# Phase grating patterning by direct laser recording on $As_2S_3$-Se nanomultilayers

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**Abstract**

Direct laser recording of surface relief gratings as phase holograms on successive evaporated $As_2S_3$ and Se nanomultilayers was studied in this paper. The surface relief gratings patterned on $As_2S_3$-Se nanomultilayers were investigated in dependence on polarization states of recording beams. It was shown that phase gratings are formed on the surface of the $As_2S_3$-Se nanomultilayers under the two linear polarized beams falling on the sample surface at (+45°)-(- 45°) relative to grating vector and the left-right circular polarizations. The influence of photo-induced mass transport on the surface relief formation process is discussed.

*Keywords:* Direct laser recording; holography; chalcogenide glass; nanostructures; phase grating; polarization states.

## 1. Introduction

Development of active and passive elements of photonics, in line with micro- and nanoelectronics, requires new materials, structures, and new methods for manufacturing those elements. Chalcogenide glasses (ChGs) are an important class of amorphous semiconductors used in this area. Applications of ChGs have mainly been based on their transparency to infrared light (passive) and sensitivity to different kinds of irradiation (active). The latter produce pronounced structural changes in ChGs that are the more important case, regarding the challenges of understanding their microscopic nature and applications as functional materials of photonics.

One of most important optically induced effects is the direct, one-step process of the surface relief (SR) formation, which was closely related to the induced mass-transport (in vertical or lateral directions) in a glassy material under non-uniform illumination. The direct light-induced fabrication of the SR by the lateral mass transport on ChG films has been widely studied for several selected compositions [1] and appropriate mechanisms have been recently proposed [2].

Multilayer structures are simplest artificial nanostructures that can be rather easily fabricated with controlled geometrical parameters and investigated as thin films. A few approaches are known for extending investigations of ChG layers towards the nanostructures, especially in the nanolayered, superlattice-like multilayer structures, but the problem of photostructural transformation dependence on the artificial nano-structuring is still not solved. In addition, all of these applications of ChGs are required a chemical developments to form an SR, i.e. cannot be fabricated by a one-step direct laser writing.

Two main types of SR gratings (SRG) induced by holographic recording, due to the excitation light modulation with near band-gap light in ChGs, can be distinguished according to their formation mechanism and their properties: 1. Small scalar SRG induced by either volume expansion or shrinkage due to different responses of the material in the bright and dark zones of the interference pattern formed. 2. Giant vectorial SRG induced by a lateral mass transport when the light polarization of the recording beams has a component along the light intensity gradient.

From the fundamental point of view, photoinduced processes in ChGs are far from being fully understood according to the existing current opinion of specialists. Still polarization dependence of these processes is of both practical and theoretical importance. This work considers of the recording properties of $As_2S_3$-Se nanomultilayers is given in the context of its contribution towards the realization of the direct relief formation applicable for fabrication of photonic elements.

## 2. Materials and methods

Although stimulated structural transformations were observed in a wide range of chalcogenide glass compositions, we selected for experiments $As_2S_3$–Se nanolayered films as well as Se and $As_2S_3$ layers because of their known parameters and well-known technology. Important criteria were low softening temperatures and low crystallization abilities of $As_2S_3$, small effects of the direct photostimulated scalar surface relief formation within the amorphous phase.

Bulk glasses $As_2S_3$ were fabricated by common melt quenching method. Amorphous $As_2S_3$–Se nanomultilayers (NML) were prepared by computer driven cyclic thermal vacuum deposition from two isolated boats with $As_2S_3$ and Se on constantly rotated glass substrate at room temperature in one vacuum deposition cycle. Prepared NML samples consist of successive evaporated through the 2 masks $As_2S_3$ and Se nanolayers. The technology allows a films deposition control within the whole sample thicknesses in the range from 0.005 up to 3.0 μm. The monitoring and determining of the total NML film thickness was carried out during the thermal evaporation by 2 interference thickness sensors at $\lambda = 0.95$ μm in transmission mode [3]. This wavelength is not actinic for NML composition.

To prevent the crystallization of Se layers, which are rather structurally unstable under heating and/or illumination, we minimized the heating of layers during the deposition by virtue of substrate rotation and reduced boat temperature. No Se nanolayers crystallization inside NML was observed by X-ray control in samples after deposition and storage at room condition for about 3 years.

An interferometric holographic recording was used to record a grating on the NML $As_2S_3$–Se like described in [4]. The holographic gratings with a period of Λ =1 μm were recorded by two symmetrical angled laser beams with respect to the sample surface normal. The intensity ratio 1:1 of the recording beams was used in order to achieve maximum interference fringes contrast. The phase shift experiments were performed by angular turning the quarter wave plates in each optical path, therefore, we can change the state of polarization of both recording beams. The two interfering beams independently pass through phase turning quarter wave plates to provide a control over the polarization state of the writing beams. Interfering beams with P–P, S–S, L–R or (+45°)–(- 45°) polarization combinations were used for SRG recording.

