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Transmission spectra modeling for nanomultilayer chalcogenide thin films

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ABSTRACT

An curve-fitting method for accurate determination of optical constants and thickness is presented for chalcogenide glasses thin films. Optical constants and thickness of nanomultilayer chalcogenide structure $\text{As}_2\text{S}_3\text{:Mn-Se}$ were obtained by Swanepoel method from transmission spectrum using PARAV software. Proposed curve-fitting method is based on numerical simulation of transmission spectrum based on film parameters obtained by Swanepoel method and comparison one with experimental transmission spectrum. It is shown that modeled spectrum is not fully corresponding to the original measured spectrum due to inaccurate absorption coefficient determination in the region of Urbach absorption. Adjusting of absorption spectrum with further numerical simulation of spectrum allows achieving the best correspondence to original transmission spectrum. It solves the problem of the accurate absorption coefficient calculation in the different absorption regions. Finally, dependencies of the absorption coefficient on wavelength of the nanomultilayer chalcogenide films structures were obtained from transmission spectra using the proposed method.

Transmission spectra, numerical simulation, chalcogenide glasses, nanomultilayer structure, optical constants, absorption coefficient.

1. INTRODUCTION

Chalcogenide glasses (ChGs) are low-phonon-energy materials and are generally transparent from the visible up to the infrared. Numerous applications of active and passive optical devices based on thin ChGs films have been proposed¹. Over the past few decades the optical properties of ChGs films have been the subject of intense study². Refractive index of ChGs films and its wavelength dispersion are of critical importance for fabricating high-quality optical devices. Due to high sensitive to the absorption of electromagnetic radiation ChGs show a variety of photoinduced effects as a result of illumination³. Various models have been put forward to explain these effects, which can be used to fabricate diffractive, waveguide and fiber structures. Next-generation devices for photonic and related applications will rely on the development of materials which possess optimized physical properties that are compatible with thin film systems. This allows suitable integration to existing film-based applications, and hence requires appropriate consideration of material choice: its optical properties depending on refractive index and absorption coefficient. Nanomultilayer chalcogenide thin films (NML) provide possibility of direct surface relief formation and are perspective in many application areas (fabrication of optical elements, recording media for digital holography, etc.)⁴⁻⁸. Such nanomultilayer structures based on ChGs were first proposed by Kikineshi⁹. Moreover for the first time possibility of direct formation of magnetic relief simultaneous with surface relief formation under optical recording using $\text{As}_2\text{S}_3\text{:Mn-Se}$ multilayer nanostructures was shown¹⁰. From general point of view the optical properties of nanomultilayer structure can be approximated by “effective” media model. Exposure of NML composite structures by actinic light causes processes of surface relief formation and this circumstance determines importance of optical constants and thickness determination.

An accurate determination of optical constants of a solid films and nanomultilayer structures is important both from fundamental and technological viewpoints. The knowledge of the actual dependence of its optical constants on spectral wavelength (spectral dispersion) enables one to elucidate optical phenomena operative in it and its band-energy structure. Further, accurate values of optical constants of solid films are necessary for design and modeling of optoelectronic devices incorporating them. Most optical studies involve measurements on thin films or multi-layered structures comprising of numerous thin solid films made from different materials, with the ultimate aim to retrieve spectral dispersion of their optical parameters¹¹⁻¹³. One needs first to measure macroscopic optical quantities of a multi-layered structure correlated to the optical parameters of its layers and applicable theoretical formulas that typify the measured

macroscopic optical quantities are then required. A reliable approach is necessary to analyze the as-measured data of such macroscopic optical quantities in view of these respective theoretical formulas. To finalize optical analysis, dispersion function and absorption models that would properly describe the spectral behavior of the optical parameters of investigated films will be supportive.

In the present paper, we proposed new method to accurate determination of optical constants (refractive index and absorption coefficient) for ChGs nanomultilayer structure, based on $\text{As}_2\text{S}_3\text{:Mn}$ and Se. Firstly, transmission spectrum of studied structure was measured with further optical constants retrieval by envelope method, proposed by Swanepoel¹⁴. Secondly, numerical simulation of optical transmission spectrum was performed using obtained optical parameters of NML structure. Comparison of modeled spectrum with experimental one showed that they were not fully corresponding to each other due to inaccurate absorption coefficient determination. Therefore, more precise method is highly desired to determine the absorption coefficient of the chalcogenide thin films in the whole studied spectrum. We applied a curve-fitting method by numerical simulation of transmittance spectra that allowed to obtain the reliable values of absorption coefficient spectrum in whole studied optical region.

