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SHORT COMMUNICATION

PHOTO-INDUCED SURFACE DEFORMATION DURING HOLOGRAM RECORDING IN a-Se FILMS

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The photo-induced deformation of a-Se film surface during hologram recording has been investigated. The surface deformation is reversible if the crystallization is not advanced.

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1. Introduction

Photo-induced effects (PE) in amorphous chalcogenide semiconductors consist in the modifications of optical parameters, viscosity, microhardness, volume and some other characteristics, which essentially depend on the structure, and its transformations in these materials [1-3]. Optical information recording may be performed due to the existence of these PE [3,4]. A large study of the photo-induced structural transformations in amorphous selenium (a-Se) has been carried out during the past few years [3-6]. As a result, have been developed new ideas regarding PE, including photo-crystallization, in a-Se [7-10], as well as the mechanism of optical recording in light-sensitive chalcogenide glasses [1-3]. Since a-Se is the simplest among the amorphous chalcogenide materials which possess PE, it is still attractive as a model for basic investigations and for applications in optoelectronics, for optical recording.

Recent models of photo-induced structural transformations contain a common first step of electron-hole pair excitation by band-gap illumination and further steps of structural re-arrangement. These steps have different possible pathways, based, for example, on the increase or decrease of average selenium coordination number [5], ring-to-chain conversion [6], ordering-disordering transformation, densification processes, etc., which possibly depend on the technology or additional treatments of the a-Se layer. Photo-induced volume changes were observed in amorphous chalcogenide thin films and a-Se layers [1]. Contraction effects were observed, first of all in obliquely deposited chalcogenide layers [11]. They are due, possibly, to the transformation of the columnar structure, which is an ordering process in as-deposited films. So the mechanism of PE and optical recording even in a-Se films with a relative simple structure seems rather incomplete. For example, despite the conclusions about the impossibility of stable optical recording in a-Se at room temperature [5,6], stable holographic gratings were recorded on a-Se layers at 294 K [3,4,7]. With the aim to look into the mechanism of such recordi ng, as well as into the details of surface relief formation on a-Se-containing multilayers [12], we have performed investigations of holographic recording combined with direct AFM measurements of the resulting surface profile on a-Se films.

2. Experimental

The samples were prepared by thermal evaporation of bulk high purity selenium in a vacuum of 10^{-4} Pa onto silica glass substrates kept at room temperature. High quality substrate surface was necessary to obtain 1 to 2 μm thick layer with a surface roughness less than 0.5 nm. The deposition rates were 1 to 3 nm/s. Structural inhomogeneity, porosity of as-deposited amorphous layer may cause large irreversible structural changes after the first thermal treatment or even during the storage at room temperature. That is why the samples were usually stored for few days at room temperature in darkness before being measured in order to decrease the possible effect of their history.

The sample was set up on a holder, which provides possibility of conventional lenseless holographic recording with He-Ne laser (λ= 0.63 μm). Readout can be carried out in transmission or reflection mode. The average light intensity at the sample surface was 0.8 W/cm^2. Gratings were recorded with spatial frequencies in the 600-1000 mm^{-1} range. Read-out was performed periodically by blocking one of the recording beams so that the hologram was irradiated for a short time with only one beam. Since the diffraction efficiency η correlates with the optical amplitude-phase modulation relief on the recorded holographic grating, indirect information about the changes of the refractive index n or thickness d of the layer can be obtained. Direct information about the surface structure and grating profile was obtained by AFM measurements. Photo-induced structural transformations in a-Se layer under the similar laser illumination were proved by Raman scattering measurements.

3. Results and discussion

The diffraction efficiency η as a function of the recording time has two distinct parts and peaks, as it is shown in Fig. 1. The first very weak peak is observed at exposition E ≈ 60 J.cm^{-2}, which is equal to the usually observed ones in chalcogenide films under the conditions of volume grating recording [3,7]. It is caused by the typical photodarkening and corresponds to the refraction index, and reflectivity change. This component of optical recording is rather small (relative transmission change τ/τ_0 equals to few percents [3]) and unstable in a-Se layers at room temperature because of the vicinity of the a-Se softening temperature ($T_g \approx$ 305-310 K) and fast (few minutes or hours) decay of ordinary photostructural transformations at 294 K.

Surprisingly, further recording allows to reach much higher efficiency of the hologram ($\eta \approx$ 1.3 % at the second peak in Fig. 1), which is very stable at 294 K (three months at least).

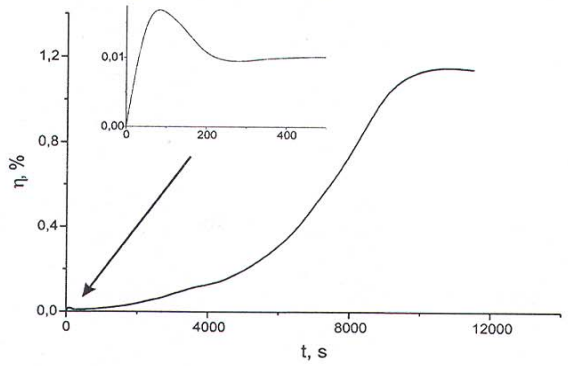

Fig. 1. Diffraction efficiency as a function of recording time (exposition) on a-Se film. The insert shows the first peak in details. Read-out was performed in reflection mode.

The a-Se film continues to dark during the second, high-exposition stage of hologram recording, but it is not the real reason of the relative high efficiency because of the small ordinary photostructural transformations, as mentioned above. The reason lies in a surface relief formation, which has sinusoidal, up to 40 nm deep profile according to the AFM measurements (see Fig. 2).