The experimental set up is sketched in Fig. 1. The diffraction efficiency η was controlled in real time by measuring the first order LD intensity in the transmission mode. The diffraction efficiency was determined as $\eta = I_{dif.}/I$ 100%, where $I_{dif}$-diffractive beam, I-transmitted beam in 0 order. During holographic recording all the changes in NML volume and surface (like those in absorption and refractive index) were controlled by measuring the transmission diffraction efficiency. Thus, the cumulative changes in diffraction efficiency of NML $As_2S_3$–Se structure were measured.

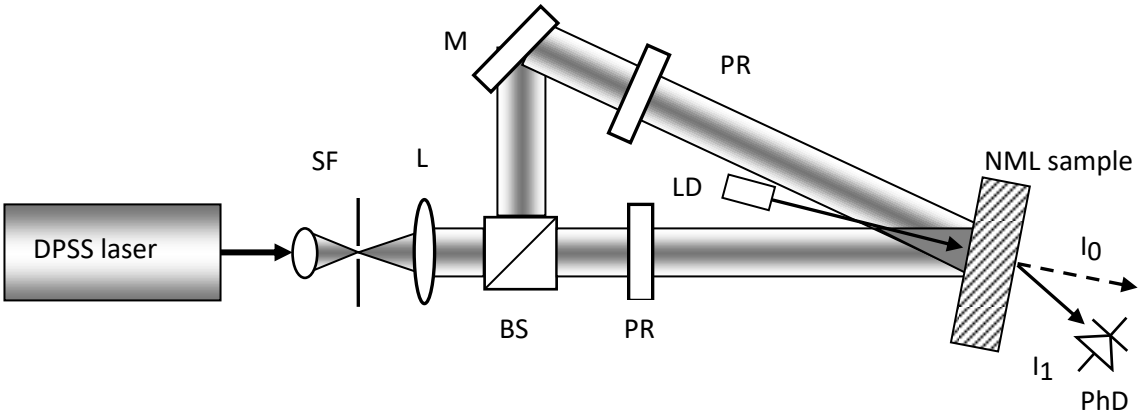


**Fig. 1**. Optical arrangement for holographic grating recording with real-time measurement of diffraction efficiency by photodetector. CW DPSS single mode laser (λ=0,532 μm, power =100 mW), M – mirror; SF – spatial filter; L – collimating lens; BS – non polarized beam splitter; LD – laser diode (λ=0,650 μm) for monitoring recording process; PR- polarization retarder; PhD – photodiode.

## 3. Results and Discussion

Diffraction gratings with 1μm period were recorded by two laser beams with S–S polarization with synchronous diffraction efficiency η measurement by LD light in the first diffraction order. Surface profile of holographic grating directly in one step recorded on NML $As_2S_3$–Se and measured by AMF is shown in Fig.2a, b.

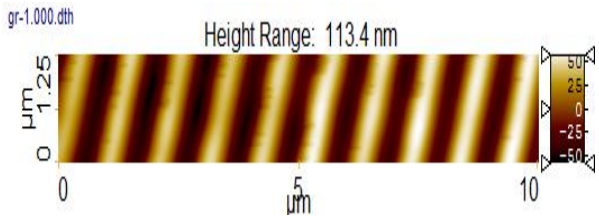


**Fig.2 a**. Surface of holographic grating recorded on NML $As_2S_3$–Se investigated by AFM.

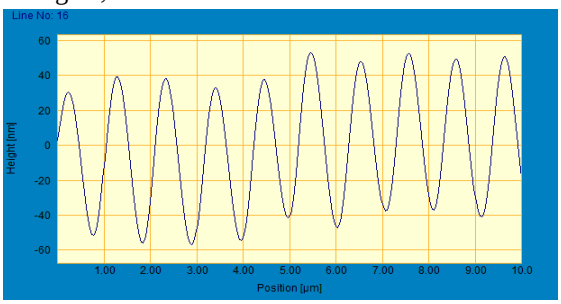


**Fig.2 b**. Profile of holographic grating (from Fig.7 a) directly recorded by two laser beams with S–S polarization on NML $As_2S_3$–Se surface.

It can be observed that grating recording on NML $As_2S_3$–Se provides high optical quality of the obtained relief with depth of the grating surface about 100 nm at total NML $As_2S3$–Se structure thickness 2500 nm and modulation period of layers N = 25 nm. It is very important to underline that in contrast to most communications about relief grating formation by laser beam in ChG films in our experiment, no wet etching was applied.

