

# Surface relief and polarization holographic grating formation in amorphous As-S-Se films

M. REINFELDE\*, J. TETERIS

*Institute of Solid State Physics, University of Latvia, 8 Kengaraga Street, LV1063, Latvia*

The results of surface relief and volume holographic grating (HG) formation in  $\text{As}_{40}\text{S}_{15}\text{Se}_{45}$  films under He-Ne laser wavelength  $\lambda=0,6328 \mu\text{m}$  light influence are examined. Recording beam polarisation and grating influence on surface relief parameters are shown. Microanalysis of chemical composition performed by SEM give evidence of As and Se concentration changes. Examination of beginning stage of recording kinetics demonstrates the presence of polarisation (vector) holographic gratings. Formation of complicated surface structures with different directions and dimensions are observed when hologram recording at opposite recording beam direction is carried out.

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## 1. Introduction

The possibility of direct surface structure formation for different materials is matter of current interest. The earlier works mainly belongs to such materials as polymer films where relations of surface relief and polarisation (vector) hologram (PH) recording were examined [1-3]. The importance of recording light polarisation was ascertained as well mass transport role for the process was indicated [2]. At the same time, the presence of surface relief formation for As, S and Se contained chalcogenide films explained by photo-structural transformations are subject of permanent interest [4-11]. The majority of studies are concerned with two component As-S(Se) compounds. In our studies we devote attention to three component  $\text{As}_{40}\text{S}_{15}\text{Se}_{45}$  films under influence of wave-light  $\lambda=0,6328 \mu\text{m}$  light belonging to bang-gap zone with absorption coefficient  $\alpha \sim 10^3 \text{ cm}^{-1}$  and we can attend more pronounced hologram (HG) volume effects. It is evident, that the phenomenon causing surface relief formation is the structure related mass transportation process during comparatively long light illumination. Specificity for direct surface relief formation is recording at orthogonally polarised light beam configuration. In such case intensity modulation is absent or nearly absent [12] and recording properties are determinated by electric field modulation. [13]. From the other side the evidence of possibility to create vector PH at orthogonally polarised light beam recording in amorphous chalcogenides was shown [14-17]. From that point of view we performed more deeply analysis for beginning part of recording process where appearance of vector PH could be expected.

## 2. Experimental

The  $\approx 2 \mu\text{m}$  thick  $\text{As}_{40}\text{S}_{15}\text{Se}_{45}$  films on glass substrate evaporated in vacuum were studied. Transparency of unexposed film  $T_0 \approx 40\%$  at  $\lambda=0,6328 \mu\text{m}$ . Two recording

schemes were used. First one-commonly used symmetric transmission scheme, Fig.1, both recording beam coincide on sample from the same side. Second - reflection scheme with recording waves coming from opposite sides. For checking up the diffraction efficiencies, we elected the slightly asymmetric configuration, with angle  $\Phi$  between reflected R1 and transmitted T2 light beams Fig.2. Half wave or four wave plates enable the elected beam polarization states. Photo diodes connected with computer are for fixation of diffracted light intensities. Diffraction efficiencies (DE) was determined as  $\eta = P_{\text{dif}}/P$ , where  $P_{\text{dif}}$  - diffracted beam,  $P$  - reading beam. Typically hologram recording was accomplished with equal light beam intensities  $I_1=I_2 \approx 0,25 \text{ W/cm}^2$  at room temperatures.

## 3. Results and discussion

Typical HG recording kinetics with surface relief formation in  $\text{As}_{40}\text{S}_{15}\text{Se}_{45}$  film under wave-length  $\lambda=0,6328 \mu\text{m}$  influence are shown in Figs. 3 and 4. Two parts in hologram recording curves can be distinguished. At the beginning for expositions not exceeding the  $50 \text{ J/cm}^2$  (inclusions in Figs. 3. and 4.) DE achieves certain maximum values. After some decreasing, values of DE begin to increase again and achieve saturation near the expositions  $10^3 \div 10^4 \text{ J/cm}^2$ . That second recording stage corresponds to surface relief formation process. Values of observed DE maximums depend on recording beam polarization states. For orthogonally orientated - s-p,  $\pm 45^\circ$ , circularly (R-L) - light beam polarizations surface relief hologram recording prevail. If recording at parallel linear and circular light beam polarization - s-s, p-p, R-R (L-R) is performed, relief formation is nearly absent, and maximum DE apparently belongs to scalar volume transmission HG, Fig. 4.

