On the optical nonlinearity in the GeSbSe chalcogenide glasses

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Abstract
Chalcogenide glasses Ge$_x$Sb$_{40-x}$Se$_{60}$ with composition 12, 25 and 30 at.%, synthesized from elements with 5N purity (Ge, Sb, Se) by the conventional melt–quenching method were studied. The spectrophotometer’s measurements were recorded to determine the optical properties. Models based on the absorbance and absorption coefficient are used to determine the optical band-gap energy. The Urbach energy is deduced in the Ge$_x$Sb$_{40-x}$Se$_{60}$ thin film alloys with composition $x = 12, 25$ and $30$ at.%. The electronic and structural properties are studied, and the influence of the Urbach energy in estimating of the nonlinearity in the optical properties of these glasses is discussed. The analysis is performed with respect to the Urbach parameter that quantifies the degree of crystallinity of the structure which can be used to understand the behavior of different diffractive, wave–transmission and fiber structures.

1. Introduction

More recently, ternary chalcogenide GeSbSe glasses were intensively studied and proposed for the development of optoelectronic devices due to their thermal, mechanical and chemical properties [1]. The melting and evaporation properties of these materials depend to a very large extent on the completeness of the chemical reaction. By varying the chemical composition of GeSbSe thin films, the network undergoes structural changes reflected in the structural phase transformations [2–4], in the effect of doping Sb on the electronic structure [5] and in the threshold transition in the physical parameters [6]. The presence of Sb and Se have strengthened in the material generated stable SbSe bonds and cross-linkages in GeSe–SbSe structural chains [7–9] with a material very versatile informing.

This paper aims to evaluate the optical constants of Ge$_x$Sb$_{40-x}$Se$_{60}$ with composition $x = 12, 25$ and $30$ at% deposited on quartz substrates held at room temperatures such as refractive index $n$, extinction coefficient $k$, absorption coefficient $\alpha$, optical band gap and the width of the tail of localized states in the band gap. The samples Ge$_x$Sb$_{40-x}$Se$_{60}$ with composition $x = 12, 25$ and $30$ at% were measured by Lambda 950 Spectrophotometer. Using the UV/Vis spectra the Urbach energy was detected. The influence of the Urbach energy in estimating of nonlinear optical properties of these glasses is discussed in this paper. The Urbach edge arises from fluctuations of short-distance forces in chalcogenides. The weak tail that appears in the absorption diagram can be described by antibonding states of wrong connections Ge–Ge or Se–Se that produce unoccupied states in the conduction band. Electronic excitations generate holes at the edge of the valence band and electrons in the conduction band, or holes in the valence band and electrons in the tail of the conduction band.

In amorphous materials, due to the absence of long-range forces, the valence and conduction bands do not have sharp cut off but have tails of localized states. Due to these band tails, the optical absorption edge is not sharp and has a tail at lower energies. When we make the Tauc plot we do not get a straight line in the whole energy range. Urbach energy has an inverse relation with enhancement in crystalline structure or with texture factor. The Urbach energy indicates the degree of crystallinity of the structure and induces nonlinearity in the optical properties of GeSbSe. These glasses have highly optically nonlinearities which are mainly reflected in devices such as optical switching. Chalcogenide layers can be obtained by different deposition methods.
2. Experimental details and results

Layers thin films Ge$_x$Sb$_{40-x}$Se$_{60}$ with composition $x = 12, 25$ and 30 at.%, were synthesized from elements with 5N purity (Ge, Sb, Se) by conventional melt-quenching method. The mixture of the elements with proper weight percent was placed in a quartz ampoule and evacuated down to a pressure of 10$^{-3}$ Pa. The ampoules were loaded in a rotary furnace and heated up to 950 $^\circ$C.

