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PECULIARITIES OF PHOTOREFRACTION EFFECT IN THICK GLASSY As_2S_3 FILMS

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Photorefraction phenomenon in thick glassy As_2S_3 films is studied. Violation of the reciprocity law for photorefraction and anomalous intensity dependence of photorefraction are observed and discussed.

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1. Introduction

The holographic recording in glassy As_2S_3 films was investigated by many authors [1-7]. Interest in this problem increased lately after the pubication of papers demonstrating new prospects of holographic recording in thick (up to 10.5 μm) non-annealed As_2S_3 films [8-12]. Recording in glassy chalcogenide films is a phase recording based on the photorefraction effect (photoinduced change of the refractive index) which, in parallel with photodarkening, accompanies the photostructural transformations. Photorefraction in As_2S_3 films was investigated both in early and in very recent studies [1,13-18], using various techniques but usually in thin film samples (0.2-1.0 μm).

In this paper we report some results of our recent research of photorefraction in thick As_2S_3 films. The data obtained allow to understand some peculiarities of holographic recording in such films.

2. Experimental

As_2S_3 films were prepared by evaporation of crushed As_2S_3 bulk glass onto oxide glass substrates from a quartz crucible in a vacuum of $\sim 5\times 10^{-6}$ Torr. The deposition rate was about 4 nm/s and the resulting film thickness was 4.0 - 5.0 μm.

An Ar^+ laser beam (λ = 515 nm, P = 3 W/cm^2) was used for photodarkening of the films. The diameter of irradiated area was ~6 mm within which the expanded laser beam had adequately uniform intensity. During exposure the As_2S_3 sample was illuminated by a normally incident He-Ne laser probe beam (λ = 632.8 nm, diameter ~1mm), and the transmitted part of this beam, was recorded as a function of time until saturation of the transmission was achieved. Similar curves of the transmission kinetics at various Ar^+ laser beam intensities allowed the determination of the beam power necessary to change the optical path in the film by $\lambda/2$ in each case. Although the absorption of the He-Ne laser beam in As_2S_3 glass is rather small [19], its intensity was kept at 0.04 W/cm^2 to minimize any effect on the recording [9].

To obtain independent values for the refractive indices they were calculated by the Swanepoel method [20] using a Hitachi U-1100 Spectrophotometer to measure the transmission spectra in non-irradiated regions and regions irradiated with different Ar^+ laser beam intensities.

In order to see if the studied film is irradiated homogeneously through its whole thickness, we investigated the rate of dissolution of the irradiated areas of the film in some solvents, using a thin film interference technique [21,22].

3. Results and discussion

Fig. 1 shows typical kinetics of relative transmission of He-Ne laser probe beam changes induced by Ar^+ laser beam irradiation. Obviously, decreasing the Ar^+ laser beam intensity results in slower kinetics. Measuring the time between two first maxima we can determine the irradiation doze necessary for changing the optical path length by $\lambda/2$ at each Ar^+ laser beam intensity.

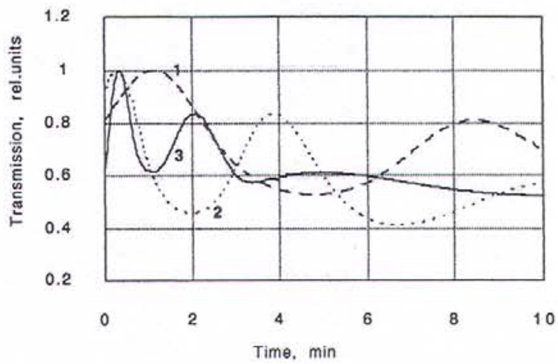

Fig. 1 Kinetics of transmission of He-Ne laser beam change induced by Ar^+ laser light with intensity of 0.35 W/cm² (1), 0.70 W/cm² (2) and 1.42 W/cm² (3).

Dependence of this dose on the Ar^+ laser beam intensity is demonstrated in Fig. 2 and shows the violation of the so called reciprocity law which assumes a constant relation between exposure and the change in the refractive index regardless of the laser power. Indeed, increasing the Ar^+ laser beam intensity from 0.3 W/cm² to 2.8 W/cm² results in a noticeable decrease of the necessary dose of irradiation, that is, in the increase of photosensitivity of photorefraction.

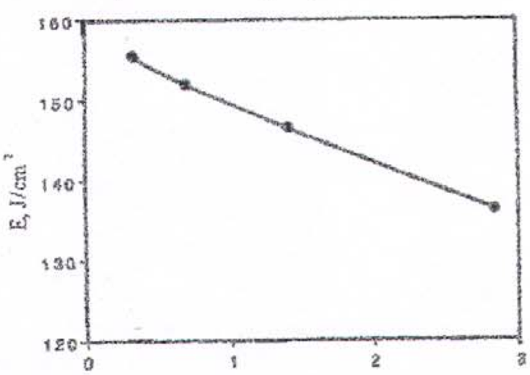

Fig. 2 Irradiation dose $E = W \times t$ changing the optical path length by $\lambda/2$ as a function of the Ar^+ laser beam intensity W.

The growth of photosensitivity of different photoinduced processes in chalcogenide glassy films with the increase of exciting light intensity was observed by many researchers. Especially a huge increase was recorded in case of intense pulsed light excitation [23-25]. Recently such increase was explained by the appearance of a two-photon effect when two photons absorbed at neighbouring atoms significantly increase the probability of structural transformations [26]. We believe that such a process starts to be effective already at much smaller light intensities leading to the violation of the reciprocity law observed in our experiments.

