects), allowing determination their acousto-optic efficiency, are of high importance.

In this work, the magnitudes of piezo-optic coefficients (POCs) were first measured in $65GeS_2$-$25GaS_3$-10CsCl ChHG by interferometry method. The elasto-optic coefficients (EOCs) were calculated at the basis of POCs, the modulation parameter (which describes the change of optical path per sample's thickness and mechanical stress) was found, and thus the acousto-optic efficiency of this ChHG was estimated. The magnitudes of refractive indexes, elastic constants and acoustic velocities were also determined.

2. Experimental

The ChHGs of $65GeS_2$-$25Ga_2S_3$-10CsCl composition (transparent in a range from ∼0.5 μm to 11 μm [13,14]) were prepared from high-

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purity raw materials (elemental Ge, Ga, S and CsCl of 5 N purity) using conventional melt-quenching route as was described elsewhere [12–14]. The purified ingredients weighed in stoichiometric proportions were inserted in a silica ampoule under 10^{-4} Pa vacuum. The sealed ampoule was placed in a rocking furnace for several hours at 850 °C and quenched in room-temperature water. The polished (λ/4) cubic sample (7 × 7 × 7 mm) was cut from the resulting ingot, this sample being annealed 10 °C below the glass transition temperature (T_g = 405 °C) for 4 h to reduce residual mechanical stress induced during the quench.

The optically isotropic materials like glasses are known to possess only two independent piezo-optic coefficients π_{im} (π_{11} and π_{12}), which can be measured by interferometry method of the half-wave (control) stress with the Mach-Zehnder interferometer set-up [24]. By using relation for difference in optical path lengths $\delta\Delta_k$ in the interferometer arm with studied sample under mechanical stress σ_m in the form of

$$\delta\Delta_k = \delta(n_i d_k) = -\frac{1}{2}\pi_{im}\sigma_m d_k n_i^3 + S_{km}\sigma_m d_k(n_i - 1), \tag{1}$$

the magnitudes of POCs π_{im} can be calculated from experimental half-wave stresses σ_{im} under condition of $\delta\Delta_k = \lambda/2$, $\sigma_m = \sigma_{im}$ [24,25]:

$$\pi_{im} = -\frac{\lambda}{\sigma_{im} n_i^3 d_k} + \frac{2S_{km}}{n_i^3}(n_i - 1) = -\frac{\lambda}{\sigma_{im}^{o} n_i^3} + \frac{2S_{km}}{n_i^3}(n_i - 1); \tag{2}$$

where $n_i = n$ is refractive index of isotropic sample, d_k is sample's thickness in light propagation direction, S_{km} is elastic compliance coefficient, $\sigma_{im}{}^{o} = \sigma_{im} d_k$ is control stress; the k, i, m indexes denote light propagation, polarization and uniaxial pressure directions, respectively.

The refractive index $n_i = n$ was determined by the interferometry-turning method using set-up at the basis of Michelson interferometer [26]. The longitudinal and transversal ultrasonic velocities V_{11} and V_{12} needed to obtain the coefficients of elastic stiffness C_{mn} and elastic compliance S_{km} were determined by impulse Papadakis method [27].

3. Results and discussion

3.1. POCs π_{11} and π_{12} determination

The value of refractive index in the studied $65GeS_2$-$25Ga_2S_3$-10CsCl glass at λ = 632.8 nm wavelength determined using the Michelson interferometer set-up occurred to be $n = 2.08 \pm 0.01$. The elastic stiffness coefficients C_{11} and C_{12} were defined as [28]

$$C_{11} = \rho V_{11}^2, \tag{3}$$

$$C_{11} - C_{12} = 2\rho V_{12}^2. \tag{4}$$

The magnitudes of these coefficients C_{11} = 24.4 GPa and C_{12} = 8.35 GPa were obtained using preliminary values of glass density $\rho = 2.92 \cdot 10^3$ kg/m^3 defined by hydrostatic weighting in toluol and ultrasonic velocities V_{11} = 2890 m/s and V_{12} = 1658 m/s for longitudinal and transversal acoustic waves, respectively, determined by impulse Papadakis method [27].

The magnitudes of elastic compliance S_{11} and S_{12} coefficients were obtained as inverse components of elastic stiffness coefficients matrix $S = C^{-1}$:

$$S_{11} = \frac{C_{11} + C_{12}}{C_{11}^2 + C_{11}C_{12} - 2C_{12}^2}, \tag{5}$$

$$S_{12} = -\frac{C_{12}}{C_{11}^2 + C_{11}C_{12} - 2C_{12}^2}. \tag{6}$$

With above C_{11} and C_{12} coefficients, the $S_{11} = 49.6 \cdot 10^{-12}$ m^2/N and $S_{12} = -12.7 \cdot 10^{-12}$ m^2/N values were obtained. The typically estimated errors do not exceed 0.3% in acoustic waves (V_{11} and V_{12}) and 2% in elastic coefficients (C_{mn} and S_{km}). Following to our previous works [29–32], we accept that realistically the latter could be somewhat greater (near 5%) due to possible inhomogeneities in mechanical stress.