For a homogeneous melting, the glassy mixture was kept at this temperature for continuously two hours by rotating the furnace. Then, the ampoules were pulled out, and the melts were rapidly cooled down in the ice water. The parts of synthesized glasses were served as parent material for deposition on the quartz substrate using a resistive crucible of 0.1mm molybdenum sheet from having the shape and size of the 0482070 from the UMICORE catalog. The degassing took place at a pressure of 10$^{-5}$N purity water. The parts of synthesized glasses were served as parent material for deposition on the quartz substrate using a resistive crucible of 0.1mm molybdenum sheet from having the shape and size of the 0482070 from the UMICORE catalog. The degassing took place at a pressure of 10$^{-5}$N purity water.

The powder material was evaporated at an electric current at least 400 mA. The optical monitoring control was done by TFCalc 3.5 software for approximately 6 min with a deposition rate of 3 nm s$^{-1}$ with 10 rot min$^{-1}$. This process was removing the impurities from the walls from the vacuum deposition installation and on the bulk. At the 4 $\times$ 10$^{-3}$ Pa pressure, the heating resistance is coupled, by ensuring the temperature of 300 $^\circ$C in the evaporation chamber while the dome rotation was kept.

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The optical properties of materials depend on parameters as the preparation technique and the cleaner surface and also the preparative conditions for surface morphology. The study of the spectral absorption coefficient gives information about the electronic states in the high energy part of the optical absorption spectra.

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Absorptance is light that is not transmitted or reflected by material but is absorbed. Having the transmission spectra and absorptance, the equation describes the theory$^{[11-13]}$ can be used to calculate the reflectance

$$ T + R + A = 1, \quad (1) $$

where $T$ is transmittance, $R$ the reflectance, and $A$ the absorptance.

The results obtained by chalcogenide layers Ge$_x$Sb$_{40-x}$Se$_{60}$ alloys with composition $x = 12, 25$ and 30 at.% are present in figure 2.

The thickness of the chalcogenide films Ge$_x$Sb$_{40-x}$Se$_{60}$ alloys with composition $x = 12, 25$ and 30 at.% was determinte from the envelope function transmission spectra using the Swanepoel method $^{[11]}$.

The optical properties of materials depend on parameters as the preparation technique and the cleaner surface and also the preparative conditions for surface morphology. The study of the spectral absorption coefficient gives information about the electronic states in the high energy part of the optical absorption spectrum, while the other lower energy part of the spectrum corresponds to the atomic vibrations $^{[11]}$. There are several applications based on optical absorption such as optoelectronics devices, sensors, display devices, solar cells.

The optical absorption spectra and the optical absorption coefficient ($\alpha$) should be studied here. The absorption coefficient $\alpha$ can be determined from the transmittance, $T(\%)$ and reflectance, $R(\%)$ by using the following formula $^{[12]}$

$$ \alpha(\lambda) = \frac{1}{d} \ln \left[ \frac{(1 - R)^2}{2T} + \left( \frac{(1 - R)^4}{4T^2} + R^2 \right)^{1/2} \right]. \quad (2) $$

where $d$ is the thickness, $T$, the transmittance and $R$, the reflectance. The dependence of the absorption coefficient, $\alpha(\lambda)$ on the photon energy $h\nu$ is represented in figure 3.

Figure 3 indicates the value of the absorption coefficient, the increase/decrease of the Ge/Sb content upon photon energy incident for all the samples of Ge$_x$Sb$_{40-x}$Se$_{60}$ alloys with composition $x = 12, 25$ and 30 at.%. The extinction coefficient is determined by (3) as shown in figure 4.

$$ k = \frac{\alpha(\lambda)\lambda}{4\pi}. \quad (3) $$

Refractive index can be obtained using the normal incidence reflectance according to Fresnel’s equations $^{[12, 13]}$ computed by relation (4):

$$ R = \frac{(n - 1)^2 + k^2}{(n + 1)^2 + k^2}. \quad (4) $$

Figure 5 shows the results obtained by the refractive index according to (4).
Figure 1. Spectral measurements by Lambda Spectro-photometer, (a) spectral transmission [10], (b) spectral absorptance. Reprinted from [10]. Copyright (2020), with permission from Elsevier.