Considering the grating profile presented in Fig. 7, one can see that the thickness changes due to surface deformations of NML $As_2S_3$–Se is (Δd/d)100% = (0.1 μm/2.5 μm) 100% ≈ 4.0%. Changing of a-Se film structure under exposure shown in [3] can trigger processes of mass transfer in chalcogenide multilayer structure due to arising structural non-uniformity according to intensity distribution during grating recording. In our experiment S–S polarizations of recording beams does not create the conditions for the vectorial mass transfer, for which the polarization along the grating vector is needed. So we can assume that 4.0% depth of the grating recorded in NML As2S3–Se structure is mainly the result of scalar deformations.

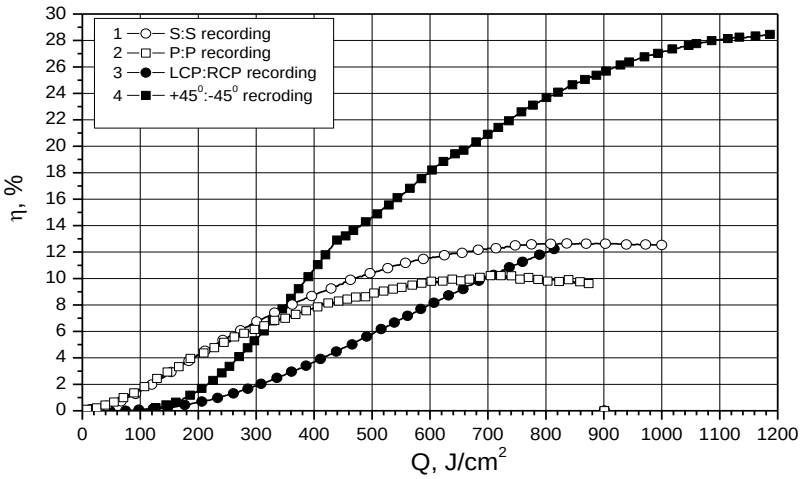


**Fig. 3.** Kinetics of holographic grating recording (grating period Λ=1μm) on NML $As_2S_3$–Se (D=2500 nm) for different light beam polarization: 1 – S:S parallel linear polarization, total light intensity 270 mW/cm$^2$; 2 – P:P parallel linear polarization, total light intensity 160 mW/cm$^2$; 3 – LCP:RCP orthogonally circularly polarization, total light intensity 220 mW/cm$^2$; 4 – +45º:45º orthogonally linear polarization, total light intensity 160 mW/cm$^2$.

Fig. 3 shows the typical diffraction efficiency η changes vs. dose value of illumination at different states of the recording beams polarizations. The light polarizations in this experiment were parallel to the grating vector (P-polarization), perpendicular to the grating vector (S-polarization), the left–right circular polarization (L–R), and the linear polarization at (+45º) direction in one writing beam and (- 45º) in the other one. Under the polarization modulated holographic recording, the two linear (P–P and S–S) polarized beams produce the similar surface relief deformations but with a different profile depth. The two linear polarized beams falling on the sample surface under angles of polarization (+45º)–(- 45º) produce the largest variation in the surface relief according to our measurements.

From Fig. 3 it is seen that not only the maximal shown diffraction efficiency η, but the kinetics of grating recording process depends on the state of polarization of the recording beams. For S–S and P–P polarizations the recording process begins just after turning on the laser illumination. It was found out that at doze of 700 J/cm$^2$ the diffraction efficiency reaches the saturation for pointed polarizations.

The control of polarization properties of recorded phase gratings has shown the changing of polarization planes at first diffraction order of transmitting light.

## 4. Conclusion

The direct one step grating surface relief formation in NML $As_2S_3$–Se structures are shown with the diffraction efficiency of 28% in value obtained at the wavelength 650 nm at (+45º)–(- 45º) orientations of polarizations. The dependence η vs. dozes for NML $As_2S_3$–Se does not reach the saturation of 1000 J/cm$^2$ and η = 25% at 800 J/cm$^2$. During the recording process on NML $As_2S_3$–Se the rate of g is slower due to mass transfer, which we suppose takes place. This process is obviously slower then the other components of photostructural changes. The possible explanation of the results obtained is under discussion, but a follow-up study is needed.

Direct one step grating recording simplifies greatly the processes of implementation of the optical elements. The obtained optical quality of relief surface is very important for realizing diffraction optical elements by printing or master hologram realization.

The creation of different nanolayered structures opens new possibilities for tuning the basic optical parameters and for stimulated structural changes which, in turn, can be used for the development of direct, one-step amplitude-phase optical recording and fabrication of the surface relief structures.

## Acknowledgements

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