2. MATERIALS AND METHODS

The experiments were performed on 1.2 μm thick $\text{As}_2\text{S}_3\text{:Mn}$ -Se NML structure deposited on rotated glass substrate by thermal evaporation from two isolated boats with bulk $\text{As}_2\text{S}_3\text{:Mn}$ (2wt%) and Se glasses. The total number of nanolayers was 220, and a thickness of each $\text{As}_2\text{S}_3\text{:Mn}$ nanolayer was 5 nm and 6 nm of Se nanolayer correspondingly. More detailed description of deposition process can be found in paper¹⁵. The geometric thickness d of the films was monitored in-situ through evaporation by an interference sensor (at $\lambda=950$ nm), installed in the chamber of deposition system.

The normal-incidence transmittance $T_{\text{exp}}(\lambda)$ of obtained NML structure was measured at room temperature as a function of spectral wavelength λ in the range 500-900 nm. Transmittance measurements were performed using a double-beam SPECORD M40 spectrophotometer. Both of the $T_{\text{exp}}(\lambda)$ spectra of the studied air/NML structure/thick glass/air and air/thick glass/air samples were recorded relative to a corrected air-baseline, where the transmittance of air was recorded with both light-beam paths were free from any sample and then normalized to 100%. Transmittance measurements were then taken with the studied sample being placed in its light-beam path, leaving the reference light-beam path uncovered.

Transmittance spectrum $T(\lambda)$ of NML structure $\text{As}_2\text{S}_3\text{:Mn}$ -Se of 1.2 μm thickness is depicted in Fig. 1. Above a threshold wavelength corresponding to optical band gap energy $E_{g_{\text{opt}}}(\lambda \sim 645 \text{ nm})$, its as-measured $T(\lambda)$ curve displays transmittance value near that of glass substrate ($T=92\%$) and exhibits well-resolved interference-fringe maxima and minima, signifying good uniformity of fabricated chalcogenide nanomultilayers structure.

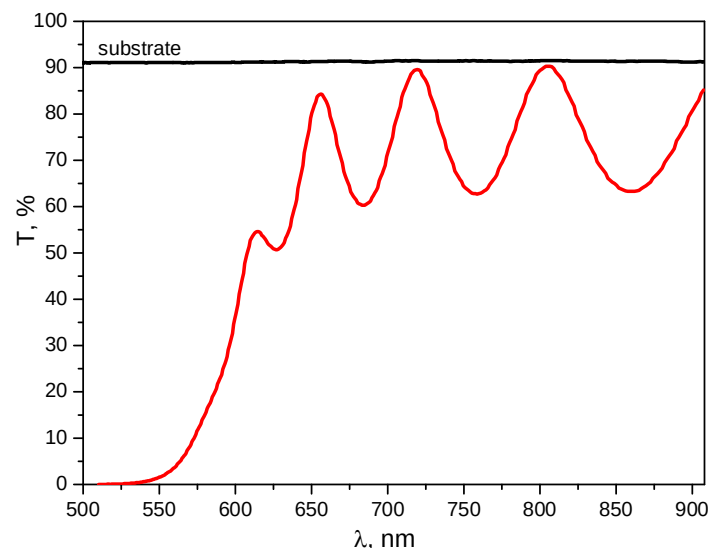


Fig. 1. Normal-incidence transmittance spectrum $T(\lambda)$ of nanomultilayer structure $\text{As}_2\text{S}_3\text{:Mn}$ -Se.

For a model solid film, i.e. linear, isotropic, homogeneous, dispersive, and uniform solid film with smooth and parallel-plane surfaces (an “ideal uniform” film), its optical properties can be completely determined by a complex index of refraction $\bar{n}(\lambda) = n(\lambda) + jk(\lambda)$, whose real part $n(\lambda)$ and imaginary part $k(\lambda)$ are, respectively, the normal index of refraction and extinction coefficient of the film, related to its absorption coefficient $\alpha(\lambda)$ as $k(\lambda) = \lambda\alpha(\lambda)/4\pi$. Therefore, in principle, two independent optical measurements are necessary at each spectral wavelength λ to solve for the unknown parameters $n(\lambda)$ and $k(\lambda)$ but, in practice, the true geometric thickness d of the film is not known in prior and is thus to be determined at the same time. So, there are three unknown parameters to be retrieved from two independent optical measurements at the same spectral wavelength, which demands adequate model suppositions or an otherwise optical analytical approach.