Figure 2. Spectral reflectance.
The value of the refractive index and extinction coefficient decrease/increase with Ge/Sb content [14–17]. The optical band gap $E_g$ was derived assuming indirect transitions between the edge of the valence and conduction band. In this work, we have used six models to determine the optical band-gap energy in the Ge$_x$Sb$_{1-x}$Se$_{60}$ thin film alloys with composition $x = 12, 25$ and $30$ at.%, represented in figure 6. These models were based on the absorbance and absorption coefficient.

The optical energy was determined by extrapolation of the linear curve of $A$, $\sqrt{A}$, $\alpha$, $\sqrt{A\alpha}$, $\sqrt{A\phi}$, and $\sqrt{A\phi\alpha}$ as a function of the photon energy $h\nu$. The intersection of the linear region on the $h\nu$ axis gives $E_g$ (figure 6).

The variation of optical band-gaps of Ge$_x$Sb$_{1-x}$Se$_{60}$ thin films alloys with composition $x = 12, 25$ and $30$ at.%, as a function of Germanium content, is presented in figure 7. The figure shows that $E_g$ grows with increasing Germanium content.

Along the absorption coefficient curve and near the optical band edge there is an exponential part called Urbach tail. This exponential tail appears in the low crystalline, poorly crystalline, disordered and amorphous materials because these materials have localized states which extended in the band gap. In the low photon energy range, the spectral dependence of the absorption coefficient ($\alpha$) and photon energy $h\nu$ is known as Urbach empirical rule, which is given by the following equation [18]

Figure 3. Absorption coefficient.

Figure 4. Extinction coefficient.
where \( A \) is the absorptance, \( \alpha \), the absorption coefficient, \( \tilde{A} \), \( \tilde{\alpha} \) are constants and \( E_u \) is the Urbach energy. The Urbach energy was deducted by the curves of \( \ln A \) (figure 8) and \( \ln \alpha \) (figure 9) as a function of photon energy \( h\nu \).

The variation of Urbach energy of Ge\(_{1-x}\)Sb\(_x\)Se thin films alloys with composition \( x = 12, 25 \) and 30 at.\% as a function of Germanium content is presented in the figure 10. The figure shows that \( E_u \) grows with increasing in Germanium content.

Wemple-DiDomenico (WD) model is using to calculate various dispersion parameters such as band-gap energy \( (E_g) \) oscillator energy \( (E_0) \) and dispersion energy \( (E_d) \) \([16, 19, 20]\). Accordingly, a graph (figure 5) is constructed with \( n\nu \) against \( h\nu^2 \). According to the single oscillator model, the data other dispersion of the refractive index can be evaluated using the equation:

\[
n^2 = 1 + \frac{E_0 E_d}{E_0^2} - h\nu^2, \tag{6}
\]

where \( n \) is the refractive index, \( E_0 \) is oscillator energy and \( E_d \) the dispersion energy.

Oscillator energy \( E_0 \) is calculated from the slope \( 1/E_0 E_d \) and dispersion energy \( E_d \) is calculated from the intercept \( E_0/E_d \) on y axis figure 11. The various dispersion parameters are also given in table 1, \( E_g \) value is an average between the model based on the absorptance and absorption coefficient, as a function of the photon energy \( h\nu \).

While the band gap energies \( E_g \) and \( E_0 \) increase, the \( E_d \) values show a tendency to decrease by increasing the Ge content, correspondingly decreasing the Sb (figure 12). This indicates the strong influence of the character and amount of chemical bonds on the GeSbSe material parameters.

### 3. Estimation of the dielectric properties

The GeSbSe, like all chalcogenides in general, are characterized by important fluctuations of the atomic distances in material as suggested in figure 13(a). The peaks in interatomic distances for Ge-Sb, Ge-Ge, Sb-Sb, and Se-Se atoms appear to be 3.80 ± 0.05 Å, 3.85 ± 0.05 Å, 3.80 ± 0.05 Å, and 3.80 ± 0.05 Å, respectively [16]. The atomic structure of GeSbSe is presented in figure 13(b).

