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Light intensity and its polarization relation to the photo-induced mass movement in thin layers of chalcogenide vitreous semiconductors

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This work is devoted to the topical issue– photo-induced formation of surface relief gratings (SRG) in thin layers of chalcogenide vitreous semiconductors (ChVS).This direct surface-relief formation during light illumination phenomenon is being discussed with special attention focused on the polarization and intensity of the corresponding light. Holographic recording setup and illumination through adjustable optical slit are used and theoretical model for light interference pattern has been built. We have showed that the efficiency of the surface relief formation strongly depends not only on the writing beam properties but also on the assisting beam. Also SRG formation and mass transfer processes which are based on the photo-induced plasticity on As_2S_3 are discussed in this paper.

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1. Introduction

Photo-lithography is one of the most common methods from all types of lithography for surface-relief structure patterning. Light induced changes of the chemical properties in a resist material enable a selective removal of regions on a thin film or on the bulk of the illuminated material by developing and etching (*e.g.* [1]). Comparatively recently a number of materials have been studied for direct surface-relief formation during the exposure process [2-5]. The physical interpretation of this incompletely understood phenomenon of light and matter interaction is interesting for many researchers in the field of soft materials like ChVS [6-9] and azo-polymers [7, 10-12]. There is a wide range of techniques used for an interpretation of the anisotropic surface-relief modulation process: some of them based on the intensity of light [*e.g.* 12], others on the direction of the electric field oscillations [*e.g.* 6]. Since the intensity of light is proportional to the squared module of the electric field, the light field experiments follow from the electric field induced anisotropic surface-relief modulation experiments but not vice versa. Thus it is necessary to discuss this anisotropic surface-relief modulation process from the point of view of the electric field.

Focused Gaussian beam experiments [*e.g.* 10] where the anisotropic mass transfer process depends on the direction of polarization are doubtful – there is a continuous mass transfer during illumination thus the adjusted focus moves into or out of the matter and the results (as shown in the [13]) can be interpreted incorrectly. The same applies to experiments where the focus is adjusted incorrectly from the beginning (distance from the surface comparable to wavelength of the writing beam). Therefore, it is necessary to use other experimental methods for a more precise investigation.

In the last few years researchers have been studying different kinds of anisotropic light induced deformations in the light sensitive materials. Under a uniformly distributed light illumination anisotropic T or X shaped scratch deformations show that only particularly oriented scratches tend to change their shape. These deformations change differently in different materials: chalcogenides show deformations in the direction parallel to the electric field vector [14] and azo-polymers – orthogonal to the electric field vector [15]. The same occurs in the experiments (shown by Tanaka [9] in chalcogenides) where uniformly illuminated free-from-substrate flakes tend to curl upwards only in the direction parallel to the electric field vector. This phenomenon can be explained by the anisotropic fluidity in the particular direction which is induced by the polarized light. The release of the tension forces in that direction leads to the flake curling in the perpendicular direction.

A new kind of investigation and interpretation of anisotropic light induced, polarization dependent deformations in ChVS are discussed in this paper. To fully understand this mass transfer process a theoretical model of the light interference is obtained. For these experiments a holographic recording setup and its modified version (one beam illumination through optical slit) are used.

2. Experimental

Amorphous As_2S_3 films were obtained by thermal evaporation in vacuum of ~$5\cdot10^{-6}$ Torr onto glass substrates. The thickness of the film was controlled in a real time by a 650nm diode laser and it was from 1 to 7μm.

One of the most common methods to investigate the surface relief grating (SRG) formation is direct holographic recording technique, thus high light gradient can be obtained maintaining constant interference pattern in the z-direction.

Recently ([2] in 2010) a huge SRG formation dependency from chosen light polarization and other factors was shown. For example, outstanding SRG occurs only in particular combinations of the writing beams properties: unusual polarization combinations (*i.e.*, orthogonal: -45 and 45 degree, left/right handed circular: LC and RC) gives the best impact on the SRG formation efficiency, *i.e.*, on a ratio of the produced surface-relief versus the time. Therefore it is necessary to discuss polarization dependent interference of the two coherent beams theoretically and compare its impact on the SRG formation. That will be discussed in this paper.