The most common, cost-effective, and non-destructive experimental techniques often used to measure macroscopic optical quantities of a solid film or layered structure composing of several films are the spectroscopic ellipsometry¹⁶ and spectrophotometry¹⁷, each with its own advantages, limitations, and drawbacks. The latter optical measuring technique, which we are concerned here, enables one to measure directly the macroscopic transmittance $T(\lambda)$ of a thin film or multi-layered structure over a wide spectral range of the light incident upon the structure¹⁸; however, the transmittance is not an intrinsic property of the film’s material and in general has no direct proportionality to its optical parameters; thus, further quantitative analysis of the $Texp(\lambda)$ spectra of the film or the structure including the film is needed to determine its geometric thickness as well as its optical constants $n(\lambda)$ and $k(\lambda)$ at each λ , and hence their dispersion functions and electronic transitions responsible for optical absorption in the film.

Many approaches, each with its own limits of applicability, were used to get geometric thickness and optical constants $n(\lambda)$ and $k(\lambda)$ of a solid film from its as-measured $Texp(\lambda)$ spectra¹⁹⁻²¹. The most common method named envelope algebraic methods, developed by Swanepoel¹³ and refined by a number of authors²²⁻²³, is based on approximate equations derived from reasonable assumptions, and is widely used to extract spectral dispersion of optical constants of a film and its geometric thickness from normal-incidence $T(\lambda)$ spectra of {thin film/thick substrate} structure, both of which do not require in prior a dispersion functional model. Using this envelope method, based on the presence of many interference-fringe maxima and minima on transmittance spectra in transparent and weak absorption regions of the film, the refractive index, thickness and absorption coefficient of the films only from their transmission spectrum could be obtained. But it cannot be applied to spectra displaying few interference fringes. This problem can be solved by curve-fitting method and demands multi-constant dispersion formulas that can be used to analyze transmittance of multi-layered structures, irrespective of the number of observed interference-fringe extrema.

3. RESULTS AND DISCUSSION

The spectral dependencies of optical constants $n(\lambda)$ and $\alpha(\lambda)$ of the studied NML structure on wavelength λ were retrieved from its experimental transmittance spectrum using software program PARAV (version 2.0)²⁴. The dispersion of refractive index, optical absorption coefficient, geometric thickness and optical bandgap, from experimentally measured transmission spectra of NML structure was obtained by this program. The retrieved $n(\lambda)$ spectrum was found to vary with wavelength in accordance with well-known Cauchy dispersion formula $n(\lambda) = \frac{A}{\lambda^2} + \frac{B}{\lambda^4} + \dots + C$, as depicted in Figure 2. Due to the small value of second term, it is sufficient to use a two-term form of this equation. It was obtained dispersion parameters $n(\lambda) = \frac{1.368 \times 10^5}{\lambda^2} + 2.195$, with a static index of refraction $n(\lambda \rightarrow \infty) = 2.195$.

To reveal physical features of observed optical absorption of the NML structure in present work, the dependency of respective absorption coefficient α was plotted against the photon energy $E = \hbar\omega$ and wavelength λ of light incident normally onto the front surface of the film, where ω is the light frequency and $\hbar$ is reduced Planck constant. In general, the optical absorption edge in amorphous semiconductors is generally not as steep as that in crystalline semiconductors, and can be divided into three parts. The relationship between the absorption coefficient and the photon energy in the region of strong absorption ($\alpha \geq 10^4 \text{ cm}^{-1}$) is given by Tauc and shows a square-root dependence $\alpha \hbar\omega \propto (\hbar\omega - E_g)^{1/2}$. For medium absorption ($10^4 \geq \alpha \geq 10^0 \text{ cm}^{-1}$), so-called Urbach edge with the form of $\alpha \propto \exp(\hbar\omega/E_U)$ appears. For weak absorption region ($\alpha \leq 10^0 \text{ cm}^{-1}$), $\alpha(E)$ shows another more gradual exponential spectrum $\alpha \propto \exp(\hbar\omega/E_w)$, which is referred to as a “weak absorption tail”. In the above, E_g represents the optical (Tauc) gap, E_U - Urbach energy.