The surface relief of the hologram, exposed up to 10^4 s, can be completely erased by annealing at T ≈ 310 K, and the recording may be repeated several times until the crystallization

effects will not restrict the relaxation and destroy the surface quality. Photocrystallization was observed on the overexposed hologram (Fig. 2b), that means twice more exposition in comparison with the second peak. This effect is more distinct and rapid in the repeated, third-fifth cycle of optical recording-thermal erasing. The spatial frequency in the mentioned limits has little influence on the η value and on the relaxation rate.

The main significance of the presented experimental results consists in the discovery of reversible, single-step surface deformation in a-Se films during hologram recording at temperatures close to the softening temperature. Being obviously connected to the known ordinary PE (at least on the first step of photon-electron interaction and atomic displacements), these deformations are much more stable even at temperatures, where PE are thermally erased during short periods. It means that additional driving forces and mechanisms must be involved, in order to explain this effect.

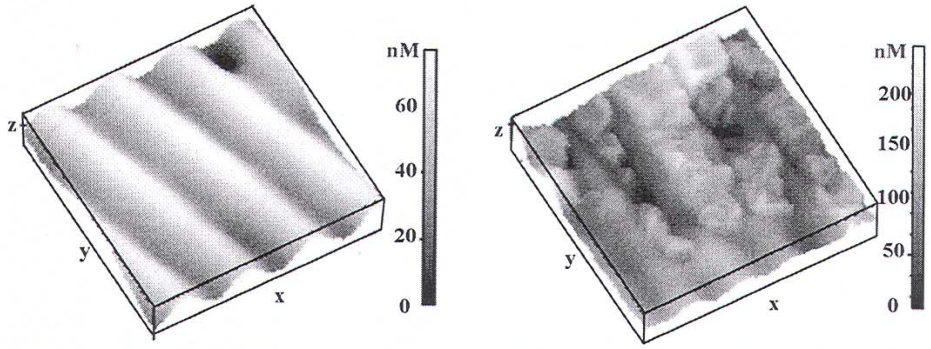

Fig. 2. a) AFM picture of the surface relief obtained on a-Se film after hologram recording up to the second maximum in Fig.1.
b) AFM picture of the surface relief in an overexposed a-Se layer.

Ablation and thermal effect of the irradiation may be neglected since the temperature in the layer does not rise more than 1-2 K in the given experimental conditions. Small volume expansion or contraction in irradiated places may be explained on the basis of previous models, taking into account the interaction of the charged neighboring structural elements of the glass [13]. Nevertheless, our experiments on a-Se layers show a pronounced, large effect of layer contraction up to 2% of total thickness d in the more intensively irradiated areas, where crystallization occurs at the final stage of illumination (see Fig. 2.b). At the same time crystallization is not apparent in the reversible cycles of recording-erasing, as it follows from the AFM measurements, although the tendency to the crystallization may be expected from the results of Raman-scattering measurements (see Fig. 3).

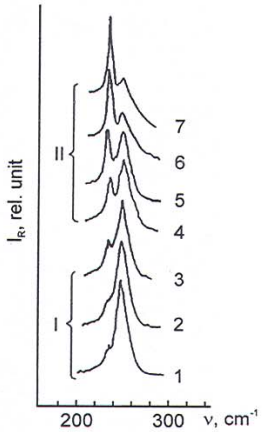

Fig. 3. Raman spectra of a-Se layer at 294 K. 1- initial state, 2-7 - after the successively increasing expositions.

The typical Raman spectrum of a-Se layer, measured at 294 K with the same radiation power density of He-Ne laser, contains a broad band with a maximum at ~250 cm^{-1} and a small "shoulder" at ~232 cm^{-2}. The spectrum does not change significantly during the first stage of exposition, which corresponds to the hologram recording in the reversible cycles, excepting a small redistribution towards the increasing "shoulder" upon illumination, or backward to the initial state after the relaxation in the dark or thermal erasing. At longer expositions, the peak at 232 cm^{-1} irreversibly increases and the peak at 250 cm^{-1} decreases. This corresponds to the second stage of hologram recording, when crystallization of a-Se layer occurs. Obviously, laser-induced local ordering process occurs in a-Se, which is similar at small expositions to the nucleation stage of the glass crystallization. Chain-type, trigonal Se structure of these nuclei follows from the development of 232 cm^{-1} peak in the Raman spectra, as it was first established in [7] and supported in [14]. As far the dimension of such nucleus does not exceed the critical radius r_c [15], it is unstable and can be amorphised (in the dark, during annealing). Greater crystallites continue to grow up to the irreversible crystallization of the layer. Since the fundamental absorption edge of the trigonal (t) Se is shifted towards longer wavelengths [15], laser illumination results in the photodarkening of the a-Se layer. This partial ordering may be also expected as a large-scale disordering, as it is often assumed for the photodarkening process. Finally the most important reversible photo-induced contraction effect follows from the difference in the densities of amorphous and trigonal selenium: $\rho_{t\text{-}Se}$-$\rho_{a\text{-}Se} \approx 4.82\text{-}4.28 = 0.54$ $g.cm^{-3}$ [15] or similar in other papers. It means more than 10% volume contraction at total crystallization. Since the volume of the sub-critical nuclei is of about 30% of the amorphous phase, the experimental results seem to be in a reasonable accordance with the proposed model of localized photo-induced contraction in a-Se layer.

4. Conclusions

Surface relief formation during hologram recording on amorphous selenium at room temperature is due to a photo-induced local ordering process and layer contraction. The process is reversible until the crystallization prevails. Thereafter, the process becomes irreversible.

Acknowledgements

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