Other group of experiments that will be discussed in this paper contains one unfocused beam writing system (see scheme in Fig.1.). This recording system allows producing of each individual grating from the gratings made by direct holographic recording system. One beam exposure through the slit as close as possible to the surface of a sample reduces the diffraction scattering effect and enables to investigate SRG formations dependency on light intensity and its polarization to the individually separated grating. To better understand the involved physics and increase the recording efficiency (like in [2]) extra illumination, *i.e.*, incoherent assisting beam was also used in this recording system. For the writing beam a 532nm Nd:YAG Coherent Verdi 8W laser and for the assisting beam other incoherent 532nm diode laser were used.

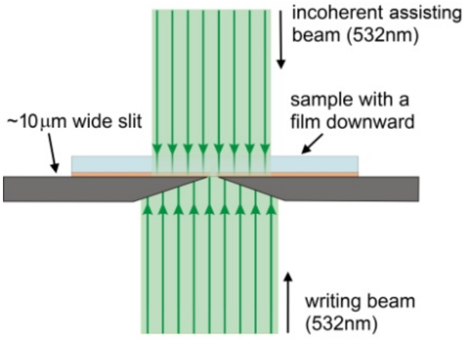


Fig.1. Holographic recording simulator, i.e., one unfocused beam illumination through adjustable optical slit enabling to produce individual grating

3. Results and discussion

According to [2] it is possible to arrange various polarization combinations by impact on direct SRG formation quality quantitatively through reflection diffraction efficiency (η_R) which means: the worst for s:p (writing beams are with s and p polarization), poor for p:p, s:s, 45°:45°, LC:LC and the best for 45°:-45°, LC:RC setup. An interesting thing (as to [2]) is that there were no polarization combinations which could give an impact on the SRG formation as *'good'* ones, in other words, no middle quality recording was possible. For future investigations this polarization-driven relief modulation phenomenon should be discussed theoretically.

The scheme for those theoretical experiments is chosen the same as it was for the direct holographic SRG recording experiments discussed above. Therefore, the Bragg equation defines the angle 2α between the corresponding k-vectors for 1μm interference period Λ by using a bandgap 532nm light:

$$\Lambda = \frac{\lambda}{2\sin\alpha} \tag{1}$$

By taking solution for a scalar wave equation in a homogeneuos medium:

$$w = A \cdot \cos(k \cdot s - \omega \cdot t) \tag{2}$$

where A – amplitude; k – wave number; s – one dimensional coordinate; ω – angular frequency; t – time, it is possible to define a vertical polarized wave that propagates along y-axes:

$$\vec{w} = \{0, s, w\} \tag{3}$$

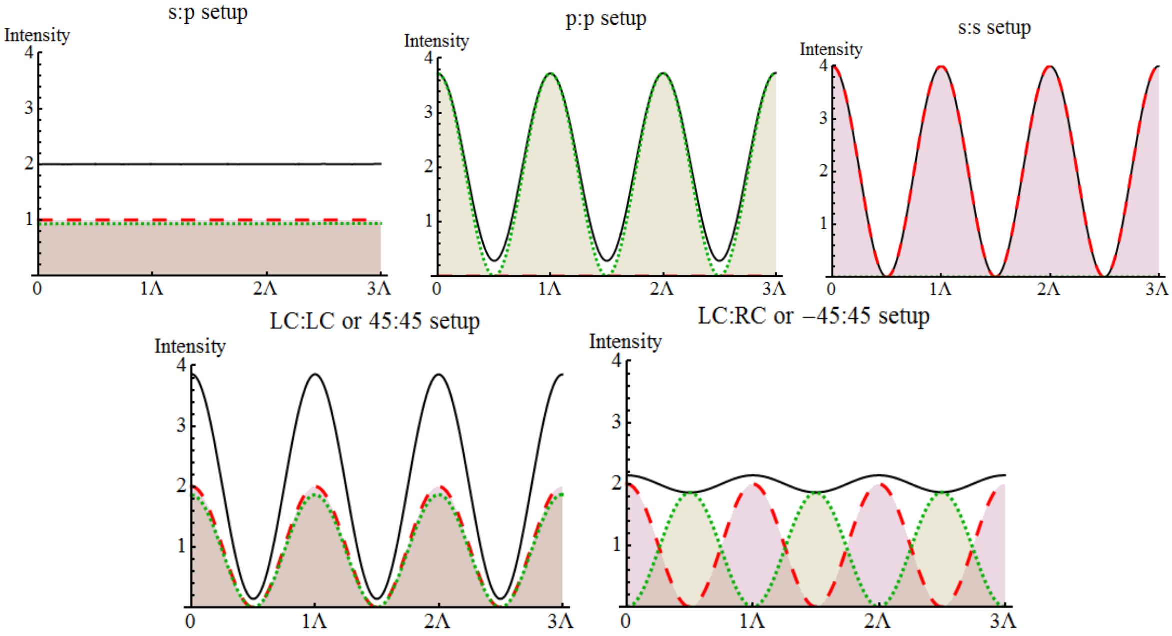
or horizontal polarized wave that propagates along y-axes:

$$\vec{w} = \{w, s, 0\} \tag{4}$$

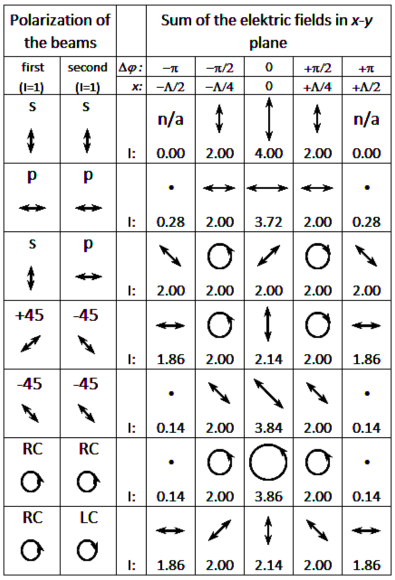
Together with the coordinate transformation matrix and a data visualization program (*e.g.* Wolfram Mathematica) it is possible to build a theoretical model of the polarization dependent interference.

Visualized polarization dependent light intensity distribution of two coherent beams interference is shown in Fig.2a. We can see that the strong light gradient which is present in most of the polarization combinations (*i.e.*, s:s, p:p, 45°:45° and LC:LC setup) is not the main factor in obtaining SRG. So, why is the recording efficiency and therefore the direct recording possibilities in s:p case the worst although in a similar case where the intensity is also uniformly distributed (45°:-45°, LC:RC in Fig. 2 (a)) the recording efficiency is excellent? The answer is in the polarization distribution. Since intensity of a light is proportional to a squared module of the electric field, instead of uniformly distributed intensity the direction of the electric field vector may vary. It is shown in Fig.2a. where two of the intensity distribution components are highlighted: dotted and dashed curves with corresponding masks under them correspond to the intensity from the polarization directions parallel (p-direction) and perpendicular (s-direction) to the plane of incidence respectively. Note that there is also the third component of intensity distribution, which is with radial direction (orthogonal to the s and p direction) but it is comparatively small and not pointed out. Besides theoretical polarization distribution of the two coherent beams interference versus polarization combinations of the interfering beams are shown in Fig. 2b. There we can see not only absolute values of the intensities of different phase shifts Δφ (-π; -π/2; 0; π/2 and π) but also corresponding polarization distribution for all combinations of the interfered light polarizations.

Now if we compare the previously arranged polarization combinations by impact on direct SRG formation quality with theoretically calculated polarization components of the intensity distribution in Fig.2a., it is clearly visible that for the best direct recording possibilities (45°:-45° or LC:RC setup) instead of high intensity gradient electric field gradients– s and p components with are a half-cycle out of phase is needed.



(a)



Polarization of the beams			Sum of the elektric fields in *x-y* plane				
first (I=1)	second (I=1)	Δφ: x:	−π −Λ/2	−π/2 −Λ/4	0 0	+π/2 +Λ/4	+π +Λ/2
s ↕	s ↕	I:	n/a 0.00	↕ 2.00	↕ 4.00	↕ 2.00	n/a 0.00
p ↔	p ↔	I:	• 0.28	↔ 2.00	↔ 3.72	↔ 2.00	• 0.28
s ↕	p ↔	I:	⤡ 2.00	○ 2.00	⤢ 2.00	○ 2.00	⤡ 2.00
+45 ⤢	-45 ⤡	I:	↔ 1.86	○ 2.00	↕ 2.14	○ 2.00	↔ 1.86
-45 ⤡	-45 ⤡	I:	• 0.14	⤡ 2.00	⤡ 3.84	⤡ 2.00	• 0.14
RC ○	RC ○	I:	• 0.14	○ 2.00	○ 3.86	○ 2.00	• 0.14
RC ○	LC ○	I:	↔ 1.86	⤢ 2.00	↕ 2.14	⤡ 2.00	↔ 1.86