Figure 3 illustrates the dependency of absorption coefficient α of investigated NML structure on the exciting photon energy E and wavelength λ in view of interband optical absorption models for disordered amorphous semiconducting samples. The regions of absorption coefficient in dependence on type of absorption are divided by dashed lines.

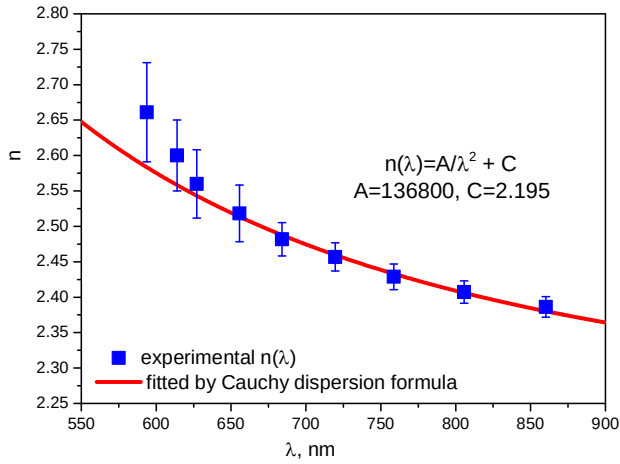


Fig. 2. Dependence of refractive index $n(\lambda)$ on wavelength λ for studied NML $\text{As}_2\text{S}_3\text{:Mn-Se}$. Symbols are data retrieved from PARAV software, and line – fitted dispersion spectrum using the Cauchy model.

The absorption coefficient in strong absorption region in many amorphous semiconductors is observed to obey Tauc relation $\alpha = \text{const} * (\hbar\omega - E_g)^{1/2} / \hbar\omega$. Therefore, plotting $(\alpha\hbar\omega)^{1/2}$ versus $\hbar\omega$ should result in a straight line. The optical gap E_g is obtained from the intersection of this line with the energy axis. The value obtained for the optical gap of NML structure $\text{As}_2\text{S}_3\text{:Mn-Se}$ was $E_g = 1.93 \pm 0.02$ eV. This value is slightly higher than that of 1.92 eV found for thermally evaporated Se films. It can be explained using the “effective” optical medium model: the layers with small optical band gap value determine the optical absorption at the average absorption edge, and the “barrier” layers with larger E_g are transparent.

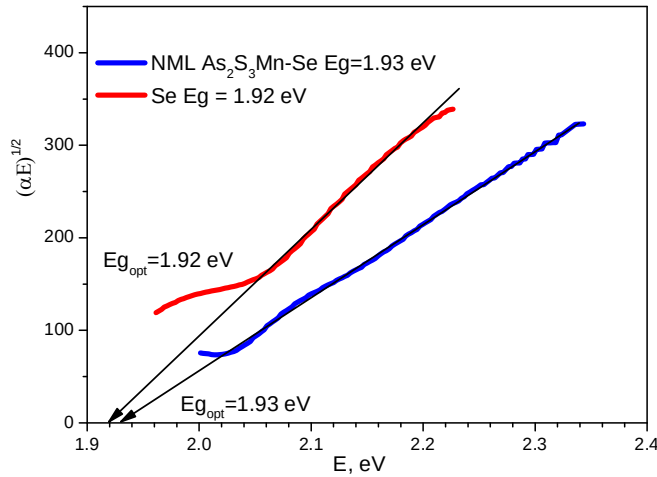


Fig. 4. $(\alpha\hbar\omega)^{1/2}$ versus $\hbar\omega$ plots for NML structure $\text{As}_2\text{S}_3\text{:Mn-Se}$ and thin Se film ($d=0.6 \mu\text{m}$). Least-squares fits (solid lines) of the linear parts on $(\alpha\hbar\omega)^{1/2} - \hbar\omega$ plots by intersection of which with the E -axis gives the respective Tauc optical bandgap energy E_g .