These fluctuations are determined by the forces of a small length of action presented in the amorphous material. The response of glasses to electric fields or other external forces within an experimentally possible length and time scale exceeds the domain of applicability of a classical theory, because it depends on the ratio \( \lambda/l \), \( \tau/\tau_0 \) or \( \lambda/l \), \( \omega_0/\omega \), where \( \lambda \) is a characteristic length of the body (atomic distance, granular distance, etc), \( l \) is the external characteristic length associated with the external forces (waves, distances over which load distribution change sharply, geometrical and surface discontinuities), \( \tau \) the time scale (or frequency \( \omega \)) which is the minimum transmission time of a signal (or a frequency), and \( \tau_0 \) is the external characteristic time or frequency) associated with the external forces \([21–23]\). For \( \lambda/l \ll 1 \) and \( \tau/\tau_0 \ll 1 \), the external forces excite
large numbers of subbodies simultaneously, so that subbodies interact and the result is a statistical average of the individual responses. For $\lambda l = 1$ and $\tau \eta_0 = 1$, the individual fields of subbodies (intermolecular and atomic forces) are important.

Forces with a short domain of action that characterize the structure of chalcogenides are identified by the distribution functions of the pairs of Ge-Se and Sb-Se atoms that can be performed using a 3D simulation in a configuration with a given number of atoms. A simulation expectation was realized in [24] for ternary films of Ge$_{15}$Sb$_{25}$Se$_{60}$ and Ge$_{35}$Sb$_5$Se$_{60}$ with a configuration of 200 atoms. The results are obtained from x-ray and neutron diffraction correlated with the Reverse Monte Carlo simulations in binary and ternary films indicate that Sb atoms incorporated in Ge$_{40}$Se$_{60}$ are covalently bonded to Se and form trigonal Sb-Se units.

The results are shown in figure 14. The distribution functions of the pairs of atoms are evidence of the presence of short-distance bonds in the amorphous material. By increasing the Ge concentration, respectively decreasing the Sb concentration, the interatomic distances in the Ge-Se bonds increase from 2.37 Å to 2.40 Å.

Figure 6. The plot of $A$ (a), $\sqrt{A}$ (b), $\alpha$ (c), $\sqrt{\alpha}$ (d) $\sqrt{\Delta h\nu}$ (e) and $\sqrt{\alpha h\nu}$ (f) versus $(h\nu)$ in Ge$_{x}$Sb$_{1-x}$Se$_{60}$ thin films alloys with composition $x = 12$, 25 and 30 at. %.

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4. Estimation of dielectric properties

Along the absorption coefficient curve and near the optical band edge there is an exponential part called Urbach tail. This exponential tail appears in amorphous materials because these materials have localized states which extended in the band gap. In the low photon energy range, the spectral dependence of the absorption coefficient ($\alpha$) and photon energy ($\nu$) is known as Urbach empirical rule, which is given by

$$\alpha = \alpha_0 \exp(h\nu/E_u),$$

where $\alpha_0$ is a constant, $h$ the Planck’s constant, $\nu$ photon’s frequency and $E_u$ the energy of the band tail or the Urbach energy, which is weakly dependent upon temperature and is often interpreted as the width of the band tail due to localized states in the normal band gap that is associated with the disordered or low crystalline materials.

Urbach energy has an inverse relation with enhancement in crystalline structure or with texture factor. So, Urbach energy is a key factor in characterizing the internal structure of GeSbSe. Therefore, the properties of GeSbSe depend strongly on the Urbach energy.

The optical absorption edge in amorphous semiconductors is generally not as steep as that in crystalline semiconductors. In general, the absorption spectrum can be divided into three parts [24] (see figure 6(c)). For $\alpha \geq 2 \times 10^4$ cm$^{-1}$, the spectrum shows a linear dependence $\alpha h\nu \approx (h\nu - E_g)$. For $2 \times 10^4 \geq \alpha \geq 10^9$ cm$^{-1}$, the Urbach edge has the form of $\alpha \approx \exp(h\nu/E_u)$. For $\alpha \lesssim 10^6$ cm$^{-1}$, a weak-absorption tail of the form $\alpha \approx \exp(h\nu/pE_u)$, $p \geq 2$, appear. We refer to the parameter $p$ as to the Urbach parameter.