(b)

Fig.2. (a) Theoretical light intensity distributions (solid curves) for two coherent beams interference (a:b symbols denote a polarization combination for the writing beams), dashed and dotted curves corresponds to the s and p polarization components of the intensity respectively; intensity of the interfered light $I_1=I_2=1$ unit, Λ- period, angle between corresponding k-vectors $\alpha=30.86^o$ (i.e., $\Lambda=1\mu m$, $\lambda=532nm$); (b) Theoretical polarization distribution at the two coherent beams interference versus polarization combinations of the interfering beams; period $\Lambda=1\mu m$ and intensity of the interfered light $I_1=I_2=1$ unit, angle between corresponding k-vectors $\alpha=30.86°$

In case of just one s or p component electric field gradient (s:s or p:p setup) or when both the components are in phase (45°:45° or LC:LC setup) or without gradient (s:p setup) it is possible to obtain rather poor SRG structures. These facts lead to some conclusions: s and p polarized light gradients with a half-cycle out of phase help each other to form a SRG, *i.e.*, on both of the periodic gradients the matter interacts differently. That is the reason why intensity distributions where both the components are in phase (LC:LC or 45°:45° setup) or without one of the components (with periodic unexposured areas like it is for s:s or p:p setup) do not give outstanding results for the direct SRG recording.

Experiments show that for some polarization combinations of the writing beams it is possible to dramatically increase the recording efficiency by optional illumination during direct SRG formation process [2]. Extra incoherent light does not interfere with the light from the writing beams and therefore it just raises the total light intensity distribution. The idea of this process is as follows: by using extra illumination to make intensity distribution as it is for 45°:-45° or LC:RC setup with a half-cycle out of phase for s and p intensity components, therefore to make recording efficiency as good as it is for the above mentioned setups. And as expected, extra illumination with polarization orthogonal to the polarization of the interfered light gives the best SRG formation enhancing effects. For example, p:p polarization setup previously arranged as *'poor'* one combination for formation of the SRG, now with extra s polarized light illumination is as good as it is for 45°:-45° or LC:RC polarization setup with or without extra illumination. In fact, extra illumination for 45°:-45° or LC:RC setup just decreases SRG formation efficiency [2]. This fact again leads to different influence on the mass movement process by s and p polarized light gradients respectively.

Due to the mass transfer, direct SRG formation experiments are possible in soft materials where light can transfer the mass to some particular places. Since the period of the holographically produced SRG is always sinusoid-like, how can it be that the geometry of the produced SRG by using p:p or s:s recording setup is the same as it is for the polarization combinations which give impact on the sample as it is for p:p and s:s setup recording simultaneously with a half-cycle out of phase distribution (as in Fig.2b. for 45:-45 or LC:RC setup)? Thus there are just two mass transfer versions possible: the mass moves parallel to the intensity gradient and in a different direction according to polarization of the intensity gradient. Therefore the best SRG were obtained with 45°:-45° or LC:RC holographic setup– s and p intensity components (Fig.2a.) which are a half-cycle out of the phase, *'help'* each other to form a ditch and a hill at the same time. To investigate this polarization dependent influence on the mass transfer process a new kind of recording system is needed where we can link the coordinate system of the substrate with one of the AFM measurements. Due to the periodicity all the obtained surface-relief structures on an AFM will look the same no matter what kind of mass transfer processes were participating. By modifying the holographic recording setup of the two beams for more precise experiments a recording setup for individual SRG period was made (Fig.1.). Using one beam illumination through the optical slit enables to determine the impact of individual electric field gradient on the mass transfer process. Therefore by illuminating a sample through ~10μm wide slit by light with polarization parallel (s-direction) or perpendicular (p-direction) to the slit will enable to investigate polarization dependency of light gradient impact on the matter. Unfortunately these experiments did not show any considerable mass transfer thus an incoherent assisting beam with the same wavelength (532nm) was used.

