Photo-induced formation of surface relief in amorphous As_2S_3 films

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Abstract. In this report we study the formation of surface relief in arsenic trisulfide films under laser light radiation with modulated polarization direction. The formation of surface relief is obtained with method what is different from holographic recording - only one laser beam with modulated polarization direction was used. Samples with different thickness were made using vacuum evaporation. The photo-induced changes on the surface were initiated by DPSS laser light (λ = 532nm). Profiles of surface reliefs were observed and relief depth dependence on film thickness and exposure was studied.

1. Introduction

Structure like surface relief grating (SRG) can be used as diffractive optical element and as Bragg reflector. The new method was used to study SRG formation in chalcogenide films. Well-known chalcogenide – amorphous arsenic trisulfide was used for this purpose. As_2S_3 is known as inorganic polymer, which is often used in holography due to large photo-induced changes of its chemical and optical properties [1.2.3]. It has been studied since 1950', a lot of interesting optical phenomena [1.2.3.4.5.6] were discovered, but still there are things to discuss. Photo-induced optical anisotropy such as birefringence and dichroism has been studied before [6] and is closely connected to photo-induced mass transport. Studying SRG formation process mainly consists of finding appropriate conditions for efficient photo-induced mass transport. Surface relief can be obtained with holographic record as well as by illumination through optical slit [8.9], but this study is based on one beam record. Since there is no interference, set-up is stable to vibrations and there is no need in high coherence. Experimental set-up consisting of polarization direction modulator is shown in Figure 1.

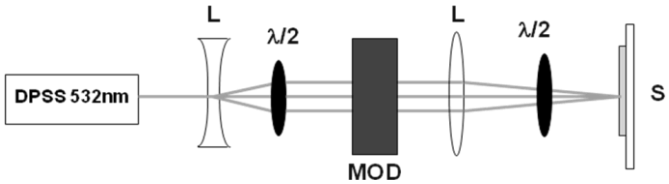

Figure 1. Experimental set-up for recording surface relief grating. **L** - lens; **λ/2** – half-wave plate; **MOD** – polarization modulating element; **S** – sample. Recording was performed with DPSS laser λ = 532nm.

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2. Experimental

Surface relief gratings (SRG) are usually obtained with holographic method based on two beam interference [8.9]. Depending on recording beam polarization direction it can provide an interference pattern with light intensity or polarization modulation what is necessary for SRG formation. In this study we observe formation of surface relief, which is obtained with new method, different from holographic recording. The method is based on modulating light polarization direction in one laser beam. Intensity modulation pattern, what forms on samples surface, is quite similar to interference pattern. By using MOD and half-wave plate polarization is modulated according to already known configurations. For example, the S-S polarization modulation is a result of two linearly polarized wave's interference. Modulated polarization directions in one laser beam and its analogy in holography are shown in Figure 2.

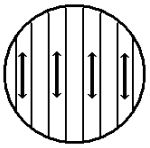
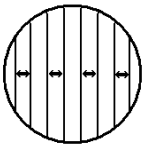
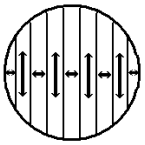
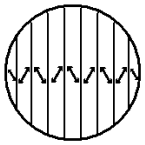
	S-S	P-P	S-P	45^0; -45^0
1				
2	Interference of two S-polarized waves	Interference of two P-polarized waves	Interference of R and L (or 45^0; -45^0) polarized waves	Interference of S and P-polarized waves

Figure 2. Simplified scheme of polarization distribution; ↕ - electric field vector oscillation direction.
1 – Modulated polarization direction in one laser beam.
2 – Similar distribution with two beams (holographic analogy).

For direct SRG record amorphous arsenic trisulfide films where used as photoresist. Samples with different thickness (from 1μm to 10μm) were prepared by vacuum evaporation onto glass substrate. Surface reliefs were recorded using modulated DPSS laser beam with 532 nm wavelength (Figure 2.). Intensity of laser beam was constant during the recordings – 3,77 W/cm^2. SRGs were recorded with a constant period – 50 μm. Depth and profile of recorded SRG was measured with profilometer. For polarization modulation change the half- wave plate is rotated, but in case of S-S or P-P configuration polarization cube is inserted after the second half-wave plate.