Dependence on recording light polarization belongs to properties observed commonly for surface relief HG recorded at different wave length [5]. Characteristic for recording at wave length  $\lambda=0.6328\mu\text{m}$  is opportunity to observe simultaneously the transmission nature of HG, Fig. 3, curves 2, 4.

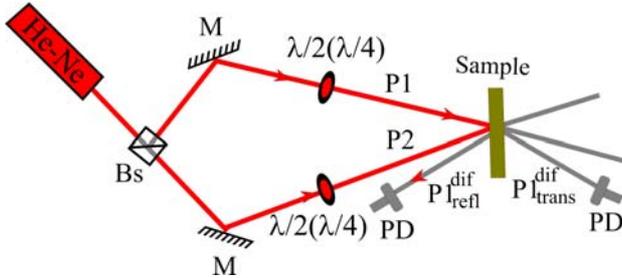


Fig. 1. Recording setup 1: P1,P2 – recording beams,  $P1^{dif}_{refl}$  and  $P1^{dif}_{trans}$  – reflected and transmitted diffraction orders respectively, Bs – beam splitter, M – mirrors, PD – photodiodes,  $\lambda/2(\lambda/4)$  – half or forth wave plate.

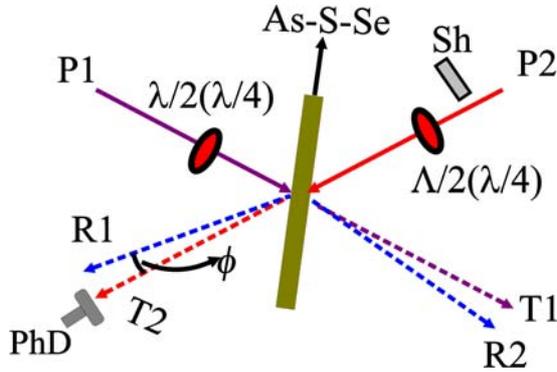


Fig. 2. Recording setup 2: P1,P2 – recording beams, T1,T2 – transmitted beams, R1,R2 – reflected beams,  $P^{dif}_{Bragg}$  – diffracted beam when shutter Sh is closed,  $\Phi$  – angle between reflected and transmitted beams, PD – photodiodes.

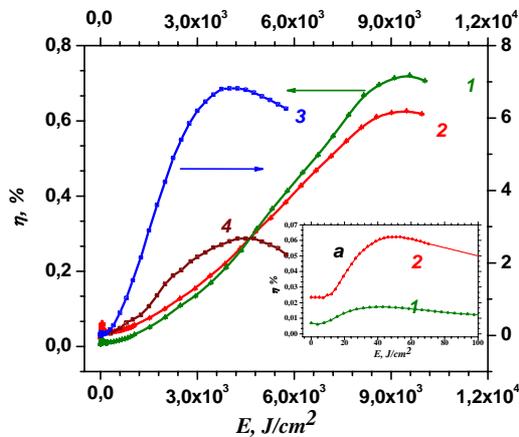


Fig. 3. Kinetics of holographic grating recording at orthogonally polarized s-p curves 1,2 and  $\pm 45^\circ$  curves 3,4 light beams: grating period  $\Lambda \approx 2.6 \mu\text{m}$ , light intensity  $I_1 \approx I_2 \approx 0.25 \text{ W/cm}^2$ ; sample thickness  $d \approx 2 \mu\text{m}$ ; a) beginning stage of recording.