The results (Fig.3.) confirm that for writing beams with different polarization the transfer of the mass process is different– for s polarization (s/p setup: polarization of the writing beam is along the slit) the mass has been transported away from the illuminated area thus forming a ditch, but for p polarization (p/s setup) vice versa– the mass has been transported into the illuminated area thus forming a hill. Note that orthogonally polarized assisting beams were used for both these cases thus making the recording environment more like it is for LC:LC or 45°:45° holographic recording setup (symbols *a/b* denote polarization for light that is going through the slit and polarization for extra illumination respectively). For the assisting beam with polarization in the same direction as for the writing beam (p/p setup in Fig.3.) the obtained gratings also formed a hill (like for p/s setup) but with more than 20 times weaker amplitude than the corresponding p/s setup. The amplitude of the gratings obtained by illumination through the slit is strongly linear with the time of the exposure (Fig.3.) but as follows from the slope coefficients formation of hills instead of ditches is slightly effective.

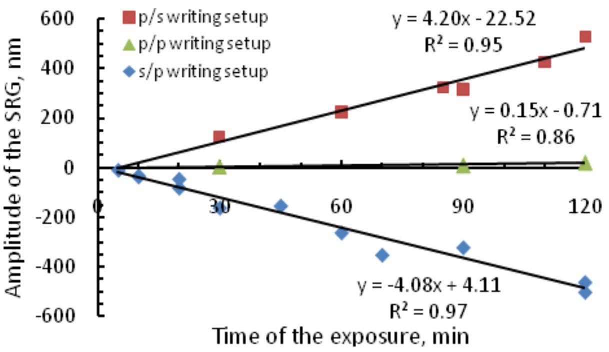


Fig. 3. Amplitude of the SRG versus time of the exposure for different polarization combinations of the writing and assisting beams (a/b symbols denote polarization for the writing and the assisting beams respectively) for optical slit experiments, intensity of the writing beam equals to $4.24 W/cm^2$ and for the assisting beam $0.37 W/cm^2$, recording performed on a $3.3 \mu m$ thick As_2S_3 film.

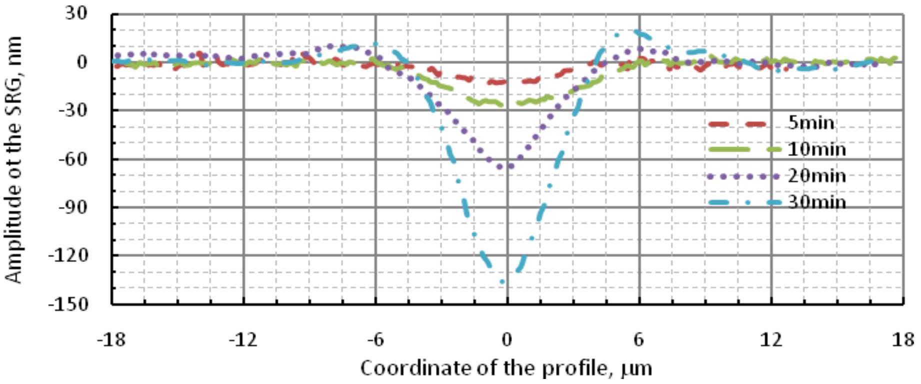


Fig. 4. Amplitude of the SRG obtained by AFM versus time of the exposure (5, 10, 20 and 30 minutes) for 4.24W/cm^2 writing and 0.37W/cm^2 assisting beam, recording performed on a 3.3µm thick As_2S_3 film, the electric field vectors of the writing and the assisting beams are parallel and perpendicular to the optical slit, i.e., s/p setup respectively

Comparing the profiles of the gratings obtained by the s/p setup for different exposure time (5, 10, 20 and 30 minutes in Fig.4.) it is clearly seen that the mass transfer in this case starts from the middle of the slit. For illumination more than 20 minutes (20, 30, *etc*) due to active mass transfer small hills were formed along the ditch thus forming a M-shaped profile. This fact confirms the theory that a long impact of the polarized light gradient on a photo-resist acts as a mass transfer in a specific direction.

4. Conclusions

The main conclusion that can be drawn from the results is that the mass transfer process for ChVS depends on the polarization of the light. It depends on the relation between the polarization and the electric field gradient of the writing beam: if the directions from both of them match, the mass will be transported into the electric field gradient those forming a hill, otherwise (both the directions will be orthogonal) forming a ditch. By using incoherent assisting beam (best results for cross-polarized assisting beam) it is possible to rise the efficiency of the mass transfer during the writing process.

A direct SRG recording technique is a comparatively new solution for lithography and as shown in this article, provides new experimental techniques for better understanding of the interaction between light and matter. The obtained gratings are very stable at room temperature, so this method can replace some of the chemical etching techniques and find practical application in the applied physics.

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