Effect of X-ray and β-irradiation on the refractive index of complex chalcogenides glasses

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The measurement of the optical characteristics and parameters of solids, namely, those prepared for measurement of samples, can be significantly affected by the optical quality of their surface, surface and bulk impurities [1, 2].

In accurate optical measurements, such an effect can be due to the oxidation of the surface and the adsorption of molecules from the environment. For such molecules, the dissociation barrier is reduced and the processes of diffusion of its fragments into volume are improved [3-5]. This can even lead to a rebuilding of the bonds in the surface layer. Therefore, at least a two-layer system similar to the film-substrate system arises. Such a film on the surface can be formed on both bulk and film samples [6-8].

The refractive index of such objects is investigated by the ellipsometric method. Its advantage is the simultaneous determination of two complex refractive indices ($N_{1,2}=n_{1,2}$-i$k_{1,2}$) of the film-substrate system and the film thickness d. The method of ellipsometry can be applied to ferroelectric materials, since the formation and inhomogeneous surface layer of them is additionally influenced by the presence and properties of the domain structure.

The optical properties, including the refractive index, will be further influenced by high-energy (ionizing) radiation [1, 9, 10].

We investigated samples of glass $Sb_{24}Se_{60}Ge_{16}$ and $Sb_{20}Se_{60}Ge_{20}$, before and after X-ray irradiation, and samples $(As_2S_3)_{1-X}(AsI_3)_X$ (X = 0.04; 0.05; 0.07; 0.08; 0.1; 0.2; 0.3) before and after β-irradiation with eight doses. The average size of the samples (≈10×6)×(1-4) mm. The source of the β-rays was the 16-capsule Sr-Y system with a maximum electron energy of 2.2 MeV (mean ~ 1.1 MeV), a flux density of ~ 109 electrons/(cm^2·s), and several hours of exposure. X-ray irradiation of the samples was carried out in 3 modes of installation. Dose-1 (360 Gy): voltage 20 kW, current 10 mA, irradiation power W = 12 Gy/min, exposure τ = 30 min. Dose-2 (600 Gy): voltage 31 kV, current 10 mA, W = 20 Gy/min, τ = 30 min. Dose-3 (1000 Gy): W = 7 Gy/min, τ = 143 min.

The refractive index n and extinction k were determined using an LEF-3M ellipsometer with a Ne-Ne laser (λ = 632.8 nm). Ellipsometric angles (Ψ, Δ) were measured in the range of laser beam incidence angles φ = 50-77° (polygonal ellipsometry method).

The effect of irradiation dose on the refractive index and thickness of the surface film (n_1, k_1, d) and sample volume (n_2, k_2) were investigated. The irradiation dose was determined by the power multiplied by the irradiation time, with only a change in β-irradiation.

The difficulty of an ellipsometric technique (solving an inverted ellipsometry problem) with a simultaneous variation of the 5 parameters of the film-substrate system is that their stepwise change in large intervals of values leads to ambiguity in their definition, especially in the domain d > 100 nm. This is because a different combination of the values of 5 parameters (n_1, k_1, d; n_2, k_2) can lead to practically the same standard deviation (σ) of the parameters $(\Psi, \Delta)_{theor}$ from $(\Psi, \Delta)_{exper}$ or to unrealistic values (n, k) (Fig. 1). Therefore, we have selected the most realistic expected intervals of the possible thickness of the surface layer, where the values of N_1 and N_2 were determined (calculated):

1) calculations in the model-1 of a THIN surface film, 10-160 nm, ($d \leq \lambda/4$);
2) calculations in the model-2 of a NON-THIN surface film, 300-450 nm, ($d \approx \lambda / 2$);
3) calculations in the model-3 of a THICK surface film, 600-900 nm, ($d \geq \lambda$).