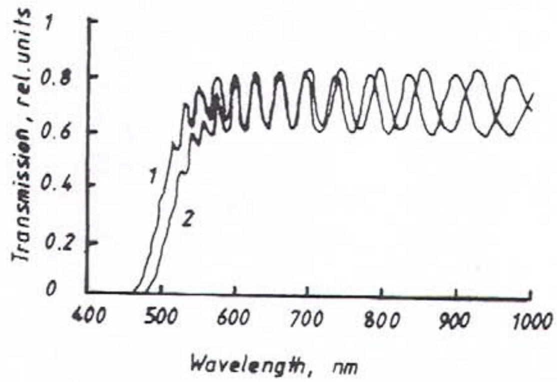

Fig. 3 Transmission spectra of non-irradiated (1) and Ar^+ laser light irradiated (2) areas.

Transmission spectra of non-irradiated and strongly irradiated areas of thick As_2S_3 film are shown in Fig. 3. Several refractive index spectra for the areas irradiated with various Ar^+ laser beam intensities and calculated from the transmission spectra, are demonstrated in Fig. 4. It is possible to conclude that irradiation with smaller intensity is accompanied by larger change of refractive index. Such anomalous intensity dependence of photorefraction is illustrated in Fig. 5 for photorefraction at λ= 632.8 nm.

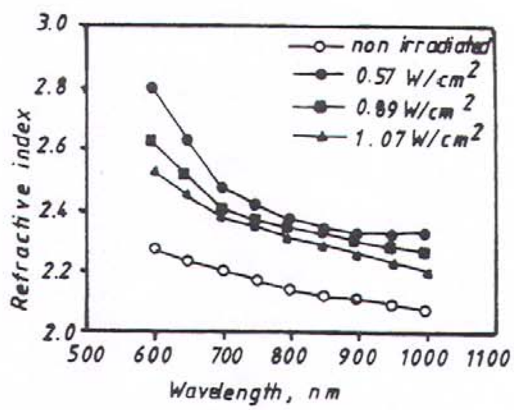

Fig. 4 Spectra of refractive index for non-irradiated film and films irradiated with various Ar^+ laser beam intensities.

This peculiarity of photorefraction in the studied films is in good agreement with data about the existence of maximum in the intensity dependence of diffraction efficiency of holograms recorded in thick As_2S_3 films [8]. Moreover, our data allow us to select between several possible reasons for the appearance of the maximum mentioned in [8]: temperature increase, laser beam fanning or non-linearity in the recording with intensity. Now we can say that the reason is the non-linearity of photorefraction. We believe that such anomalous intensity dependence of photorefraction is due to strong photodarkening of the films when larger time of irradiation must be used in order to treat the whole volume of the film. This effect is especially noticeable in the thick films that were studied in our research.

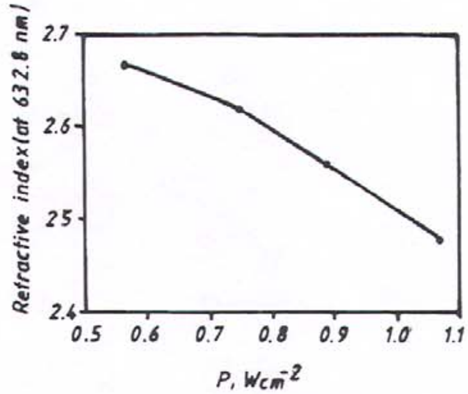

Fig. 5 Refractive index at λ= 632.8 nm as a function of the Ar^+ laser beam intensity.

The results of study of photorefraction homogeneity through the whole film thickness are shown in Fig.6 where the intensity of the He-Ne laser beam transmitted through the As_2S_3 film during its etching in the positive selective developer, based on dimethylamine, is presented. The constancy of the period of the interference pattern testifies that the etching rate is constant during the whole dissolution process of the irradiated film, confirming a homogeneity of photostructural transformations. This result indicates that we obtained the real refractive index values and not the average values. Large difference in the etching rate for non-irradiated and irradiated films is also demonstrated.

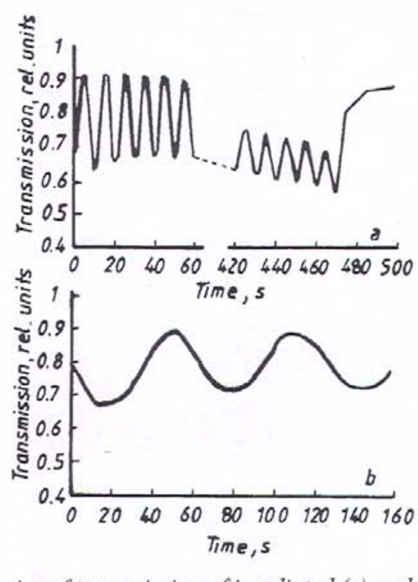

Fig. 6 Kinetics of transmission of irradiated (a) and non-irradiated (b) thick As_2S_3 films in process of their etching (for non-irradiated film only initial periods of etching are shown).

4. Conclusion

We investigated some characteristics of the photorefraction phenomenon in thick As_2S_3 films successfully used in holographic recording and showed that the Ar^+ laser beam at $\lambda = 515$nm is capable of homogeneously exciting the whole thickness of the film. Two peculiarities of photorefraction were observed: the violation of the reciprocity law and the anomaly in photorefraction when smaller light intensity irradiation is accompanied by a larger change of refractive index. These peculiarities are connected with characteristic properties of photostructural transformations in chalcogenide glasses.

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