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SHORT COMMUNICATION

PHOTOINDUCED EXTENSION AND OPTICAL RECORDING IN a-Se/As_2S_3 MULTILAYERS

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1. Introduction

Photoinduced structural transformations (PST) in AsSe, As_2S_3, GeS_2 and analogous chalcogenides are rather well known and reviewed in a number of papers (see, for example [1-3]). Large changes of optical absorption and reflection, as well as the change of the refractive index Δn at PST in chalcogenide glasses are the basis for amplitude-phase optical recording, which is performed usually in 1-2 μm thick homogeneous layers, without or with a subsequent chemical etching. The last is necessary for producing deep surface relief, for example holographic diffraction gratings (HDG) [4]. In spite of some contradictory experiments [5,6] amorphous selenium (a-Se) layers also undergo reversible PST at 293 K, if photocrystallization is avoided. It is desirable to use this simple material for optical recording, and for adding new ideas to the mechanism of recording.

The mechanism of PST was successively enlarged, especially taking into account the novel ideas and experimental possibilities [6,7,8], but the known effects of 0.3-0.6% reversible volume extension or contraction at PST are not unambiguously considered [9-11]. Besides AsSe or As_2S_3, were a-Se could be one of the basic model materials to explain and apply such properties, but amorphous selenium layers easily undergo photo- or thermally induced crystallization at room temperature. The stability of a-Se might be essentially increased in nanolayered structures [12,13] and the a-Se/As_2S_3 multilayers (ML) appeared as highly effective for holographic recording [14]. Further investigations of the interconnections between photoinduced changes of optical and dimensional parameters in such multilayer structures are presented in this paper.

2. Experimental

Samples were prepared by cyclic vacuum thermal evaporation of a-Se and As_2S_3 from separate sources. Nanolayered structures with optimum period = 10-14 nm and total thickness d = 0.5-2.5 μm were deposited on silica glass substrata with high quality surfaces (roughness less than 1 nm). The periodicity was controlled by Small Angle X-ray Diffraction method, the surface structure and relief were investigated by Atomic Force Microscopy. Homogeneous layers of a-Se and As_2S_3 with the same total thickness as the sum of corresponding sub-layers' thickness in ML were prepared simultaneously for the comparison of PST in homogeneous and nanolayered structures.

Polarized illumination or simple HDG recording-readout was performed by He-Ne laser (λ = 0.63 μm, P = 0.6 W/cm^2). Taking into account the fact that the fundamental absorption edge positions in a-Se (~2.0 eV) and As_2S_3 layers (~2.4 eV) are shifted towards shorter wavelengths in

ML [12,13], a-Se was optically excited and PST occurred first of all in these sublayers. The optical transmission change $\Delta T/T$ was measured at the same laser wavelength and compared with the data obtained from optical transmission spectra. The refractive index, as well as thickness change were determined from the interference patterns of these spectra.

3. Results and discussion

The first important and easily observed peculiarity of PST in a-Se/As_2S_3 is that not photodarkening but photobleaching occurs in all as-deposited samples, while in homogeneous a-Se layers it was never observed. For As_2S_3 it was detected only in extremely non-equilibrium layers [15], that does not correspond to ML. The optical absorption edge shifts for $\Delta\lambda \approx 10$ nm towards shorter wavelengths in the 0.55 - 0.65 μm spectral range, that means a relative transmission change of $\Delta T_{max} / T = 5.5\%$ at the recording wavelength $\lambda = 0.63$ μm.

The second peculiarity consists in the giant extension of the ML in the direction of Z axis, which is normal to the ML surface and coincides with laser beam direction. The light-induced volume (layer thickness) change in amorphous chalcogenides is usually ignored in optical experiments, first of all, possibly, because of small values (0.3 - 0.6 % [2]) and difficulties in measurements. HDG recording gave us an unique possibility to investigate the kinetics of the localised layer deformation through the measurements of the HDG efficiency $\eta = I_{T,R}/I_0$, where I_0 is the intensity of the readout light and I_T or I_R are intensities of the light transmitted or reflected to the first order diffraction peak. Of course, it was necessary to prove independently the sign and the extent of the layer thickness change. This was made by the direct AFM measurement of the resulting surface relief on HDG (Fig. 1): the height of the sinusoidal profile achieved 100 nm in 2.5 μm-thick ML, that means 4% average thickness change. The same changes were determined from the optical transmission spectra (Fig. 2, curve 1).