The linear and nonlinear dielectric constants are evaluated with respect to $p$. The linear dielectric constants are the dielectric constant $\varepsilon_1 = \varepsilon_0 = n^2 - k^2$ and the optical susceptibility $\varepsilon_2 = \chi_2 = n^2 - 1$, where $n$ is the refractive index. The nonlinear dielectric constant is the third-order optical susceptibility $\varepsilon_3 = \chi_3$ defined by the third harmonic generation that cannot be experimentally predicted [25, 26].

For estimating theoretically the dielectric constants, the theory of surface tension of Hilliard is applied [27, 28]. The local energy calculated from the interaction of atoms is the predominant term in the atomic pseudopotentials of Ge, Sb and Se atoms.

The distribution functions allow the representation of the nearest neighbours of each atom in a given reference system, necessary to calculate the energy for an amorphous material as the GeSbSe glass. Good identification of the distribution functions of the pairs of Ge-Se and Sb-Se atoms can be done by 3D simulations of a small configuration of 200 atoms in the glass [25].

The components $D_i, i = 1, 2, 3$ of the dielectric displacement vector are described in terms of the components $\varepsilon_{ij}, i, j = 1, 2, 3$ of the dielectric constant tensor, and the components $E_j, j = 1, 2, 3$, of the electric field vector $E$ in an arbitrary coordinate system, in a linear theory

$$D_i(\lambda)/\varepsilon_0 = \varepsilon_{ij}(\lambda)E_j,$$

with $\varepsilon_0 \approx 8.854 \times 10^{-12}$ farad/m is [10, 25–27]. This nonlinearity can be described by non-dispersive quadratic and cubic laws [10, 29]. The dielectric displacements $D_i, i = 1, 2, 3$ are
Figure 8. The variation of Urbach energy deduced by ln $A$ of (a) Ge$_{12}$Sb$_{28}$Se$_{60}$, (b) Ge$_{25}$Sb$_{15}$Se$_{60}$, (c) Ge$_{30}$Sb$_{10}$Se$_{60}$, (d) Ge$_x$Sb$_{40-x}$Se$_{60}$ ($x = 12, 25$ and $30$ at. %) thin film as a function of photon energy $h\nu$. 

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Figure 9. The variation of Urbach energy deduced by \( \ln \alpha \) of (a) \( \text{Ge}_{12}\text{Sb}_{28}\text{Se}_{60} \), (b) \( \text{Ge}_{25}\text{Sb}_{15}\text{Se}_{60} \), (c) \( \text{Ge}_{30}\text{Sb}_{10}\text{Se}_{60} \), (d) \( \text{Ge}_{x}\text{Sb}_{40-x}\text{Se}_{60} \) \((x = 12, 25, 30 \text{ at.\%})\) thin film as a function of photon energy \( h\nu \).
The pseudopotential energy per unit volume $W$ is

$$W = \frac{V}{\Omega} = D_l E_k \delta_{ik},$$

and

$$D_l / \varepsilon_0 = \varepsilon_{ij} E_j + \frac{1}{2!} \varepsilon_{ijkl} E_k E_l + \frac{1}{3!} \varepsilon_{ijklm} E_k E_l E_m + \ldots$$

The pseudopotential energy per unit volume $W$ is

$V = D_l E_k \delta_{ik},$
where $V$ is the pseudopotential energy, and $\Omega$ the cell volume. From (9) and (10), $V$ we have

$$V = D_i E_i \delta_{in} + \frac{1}{2!} \varepsilon_{ij} E_j E_i + \frac{1}{3!} \varepsilon_{ijkl} E_k E_l E_i + \frac{1}{4!} \varepsilon_{ijklm} E_m E_n E_l E_i + \ldots$$  \hspace{1cm} (11)
The constants are determined from (11) as

\[ \varepsilon_{ij} = \frac{1}{2!} \frac{\partial^2 V}{\partial \alpha_i \partial \alpha_j} \bigg|_{\alpha = 0}, \quad \varepsilon_{ikl} = \frac{1}{3!} \frac{\partial^3 V}{\partial \alpha_i \partial \alpha_k \partial \alpha_l} \bigg|_{\alpha = 0}, \]

\[ \varepsilon_{iklm} = \frac{1}{4!} \frac{\partial^4 V}{\partial \alpha_i \partial \alpha_k \partial \alpha_l \partial \alpha_m} \bigg|_{\alpha = 0} \ldots \]  \hspace{1cm} (12)