Influence of grating period on surface relief hologram formation process and light sensitivity in dependence of grating period was studied as well, Fig. 5. Recording sensitivity  $1/S = \Delta E / \Delta \eta$  (exposition necessary per 1% DE) in dependence of grating period  $\Lambda$  was determinate from linear part of recording curve. It is clearly seen that decreasing of grating period below some values makes more difficult the surface relief formation process. Surface relief scanning by AFM marks the relief depth dependence on grating period, Fig. 6, Tab. Such course of dependence on grating period could indicate on influence of surface tension. The question arise how does the components of basic compound are affected. For this reason the surface relief of period  $\Lambda = 33 \mu\text{m}$  was prepared and SEM\* microanalysis of chemical composition for As-S-Se film was performed. One can see from Fig. 7, that As and Se shows the periodic oppositely directed concentration changes accordingly to grating period. Obviously the real motion of these components are opposite, too. As to concentration of S, there was not observable change of this component (not shown).

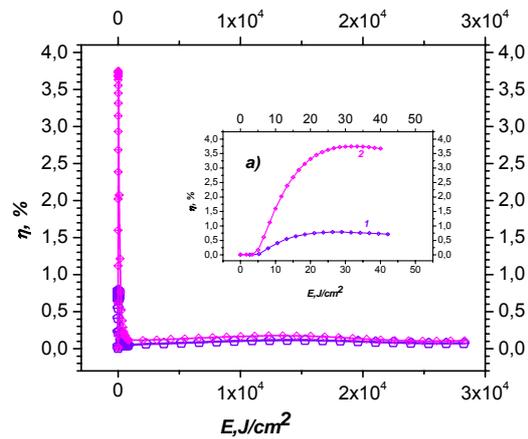


Fig. 4. Kinetics of holographic grating recording for mutually parallel (p-p) light beam polarization: grating period  $\Lambda = 2.6 \mu\text{m}$ , light intensity  $I_1 \approx I_2 \approx 0.25 \text{ W/cm}^2$ ; sample thickness  $d \approx 2 \mu\text{m}$ ; a) beginning stage of recording in enlarged mode.

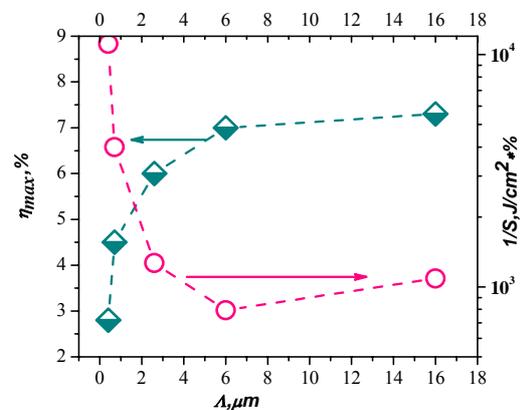


Fig. 5. Recording sensitivity and maximum diffraction efficiency dependence of on grating period for holograms recorded with  $\pm 45^\circ$  polarized light beams.

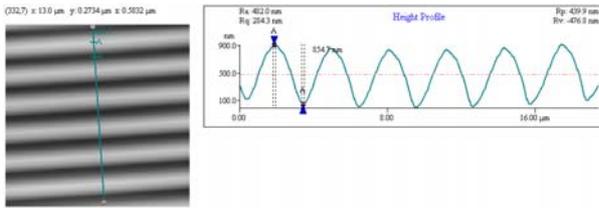


Fig. 6. AFM picture and relief profile of hologram grating with a period  $\Lambda=2.3 \mu\text{m}$ .