Two independent control stresses $\sigma_{11}{}^{o}$ and $\sigma_{12}{}^{o}$ defined experimentally with typical errors no more than 10% were + 12.2 and + 7.9 kG/cm, respectively.

Using Eq. (2) with control stresses ($\sigma_{11}{}^{o}$ and $\sigma_{12}{}^{o}$) and elastic coefficient S_{12}, the magnitudes of two independent POCs can be presented (in Br units, 1 Br = 1 Brewster = 10^{-12} m^2/N) as $\pi_{11} = +2.8 \pm 0.6$ (21%) and $\pi_{12} = +6.0 \pm 0.9$ (15%). The errors in POCs were found as mean square values from first and second components in Eq. (2) by assuming that error in the first component is formed by 10% error in the control stress $\sigma_{im}{}^{o}$, and error in the second component is formed by 5% error in the elastic coefficient S_{12}.

Noteworthy, the $65GeS_2$-$25Ga_2S_3$-10CsCl glass overcomes by POCs magnitudes many known acousto-optic media. Thus, the maximal magnitudes of principal POCs π_{im} (i, m = 1, 2, 3) are known to be from 0.66 to 0.8 Br for lithium niobate $LiNbO_3$ [32,33], 1.44 Br for gallium phosphide GaP [30], 1.86 Br for calcium wolframate $CaWO_4$ [29,34], 3.11 Br for crystalline quartz [35], 3.34 Br for lead molybdate $PbMoO_4$ [36], 3.7 Br for barium β-borate β-BaB_2O_4 [37,38]. The high magnitude was also obtained for rotation-shift coefficient $\pi_{44} = -(3.2 \pm 1.1)$ Br defined as $\pi_{44} = \pi_{11} - \pi_{12}$ in respect to POCs matrix of isotropic solids [39,40].

Let's estimate the photoelastic efficiency of this glass due to $\delta\Delta_k/(d_k\sigma_m)$ parameter, which describes the change in the optical path per sample's thickness d_k and mechanical stress unit σ_m. This parameter introduced in [30,32] can be presented at the basis of Eq. (1) as

$$\frac{\delta\Delta_k}{d_k\sigma_m} = -\frac{1}{2}\pi_{im} n_i^3 + S_{km}(n_i - 1). \tag{7}$$

For real experimental conditions $m = 1$, $k = 2$, $i = 1$, and change in optical path is formed by $\pi_{im} = \pi_{11}$ and $S_{km} = S_{12}$ coefficients. Respectively, it is found that:

$$\frac{\delta\Delta_2}{d_2\sigma_1} = -\frac{1}{2}\pi_{11} n_i^3 + S_{12}(n_1 - 1) = -12.6(48\%) - 13.7(52\%) = -26.3\,\text{Br}. \tag{8}$$

The larger value of this parameter is character for $m = 2$, $k = 3$, $i = 1$ (corresponding to $\pi_{im} = \pi_{12}$, $S_{km} = S_{32} = S_{12}$):

$$\frac{\delta\Delta_3}{d_3\sigma_2} = -\frac{1}{2}\pi_{12} n_i^3 + S_{12}(n_1 - 1) = -27.0(66\%) - 13.7(34\%) = -40.7\,\text{Br}. \tag{9}$$

In respect to calculated $\delta\Delta_k/(d_k\sigma_m)$ parameters, the studied $65GeS_2$-$25Ga_2S_3$-10CsCl ChHG is better than such known crystalline photoelastic materials as GaP and $PbMoO_4$, possessing maximal values of these parameters (+ 20.3 and − 24.9 Br, respectively [30,36]). So this glass belongs to better photoelastic materials in view of POCs magnitudes and high values of modulation parameter $\delta\Delta_k/(d_k\sigma_m)$.

Note that percentages in parenthesis in Eqs. (8) and (9) correspond to inputs into overall effect from piezo-optic (first component) and elastic (second component) contributions. The piezo-optic contribution in Eq. (9) is twice greater than the elastic one. The negative magnitudes in Eqs. (8) and (9) mean that optical path $\delta\Delta_k$ decreases under positive (expansion) mechanical stress.

3.2. EOCs p_{11} and p_{12} determination

The p_{11} and p_{12} EOCs are calculated at the basis of experimental POCs (π_{11} and π_{12}) and elastic stiffness coefficients C_{11} and C_{12}, using known tensor expression $p_{in} = \pi_{im}C_{mn}$ [39–41], which can be written for isotropic media as

$$p_{11} = \pi_{11}C_{11} + \pi_{12}C_{21} + \pi_{13}C_{31} = \pi_{11}C_{11} + 2\pi_{12}C_{12}, \tag{10}$$

$$p_{12} = \pi_{11}C_{12} + \pi_{12}C_{22} + \pi_{13}C_{32} = \pi_{11}C_{12} + \pi_{12}(C_{11} + C_{12}). \tag{11}$$