For 1D case, (9) becomes

\[ D / \varepsilon_0 = \varepsilon_1^d E + \varepsilon_2^d E^2 + \varepsilon_3^d E^3, \]  \hspace{1cm} (13)

where \( \varepsilon_1^d = \varepsilon_0 = n^2 - k^2 \) is the linear dielectric constant, \( \varepsilon_2^d = \chi_2 = n^2 - 1 \), is the linear optical susceptibility, \( n \) is the refractive index and \( \varepsilon_3^d = \chi_3 \) is the third-order optical susceptibility measured by the third harmonic generation [28].

In order to estimate \( \varepsilon_{ij}, i, j = 1, 2, 3, \varepsilon_{ikl}, i, k, l = 1, 2, 3, \varepsilon_{iklm}, i, k, l, m = 1, 2, 3, V \) is computed by using the atomic pseudopotentials of Ge, Sb and Se atoms, respectively. The atomic pseudopotentials represent the interactions between individual atoms \( i \) and \( j \) linked by atomic bonds \( b_{ij} \), \( i, j = 1, \ldots, n \). Here, \( n \) is the number of Ge, Sb and Se atoms in a representative cell of the GeSbSe glass.

The computing of \( V \) needs a unit cell with a known number of atoms and bonds. The cell can be built from the distribution functions of the pairs of atoms Ge-Sb, Ge-Se and Sb-S. An example of this cell is represented by 25 nearest neighbours of a green atom \( \alpha \) in the reference coordinate system \((X, Y, Z)\) for Ge\(_{12}\)Sb\(_{28}\)Se\(_{60}\), where \( \theta \) is the angle between Ge and Se bonds, and \( Z \) is the direction \((100)\) [10] (figure 15).

The model of Jankowski and Tsakalakos for the energy \( V_i \) of an atom \( i \) (the green atom) is adopted [27, 28]

\[ V_i = \frac{1}{2} \sum_{j \neq i} V_{ij}(0) = \frac{1}{2} \alpha \sum_n \exp(-\beta R_n^{(0)}), \]  \hspace{1cm} (14)

where \( V_{ij} \) is the potential between the atom \( i \) and its neighbour \( j \), and \( r_{ij} \) is the distance from atom \( i \) to the neighbour atom \( j \). \( \alpha \) is the repulsive energy parameter, and \( \beta \) is the repulsive range parameter which depends on \( b_{ij} \), \( i, j = 1, \ldots, n \). The sum \( \frac{1}{2} \alpha \sum_n \exp(-\beta R_n^{(0)}) \) is extended to the neighbours points \((X, Y, Z)\) located at distances \( R_n^{(0)} \), \( R = \sqrt{X^2 + Y^2 + Z^2} \) with respect to the green Se atom. In view of (14), \( V \) can be interpreted as a measure of the Born-Mayer repulsive energy. The parameter \( \alpha \) is measured in Ryd (Rydberg) 1 Ryd = 13.6 eV = 2.092 × 10\(^{-18}\) J, and the parameter \( \beta \) in atomic units [\( \text{au} \)]. Total atomic energy \( V \) is computed from

\[ V (r_1, r_2, \ldots, r_N) = \sum_i V_i, \]  \hspace{1cm} (15)

with respect to atomic coordinates \((r_1, r_2, \ldots, r_N)\). Forces on each atom are given by

\[ f_i = -\frac{\partial V (r_1, r_2, \ldots, r_N)}{\partial r_i}. \]  \hspace{1cm} (16)