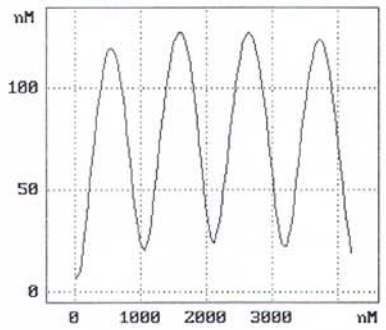

Fig.1 Sinusoidal surface relief profile of HDG, measured with AFM on a-Se/As_2S_3 ML

The $\Delta d/d$ value is somewhat smaller than the HDG relief height at the same exposure E. This fact, as well as the shift between the saturation points of curves 1,2 and 3,4 on Fig. 2 may be attributed to the difference between uniform (curves 1,2) and periodical (in the ML plane) illumination of the layer. In the last case, additional driving forces may appear along X direction in the ML plane, perpendicularly to the grating rows. These driving forces seem to be the same as for the total thickness change and connected mainly with built-in stress and its relaxation during illumination [13,14]. Mainly, but not entirely, because a small reversible extension-contraction effect may be caused by polarized light [16] even in thick, annealed, homogeneous amorphous layers.

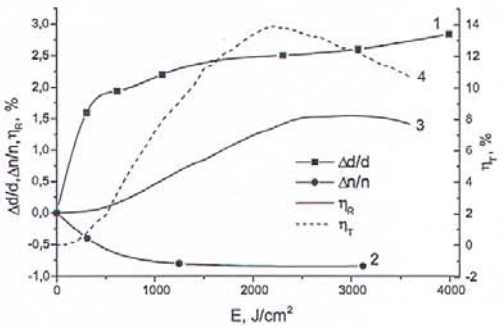

Fig. 2 Experimental dependencies of the relative thickness (1), refractive index (2), diffraction efficiency, measured in reflection (3) and transmission (4) mode, on exposure of the a-Se/As_2S_3 multilayer to the laser irradiation.

Few initiating mechanisms of local structural rearrangement in such layers were proposed and connected with the position of bridge chalcogen atoms, with the change of the intermolecular distances and with localized centres in chalcogenide glasses [1,2,9,17]. The thermally or light-stimulated structural relaxations erase these changes [13,18], and promote the stress decrease in the layer.

Homogeneous, annealed layers probably possess weak built - in stress and, therefore, relatively small deformations, as it was found in our a-Se and As_2S_3 layers. Obviously, the structure of the given a-Se/As_2S_3 ML-type is just favourable for stress accumulation and its relaxation under illumination because of the differences in $\Delta d/d$ [2] and effective photostimulated fluidity of a-Se, which is similar to chalcogenide glasses [19,20] just below the softening temperature of a-Se ($T_g \approx 305$ K in bulk samples and ≈ 350 K in a nanostructure [13]). The recorded relief may be thermally erased in this temperature range, but the next recordings are not so effective in ML.

Photobleaching in a-Se /As_2S_3 ML supposes negative $\Delta n/n$ contrary to the positive one in homogeneous layers, where photodarkening is usually observed. Our experiments showed negative $\Delta n/n$ (curve 2 on Fig. 2). If PST in ML are connected with a sensitive set of nanometer-thick a-Se sub-layers, the partial $\Delta n/n$ and $\Delta d/d$ are even greater as the above-mentioned values. The details of this complex mechanism of PST and optical, mechanical parameters' change in ML nanostructures need further investigations, but its importance is obvious for applied optics.

Both Δn and Δd modulations contribute to the efficiency η of the HDG recorded in the real time-scale. The readout may be performed in transmission or reflection mode. The last is better when self-absorption must be excluded. Maximum η in this case may be ~33 %, but the low reflection and some light scattering reduce η to few percents [14], what is in accordance with our results (Fig. 2, curve 3). The phase modulation of the incident light beam is more effective in the transmission-mode readout of such HDG, especially when the self-absorption is low. Our calculations were made according to the simple equation for a thin hologram [21]:

$$\eta = T \cdot J_1^2(\phi) \tag{1}$$

where ϕ is the centre-to-peak excursion of the phase shift in radians, T - optical transmission, J_1 – first order Bessel function. Taking into account two components of the phase shift:

$$\phi = \frac{2\pi}{\lambda}\left[\Delta n(d+B) + (n-1)B\right], \tag{2}$$

where λ - wavelength, B – relief depth. The resulting η = 10.3 % for the relief depth presented in the Fig. 1 (≈100 nm) and average initial refractive index n = 2.5 for ML, is in good accordance with the experimental results (Fig. 2, curve 4). This value was reached after rather large total exposure and it must be corrected for the low absorption at the recording-readout wavelength. Still the surface relief formation needs much longer time and longer exposure than, for example, the simple process of photodarkening in a-Se or AsSe at the same room temperatures [3]. Obviously, these processes have

common roots, but differ in mechanisms. The stability of the recorded surface relief is high: it is maintained in normal day-light conditions more than an year, so the investigated ML may be used for producing archive memory or elements with surface relief.

4. Conclusion

Giant photoinduced extension in a-Se/As_2S_3 nanolayered structures was observed and investigated. This effect is closely related to optical recording in such structures. The basical mechanism of PST and optical relief formation extended taking into account additional driving forces: stresses in such structures and stimulated deformations. This new type of light-sensitive material is useful for the surface relief recording in the real-time-scale.

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