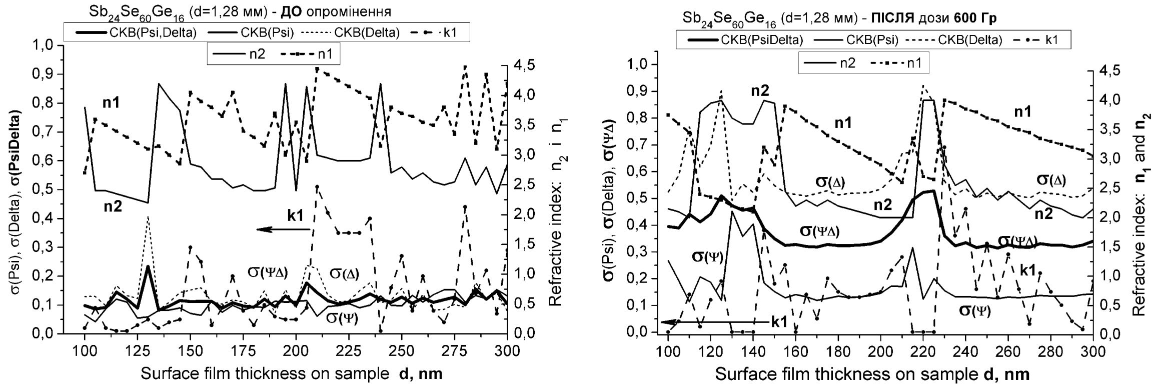

Fig. 1. The dependence of the root mean square deviation [σ(Ψ), σ(Δ) and σ(ΨΔ)] from the thickness, as well as the corresponding values of the indexes of the substrate (sample, n_2, k_2) and the film possible on it (n_1, k_1) for the minimum value σ at the given thickness.

Below is a part of the results of measurements of the dependences (Ψ, Δ) on the angle φ, which determined the parameters of the samples in the specified intervals of the possible thickness of the surface layer.

Sample-1 with a total thickness of 1.28 mm (Fig. 2). Prior to irradiation, the thickness of the surface layer could be up to 20 nm (bold), and after irradiation with a dose of 600 Gy it increased several times (80-100 nm) or even up to 300 nm. Accordingly, the refractive index has changed significantly. A quasi-periodic change in the dependence Ψ(φ) is observed.

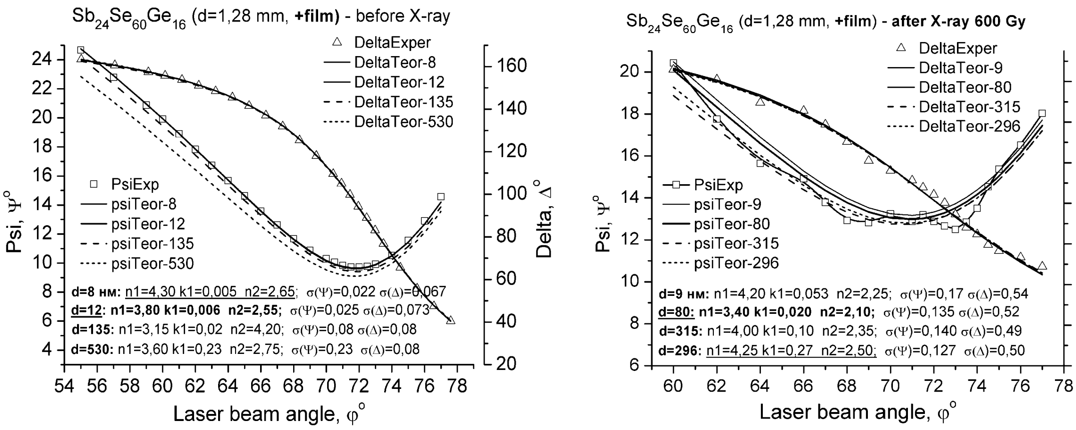

Fig. 2. Changing the refractive index and the thickness of the surface layer of the sample $Sb_{24}Se_{60}Ge_{16}$ after its X-ray irradiation (600 Gy dose).

For the samples of the composition $(As_2S_3)_{1-X}(AsI_3)_X$, similar studies were performed, both depending on the concentration of the component (AsI_3) and the dose of β-irradiation. Part of the results are presented in Fig. 3 and Table 1. We see that here also the experimental dependences Ψ(φ), as well as Δ(φ), after β-irradiation, their quasi-periodic change is more clearly observed.