To elucidate more details on the dependency of the absorption coefficient in the sub-bandgap region for NML structure studied in the present work, logarithmic dependence $\ln(\alpha)$ vs E (Urbach-tail) was obtained (Fig. 5). The slope,

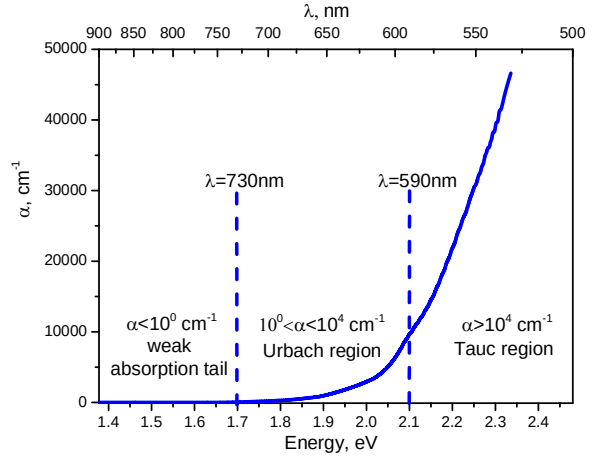


Fig. 3. Dependence of absorption coefficient α on photon energy E and wavelength λ for studied NML $\text{As}_2\text{S}_3\text{:Mn-Se}$, retrieved from PARAV software using its normal-incidence transmittance spectrum.

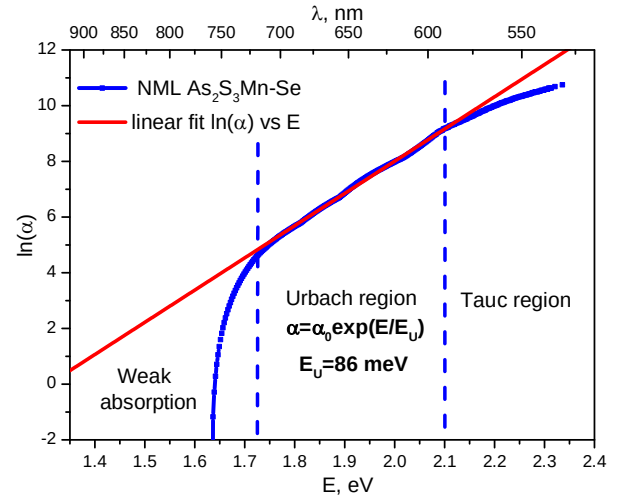


Fig. 5. $\ln(\alpha)$ versus E (Urbach-tail) plots for NML structure $\text{As}_2\text{S}_3\text{:Mn-Se}$. Slope of least-squares fits of the linear part of such semi-log plot gives Urbach energy E_U .

obtained from least-squares fitting of the straight line section of such semi-log plot, gives Urbach energy $E_U=86$ meV for the studied NML structure.

Obtained values of the Tauc and Urbach energies E_g and E_U correspondingly, which are all related to the film structure and material properties, suggest that the studied NML structure $\text{As}_2\text{S}_3:\text{Mn}-\text{Se}$ had almost same amorphous structure and structural disorder as a-Se films. Obtained Tauc bandgap energy E_g is in good agreement with E_g values reported for a-Se films by other authors, who employed a rather different optical analysis. But obtained E_U value for NML structure ($E_U=86\text{meV}$) is much higher as for a-Se films ($E_U\approx 40\text{meV}$), suggesting larger structural disorder in our fabricated nanomultilayer structure.

A common approach for verification of retrieved optical constants $n(\lambda)$ and $\alpha(\lambda)$ and their spectral dependencies is the indirect (inverse) synthesis method, which is based on modeling for transmittance spectrum $T(\lambda)$ inserting multi-constant dispersion function for $n(\lambda)$ and $\alpha(\lambda)$ into theoretical formulas of transmittance, and then use a curve-fitting method to compare of simulated spectrum and as-measured one²⁵. The dispersion functions for $n(\lambda)$ and $\alpha(\lambda)$ can be adjusted to achieve optimal correspondence between simulated curve $T(\lambda)$ and measured one $T_{\text{exp}}(\lambda)$. This curve-fitting procedure can be implemented manually or automatically in the sense that the parameters of the adopted dispersion model and other initially selected fitting parameters can be varied manually, by typing new values, or automatically by using such method known as the downhill simplex minimization technique.