3. Results and discussion.

First, SRG recording with different polarization modulations was studied. Recording with all four configurations gave positive results (Figure 3.), but SRG recorded with S-P modulation was the deepest.

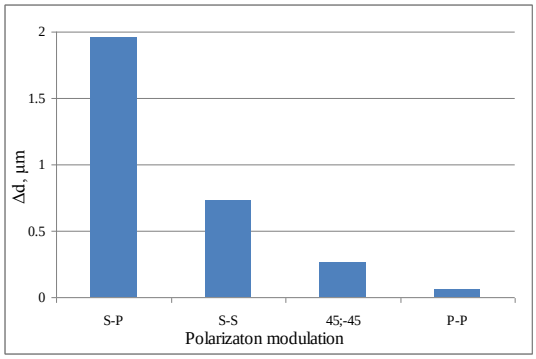
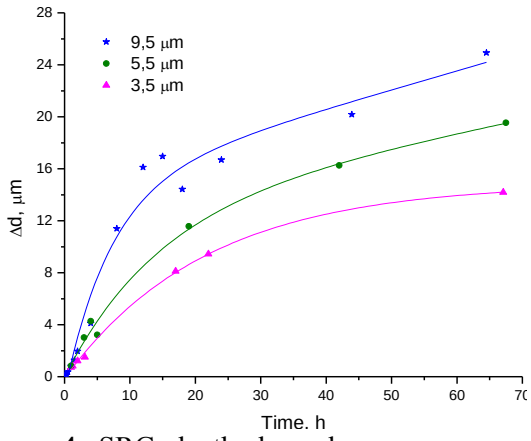


Figure 3. Δd – SRG depth in micrometers. Recording beam intensity – 3, 77 W/cm^2; Exposure time – 2h.

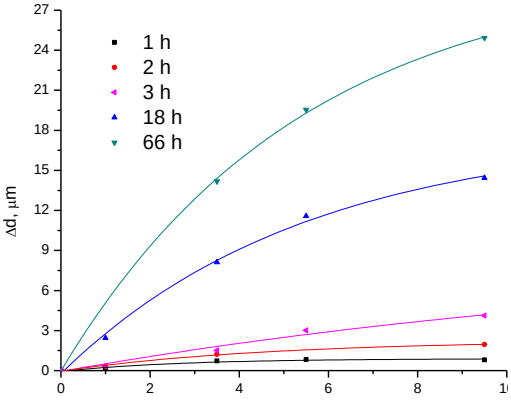
Intensity distribution is the same for S-P and 45^0;-45^0, but the results differ ~8 times. S-S and P-P configurations are similar, but still there is a big difference in results. It means that electric field vector

oscillation direction is principal photo-induced mass transport mechanism. In SRG formation process polarization modulation plays the key role. S and P – polarizations are orthogonal, so the electric field gradient has biggest value. It is electric field gradient [8] what induces mass transport, but mass transport direction depends on polarization direction. The velocity of mass transport process is not constant and growth of SRG is not infinite process. SRG formation process, increasing exposure, is shown in Figure 4. SRG formation process consists of two parts: linear, fast growth (t = [0..12]h) and saturation process, the part of slow growth(t = [12..70]h). It supposes to reach the maximum, i.e., biggest possible SRG depth value and after that growing stops. Thinner films are most likely to reach the maximum faster, because there is same amount of received energy, but less mass to transfer. Form of graphic, process tendency, is the same for all three films with different thickness. Recording SRG in 1 μm thick film is possible at smaller exposure or intensity. At 10 h exposure with 3,77 W/cm^2 intensity SRG starts to disband. Damage of SRG can be caused by large amount of received energy, illuminated area heating or large period influence.