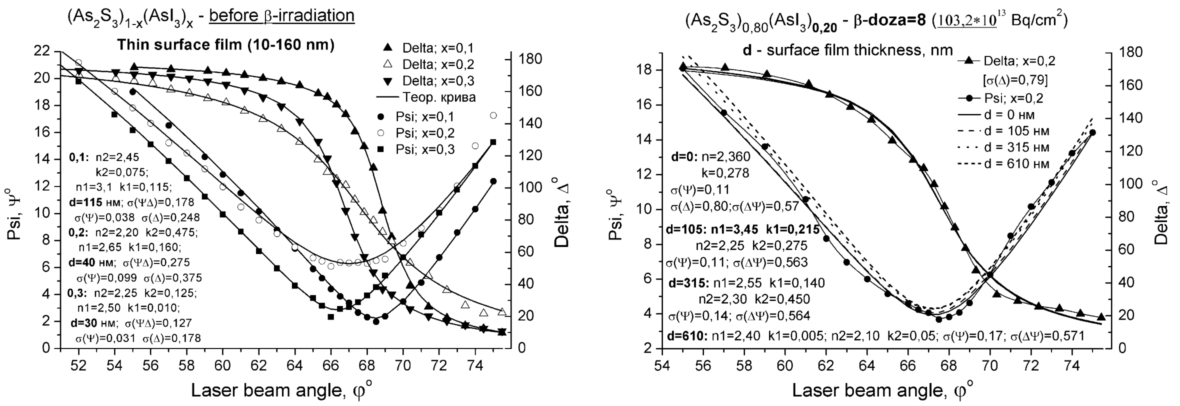

Fig. 3. Dependences of Ψ(φ), Δ(φ) of samples $(As_2S_3)_{1-X}(AsI_3)_X$ (X = 0.1; 0.2; 0.3) and calculated values (*n*, *k*, *d*) before irradiation (a) and after β-irradiation (b) at a dose-**8** ($103.2\cdot10^{13}$ Bq/cm^2; the result with the smallest values of σ is given in three intervals d only for the sample X = 0.2).

Table 1. Dependence of parameters (*d*, *n*, *k*) of samples $(As_2S_3)_{1-X}(AsI_3)_X$ on the dose of β-irradiation, Bq/cm^2: dose **6** = $55.2\cdot10^{13}$, **7** = $79.2\cdot10^{13}$, **8** = $103.2\cdot10^{13}$

	***d*, нм**				***n***				***k***			
Доза:	**0**	**6**	**7**	**8**	**0**	**6**	**7**	**8**	**0**	**6**	**7**	**8**
Зразок	**MODEL 1 (10-160 nm)**											
x=0,04(d,n_1,k_1)	10	90	155	115	3,55	3,55	3,00	2,70	0,025	0,005	0,020	0,050
x=0,04 (n_2,k_2)					2,60	2,50	3,50	2,50	0,325	0,025	0,175	0,100
x=0,10(d,n_1,k_1)	115	120	95	100	3,10	3,05	3,35	2,55	0,115	0,060	0,025	0,060
x=0,10 (n_2,k_2)					2,45	2,70	2,45	2,80	0,075	0,425	0,075	0,025
	MODEL 2 (300-450 nm)											
x=0,04(d,n_1,k_1)	380	420	415	410	2,95	3,20	3,25	2,55	0,185	0,250	0,250	0,150
x=0,04 (n_2,k_2)					3,20	2,55	2,60	2,65	0,500	0,350	0,400	0,226
x=0,10(d,n_1,k_1)	415	300	430	440	2,65	2,75	3,10	3,05	0,055	0,095	0,280	0,235
x=0,10 (n_2,k_2)					2,75	3,05	2,50	2,50	0,150	0,050	0,275	0,225
	MODEL 3 (600-900 nm)											
x=0,04(d,n_1,k_1)	610	690	630	890	2,85	2,95	2,90	2,60	0,200	0,270	0,205	0,095
x=0,04 (n_2,k_2)					2,70	2,55	3,25	2,50	0,400	0,375	0,350	0,100
x=0,10(d,n_1,k_1)	780	640	900	670	2,65	2,60	2,75	2,75	0,085	0,095	0,275	0,170
x=0,10 (n_2,k_2)					2,60	2,45	2,90	3,00	0,175	0,125	0,475	0,250

A multi-angle ellipsometry technique is applied to the study of the surface of samples of complex chalcogenide glasses and the effect of β- and X-ray on the refractive index. The analysis of the results shows that the consideration of the surface layer gives a better agreement of the experimental dependences Ψ (φ) and Δ(φ) with the theoretically calculated ones. This confirms the presence of such optically different volume layers.

The magnitude and nature of the change in the thickness of the layer d and the optical parameters (n_1, k_1), as well as the volume (n_2, k_2), under the action of β- and X-rays, depends on the radiation dose and its power (for X-rays), when this absorbance k and the thickness of the surface layer d may increase several times. However, a large dose of X-irradiation (1000 Gy) but low power (long exposure time) causes smaller changes in the refractive index and may even decrease (n_1, k_1, *d*) compared to the values before irradiation.

After the action of powerful X-irradiation, relaxation processes of 10-20 days are observed on the $Sb_{20}Se_{60}Ge_{20}$ and $Sb_{24}Se_{60}Ge_{16}$ samples, and occur more rapidly at lower irradiation power.

High-energy quanta (high-energy ionizing irradiation) are able to ionize atoms in the glass grid, reorienting their interatomic bonds [11]. Because the binding energy of our Sb-Se-Ge glasses is the smallest between Sb-Sb and Ge-Sb atoms, it is likely that ionization of bondreversed atoms can occur under irradiation or that Ge and Sb atoms can cause defects implementation and jobs. These causes, by accumulating defects of this kind at high dose and irradiation power, lead to a change in the refractive index.

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