First, we tried to model as-measured normal-incidence transmittance spectrum of our NML sample by known expression for uniform solid film with smooth and parallel-plane surfaces $T(\lambda)=Ax/(B-Cx\cos\varphi+Dx^2)$, where $A=16n^2s$, $B=(n+1)^3(n+s^2)$, $C=2(n^2-1)(n^2-s^2)$, $D=(n-1)^3(n-s^2)$, $\varphi=4\pi nd/\lambda$, $x=\exp(-ad)$, s – refractive index of glass substrate, d – NML thickness, $n(\lambda)$ and $\alpha(\lambda)$ – “effective” refractive index and absorption coefficient of NML structure respectively. The refractive index s of the substrate is acquired using the data from the transmission spectrum of the substrate alone $T_s(\lambda)$ by formula $s=1/T_s+(1/T_s^2-1)^{1/2}$.

The red curve in Fig. 6 is a plot of simulated transmittance spectrum $T(\lambda)$ as calculated by expression given above using the postulated properties of the NML structure $n(\lambda)$ and $\alpha(\lambda)$ obtained by PARAV software. For comparison by black line is shown the as-measured transmittance spectrum $T_{\text{exp}}(\lambda)$ and by green line the simulated transmittance spectrum $T(\lambda)$ using only $n(\lambda)$ and assuming that $\alpha(\lambda)=0$.

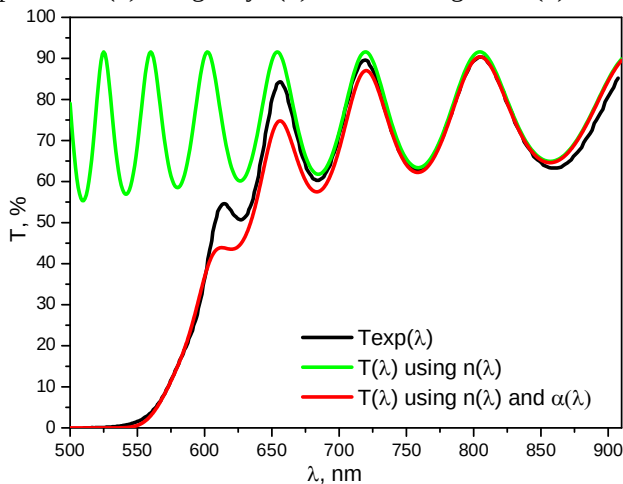


Fig. 6. As-measured transmittance spectrum $T_{\text{exp}}(\lambda)$ (black curve), simulated spectrum using only $n(\lambda)$ and assuming that $\alpha(\lambda)=0$ (green curve) and simulated spectrum using $n(\lambda)$ and $\alpha(\lambda)$ obtained by PARAV software (red curve).

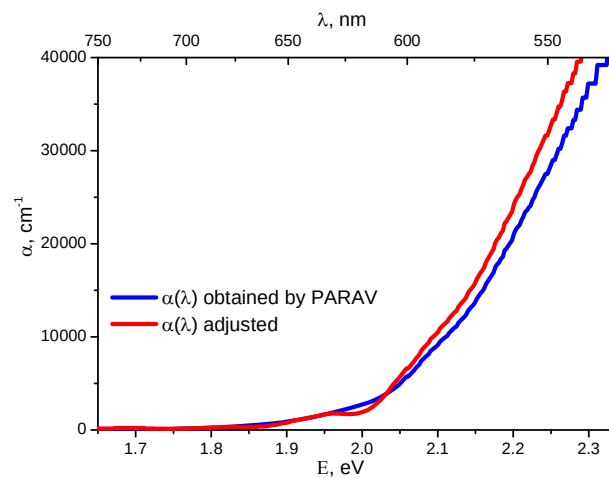


Fig. 7. Absorption coefficient spectrum $\alpha(\lambda)$ obtained by PARAV program (blue curve) and adjusted one $\alpha_{\text{adj}}(\lambda)$ (red curve).