Grating period, $\mu\text{m}$	Relief profile depth, nm
0.7	82
1.2	240
2.6	855

More complicate relief structure develops if recording is fulfilled at reflection scheme as shown at Fig. 2. We expected the some appearance of reflection HG on the base of structure formation inside the volume of recording material. For checking up the diffraction efficiencies at Bragg's angle, we elected the slightly asymmetric scheme. Recording wave-beams coming from opposite sides at the intersection takes mutually orthogonal polarization. From the point of view on recording kinetics we obtained relationships surprisingly resembling the previously described for transmission scheme recording as at Fig. 3. The only difference was appearance of maximum values for DE at expositions approximately four times exceeding the correspondent values at transmission scheme. Besides, for angle  $\Phi \approx 1,3^\circ$  we observed large amount of diffraction maxima - we counted up to 20 maximum aside the Bragg's one - and symmetrically around reading beam arranged eight-petal like light scattering. Increasing the angle  $\Phi$  the number of maximums as well petals decreased. Such results make us to speculate, that we observe the HG recording due to mutual interaction of transmitted and reflected inside the film's volume light beams. Confirmation of such assumption we received from optical and AFM measurements shown at Fig. 8. We can see there composite surface grating structure of different period, direction and relief profile.

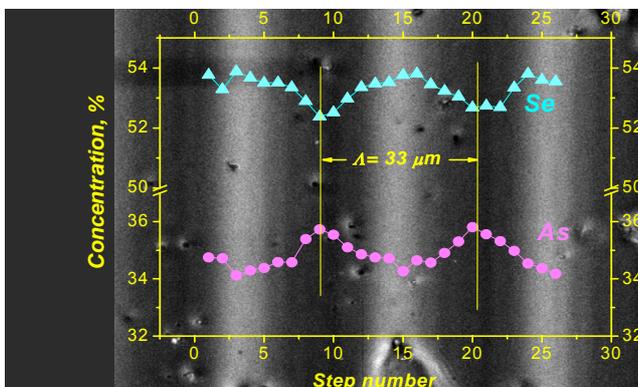


Fig. 7. AFM picture and relief profile of hologram grating with a period  $\Lambda=2.3 \mu\text{m}$ .

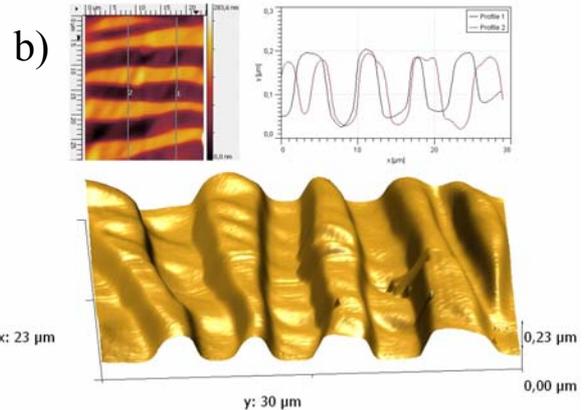
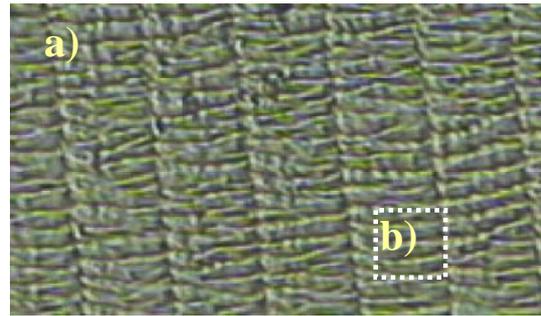


Fig. 8. Optical microscope – (a) and enlarged region in (b) - AFM pictures and relief profile for hologram recorded by reflection scheme;  $\Phi \approx 1,3^\circ$ .

Now let we consider more deeply beginning part of recording curves – insets a) in Figs. 3. and 4. As we noted previously, when recording are performed at linear or circular polarization configurations, scalar volume holographic grating appears due to intensity modulation. If compare maximum DE for that part of curves, we can note that at parallel linear and circular light beam polarization recording DE achieve far more higher values as at orthogonally light beam polarization. Taking in account, that for latest case intensity modulation is absent or nearly absent [12] we can suppose that we observe polarization (vector) holograms.