Equation	$y = A_1 e^{-\frac{x}{t_1}} + A_2 e^{-\frac{x}{t_2}} + y_0$					
	* 9,5	±Δ	● 5,5	±Δ	▲ 3,5	±Δ
y_0	14.9	3.0*10^{-16}	138.2	11.9	39.2	2.0
A_1	-7.5	7.9*10^{-9}	-123.6	11.9	-11.8	0.1
t_1	21.7	2.8*10^{-8}	800.2	81.4	13.5	0.1
A_2	-7.5	7.9*10^{-9}	-15.8	1.9*10^{-2}	-27.4	1.8
t_2	21.7	2.8*10^{-8}	7.0	1.1*10^{-2}	201.5	19.5
R^2	1		1		1	

Figure 4. SRG depth dependence on exposure. Δd – SRG depth in micrometers; samples with thicknesses 3,5 μm, 5,5 μm and 9,5 μm were used; recording beam intensity – 3, 77 W/cm^2. Second order exponential decay function is used to fit experimental data.



Equation	$y = Ae^{-\frac{t}{x}} + y_0$			
Coeff.	A	t	y_0	R^2
▼ 66 h	-30.23	5.40	30.20	0.99966
±Δ	1.11	0.41	1.13	
▲ 18 h	-18.01	5.49	17.78	0.9975
±Δ	1.34	0.89	1.42	
◀ 3 h	-9.55	15.71	9.41	0.98172
±Δ	7.69	17.11	7.82	
● 2 h	-2.32	4.74	2.28	0.99591
±Δ	0.24	1.25	0.26	
■ 1 h	-0.97	2.66	0.90	0.93978
±Δ	0.19	1.53	0.17	

Figure 5. SRG depth dependence on film thickness at different exposures; Δd – SRG depth in micrometers; b – film thickness. Exponential fit is used.

Depth of recorded SRG depends on films thickness, too. SRG formation process versus film thickness is shown in Figure 5. Process is exponentially growing. To evaluate the effectiveness of each

thickness, the ratio $\frac{\Delta d}{b}$ was calculated. Most efficient film thicknesses are from 3,5 μm to 5,5 μm, with value of ratio 0,21 (1 h exposure) – 4,05 (66 h exposure). For thicker films, as there is a lot of mass to transport, process is slower and they are efficient at long exposures. Deeper SRG can be recorded, but it takes more time. Example of recorded SRG and its profile are shown in Figure 6.

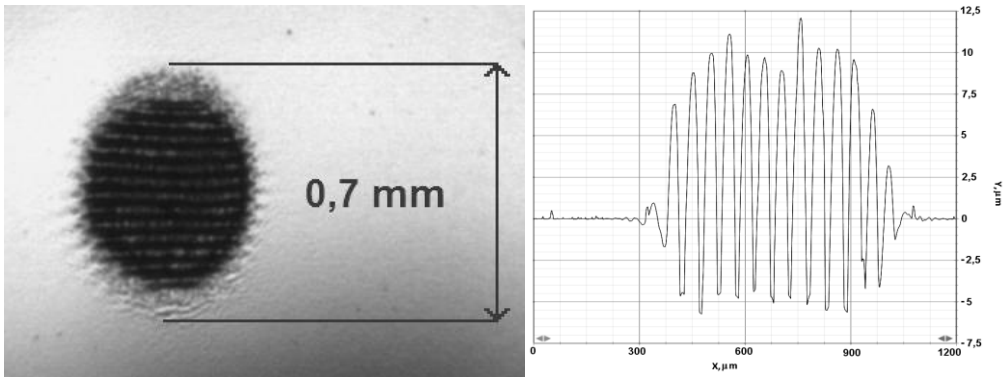


Figure 6. Picture and profile of 17 μm deep SRG recorded in 5, 5 μm thick film. Measured with profilemeter.

4. Conclusions

The new method is applicable for studying photo-induced mass transport in chalcogenide films. It is easier and cheaper way of recording SRG then holographic record. The quality of SRG is lower, but for research purposes this method is very good. For efficient recording S-P polarization is necessary. Recording beam intensity may vary but for thinner films ~1 μm it should be less then 3,77 W/cm^2 to prevent damaging of SRG. Optimal film thickness for SRG recording is ~ 4,5 μm.

Acknowlegements.
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