As we can see from Fig. 6, simulated spectrum using only $n(\lambda)$ and assuming that $\alpha(\lambda)=0$ shows a good coincidence with experimental measured spectrum in weak absorption region 730-900 nm, that confirms accuracy of $n(\lambda)$ retrieval. But simulated spectrum using combination of obtained $n(\lambda)$ and $\alpha(\lambda)$ data gave bad fits for as-measured transmittance spectrum in the Urbach region. We believe that absorption coefficient $\alpha(\lambda)$ was not adequately obtained by PARAV software in the Urbach region. To obtain accurate and reliable data of optical properties absorption coefficient $\alpha(\lambda)$ was manually adjusted to achieve optimal consistency between theoretically simulated curve and measured one. Adjusted

value of absorption coefficient $\alpha_{adj}(\lambda)$ is shown in Fig. 7. The full coincidence of experimental and simulated spectra based on adjusted $\alpha_{adj}(\lambda)$ is shown in Fig. 8. The corresponding absorption regions are separated by dashed lines.

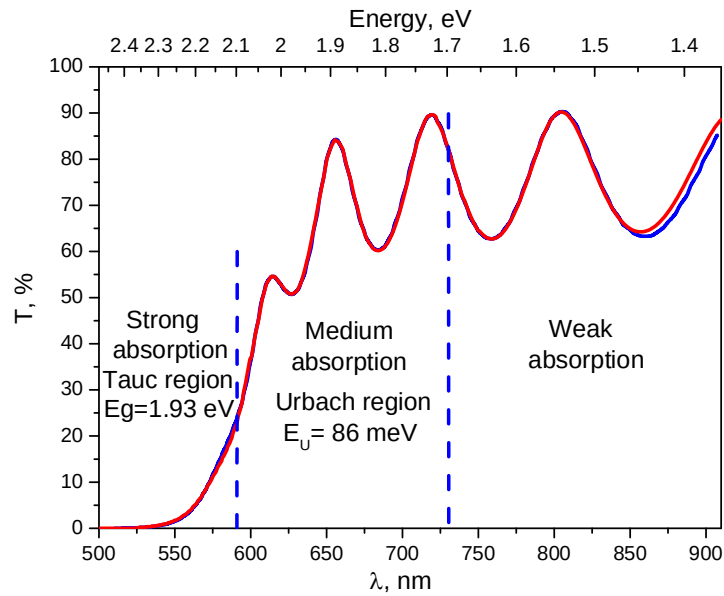


Fig. 8. Measured transmission spectrum $T_{exp}(\lambda)$ (blue curve) and modeled transmission spectrum $T(\lambda)$ based on adjusted absorption coefficient $\alpha_{adj}(\lambda)$ (red curve).

4. CONCLUSIONS

Optical properties of chalcogenide nanomultilayer structure $\text{As}_2\text{S}_3\text{:Mn-Se}$ obtained by thermal vacuum evaporation were studied. The spectral dependencies of the optical constants, namely refractive index $n(\lambda)$ and absorption coefficient $\alpha(\lambda)$ of the studied NML sample were retrieved from transmittance spectrum using the PARAV software based on envelope Swanepoel method. The indirect synthesis method based on modeling of transmittance spectrum $T(\lambda)$ inserting obtained multi-constant dispersion function for $n(\lambda)$ and $\alpha(\lambda)$ into theoretical formulas of transmittance was applied to prove their reliability. Simulated spectrum using combination of obtained $n(\lambda)$ and $\alpha(\lambda)$ data gave bad fits for as-measured transmittance spectrum in the Urbach region of absorption, so absorption coefficient $\alpha(\lambda)$ was not adequate obtained by PARAV software. To obtain accurate and reliable data of optical properties absorption coefficient $\alpha(\lambda)$ was manually adjusted to achieve optimal consistency between theoretically simulated curve and measured one. This curve-fitting method corrects the abnormal value of absorption coefficient, which proves to be a powerful curve-fitting method for analyzing optical spectra of nanomultilayered structures by comparing theoretical transmittance spectrum with experimental obtained one. Finally, we obtained the reliable refractive index dispersion and absorption coefficient dependence in whole studied optical spectra for chalcogenide nanomultilayer structure $\text{As}_2\text{S}_3\text{:Mn-Se}$. Applied curve-fitting and envelope methods will be helpful and complementary in study of optical properties of chalcogenide nanomultilayer structures.

Acknowledgements

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