The parameters \( \alpha \) and \( \beta \), the angle \( \theta \), and the atomic bonds \( b_{ij} \), \( i, j = 1, \ldots, n \) for Ge-Se, Se-Sb and Ge-Sb have computed from the condition of zero applied forces and zero imposed displacements on specific atoms in the equilibrium atomic state. These parameters depend on \( p \) as shown in table 2. The dielectric constants are determined from (15) with the following formulae with no summation over repeating indices.
The computing of $\frac{\partial \eta}{\partial E_j} = X_{ij}$ is based on the inverse square relationship between $E$ strength and $\eta$. The dielectric constants $\epsilon_i^d$, $\epsilon_i^d$ and $\epsilon_i^d$ are presented in tables 3 and 4 for $p = 1$ and $p = 2$ respectively. The difference in values appears to the second decimal place in all cases. It is noticeable that the values are lower for $p = 2$ (in absolute value).
5. Conclusions

The thin films $\text{Ge}_x\text{Sb}_{40-x}\text{Se}_{60}$ with composition $x = 12, 25$ and $30$ at.%, have been synthesized from elements with 5N purity (Ge, Sb, Se) by the conventional melt-quenching method. The mixture part of the elements served as parent material for deposition on the quartz substrate. Models based on the absorbance and absorption coefficient are used to determine the optical band-gap energy. Urbach energy is the primary means for study the nonlinear optical properties of the glasses. Novel optical switching, for example, is an application of chalcogenide glasses in which the optical nonlinearities is the key for the optimal functionality of the femtosecond switches. The paper proposes a theoretical formalism for evaluating the optical properties for $\text{Ge}_x\text{Sb}_{40-x}\text{Se}_{60}$ thin films with composition $x = 12, 25$ and $30$ at.%, with respect to a parameter which quantifies the Urbach energy. The algorithm addresses new results regarding the induced nonlinearities and the possibility to enhance them to be used in optical nonlinear devices.

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Table 4. Dielectric constants $\varepsilon_1$, $\varepsilon_2$ and $\varepsilon_3$ with respect to wavelength, for $p = 2$.

| $x$ | $\varepsilon_1$ [farad/m] | $\varepsilon_2$ [farad/volt] | $\varepsilon_3$ [farad m volt$^{-1}$] | Wavelength $\times 10^{-9}$[m] |
|-----|--------------------------|-----------------------------|-----------------------------------|-------------------------------|
| $x = 12$ at.% $p = 2$ | 1.31 | 6.30 | 3.87 | 200 |
| 9.20 | 1.90 | 3.84 | 400 |
| 13.05 | −6.31 | −0.70 | 600 |
| 10.61 | −13.41 | −1.32 | 800 |
| 9.86 | −18.33 | −18.61 | 1000 |
| 9.36 | −15.44 | −11.47 | 1200 |
| 9.12 | −2.42 | −4.06 | 1400 |
| 9.03 | 1.96 | 0.88 | 1600 |
| 9.00 | 3.40 | −2.42 | 1800 |
| $x = 25$ at.% $p = 2$ | 1.43 | 2.90 | 0.20 | 200 |
| 5.40 | −3.51 | −2.49 | 400 |
| 12.01 | −8.24 | −3.32 | 600 |
| 9.40 | −14.01 | −10.86 | 800 |
| 8.86 | −17.00 | −16.96 | 1000 |
| 8.38 | −14.02 | −12 | 1200 |
| 8.35 | −4.06 | −3 | 1400 |
| 8.14 | −4.00 | 5.88 | 1600 |
| 8.13 | −0.94 | 5.90 | 1800 |
| $x = 30$ at.% $p = 2$ | 6.20 | 6.32 | 2.05 | 200 |
| 6.00 | −0.60 | 2.46 | 400 |
| 9.86 | −6.60 | 2 | 600 |
| 8.13 | −12.34 | −6.13 | 800 |
| 7.81 | −14.82 | −14.20 | 1000 |
| 7.63 | −3.70 | −9.94 | 1200 |
| 7.37 | 1.23 | −4.03 | 1400 |
| 7.33 | −1.15 | 2.48 | 1600 |
| 7.30 | −4.11 | 2.49 | 1800 |
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