To ascertain on appearance of vector polarisation hologram in As-S-Se films, we inspected polarisation properties of  $\pm 1$  order transmitted as well reflected diffracted beams of s-p and  $\pm 45^\circ$  polarised 0, 6328  $\mu\text{m}$  light beam recorded gratings at different expositions. As we expected, in both cases we observed the properties characteristic for vector polarisation holograms: the restored diffracted beams - transmitted as well reflected shows the opposite polarisation than a reading beam. Nevertheless it should be note that observation and definition of PH nature is related with some difficulties. Firstly, it is due to very low diffraction efficiencies distinguishable at first maximum. Secondly, the specific reversibility take place, Fig. 9, – while reading, hologram is going very fast. And thirdly, with surface relief formation process phase hologram recording begins to dominate thus overlapping PH.

If compare s-p and  $\pm 45^\circ$  polarised light beam recording we should underline some difference between

both cases: 1) for s-p polarised light beam recording we achieved  $DE_{ref} \approx 2 \times 10^{-4}$  and  $DE_{tr} \approx 2 \times 10^{-3}$  for transmitted and reflected beams respectively. Meanwhile for  $\pm 45^\circ$  polarised light beam recording values  $DE_{ref} \approx 3 \times 10^{-3}$  and  $DE_{tr} \approx 8 \times 10^{-3}$  nearly an order of magnitude exceed the corresponding values for s-p polarised light beam recording; 2) PH properties i.e. reconstruction of appropriate recording beam polarisation for s-p recording we can observe not only up to achieving first diffraction maximum but even at beginning stage of surface relief formation process. For  $\pm 45^\circ$  polarised light beam recording polarisation properties we could behold only in very beginning stage of recording – at expositions less than necessary for achieving first diffraction maximum values. From analysis of recording conditions for these two cases under consideration we can conclude that for s-p polarised light beam recording tiny diffraction maximum in transmission mode and in reflection one at the beginning stage of recording curve belongs to PH and arises due to electric field modulation. For  $\pm 45^\circ$  polarised light beam recording with the electric field modulation slight light intensity modulation takes place too [12]. Presumably, that leads to stronger as PH scalar volume hologram recording what not allow visual checking of polarisation properties.

Using the method described in [18] for separation PH and surface relief gratings we carried out some experiment for circularly orthogonal polarized light beam recording. As it is possible to see from Fig. 10, slight difference in  $\pm 1$  order diffracted beam time dependence curves is possible to distinguish. We should note on similarity in course of recording curves shown in Fig. 9-2) for transmitted diffraction beam and Figs. 11–3: fast increasing up to some maximum values, decreasing and then more slowly growing again. In addition, values in first maximum are comparable –  $\sim 10^{-3}$ . From here we can speculate on presence of PH initiated by optically induced anisotropy. Decreasing and further part of curves presumably is related with surface relief formation process.

#### 4. Conclusions

The results for As-S-Se films demonstrate, that at wave-length  $\lambda = 0,6328 \mu\text{m}$ , scalar volume transmission, polarisation as well surface relief HG recording is possible to stimulate. What kind of HG will be dominating depends of light beam polarisation. If the polarisation for both recording waves takes the same direction (linear or circular), scalar transmission HG recording occurs. In this case light expositions necessary for achieving maximum values of diffraction efficiencies ( $DE(\text{max})$ ) not exceed  $50 \text{ J/cm}^2$ . At the opposite (orthogonal) light beam recording surface relief formatting takes place and we achieved DE up to 8% in the case of circularly opposite and  $\pm 45^\circ$  polarisations. Necessary light expositions for this process are rather high – up to  $10^4 \text{ J/cm}^2$ . AFM measurements have shown, that relief depth could achieve even more than  $0,8 \mu\text{m}$  depth. Nevertheless, at the same

expositions  $\leq 50 \text{ J/cm}^2$  as for linearly polarised light slight diffraction maximum at orthogonal light beam recording can be observed. Analysis for that part of recording curve indicates on presence of polarisation holographic gratings.

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\*Corresponding author: Mara.reinfelds@cfi.lu.lv