Surface relief formation during holographic recording

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In this report the study of direct holographic recording of the surface relief gratings on amorphous chalcogenide thin (0.5-10μm) films is presented in view of the light polarization state. Recording was performed on As_2S_3 by 532nm wavelength laser light. Because of direct surface relief formation, its efficiency may also depend on the softening temperature of the film material, which was studied using additional incoherent laser light for extra illumination during holographic recording. The results have shown that the efficiency of surface relief formation is many times higher in the case of extra illumination by additional incoherent light during recording. The mechanism of the direct recording of surface relief on amorphous chalcogenide films based on the photoinduced plasticity is discussed.

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1. Introduction

The key element for the production of surface relief holographic optical elements is photoresist or light sensitive material. The changes in local chemical properties, optical parameters, microhardness, viscosity and some other characteristics [1-3] induced in a resist material by light or e-beam exposure enable the surface relief structuring by *wet* or *dry* etching. Therefore this process includes two steps: recording and development by etching. Due to polarization-dependent anisotropic changes in resist material, such as opto-mechanical [7], M-shaped [8], or anisotropic crack [9] deformations during exposure it is possible to obtain a structured surface relief optical elements without etching.

A number of organic and inorganic materials have been studied for direct surface relief formation during a process of exposure by light or e-beam [4-6]. This is very promising for practical applications, since makes it possible to simplify the technology of surface patterning.

Apart from that, we report here the surface relief formation during holographic recording in amorphous chalcogenide semiconductor thin films. A number of studies have been carried out on the photoinduced structural transformation in amorphous chalcogenides during the past few years [4, 10, 11], but in this case particular attention is given to the polarization state of exposure light.

Intensity distribution of the two coherent equal intensities P polarized beams interference we can approximate by *sin* function: $I_{local}=I_{max}\cdot sin(2\cdot\Lambda\cdot\pi\cdot x)$, where Λ- period of the light interference and *x*- profile coordinate. From this formula can see that the local intensity of the light periodically vary from zero to I_{max}, i.e. exist places where light intensity equals to zero. In case of holographic recording and direct surface relief formation during the exposure process that mean that always exist local unexposured places on the sample whose do not participate in surface relief formation process and moreover- disturb relief formation efficiency. If and how we can change this situation? What happens if we extra illuminate sample by additional incoherent laser light with intensity I_0 during recording process? We keep the same local light gradient and lift up total local intensity in the range from I_0 to I_0+I_{max}- exposure also places where local intensity of the recording beams are equal to zero. In this way decreasing softening temperature and probably increasing efficiency of the surface relief formation during holographic recording. Since there is a direct surface relief formation, its efficiency can also depend on the softening temperature of the sample. The main task of this report was to investigate this dependence using additional incoherent laser light for extra illumination during holographic recording.

2. Experimental

Amorphous films were obtained by thermal evaporation in vacuum onto glass substrates. Thickness of the sample was controlled in real time by diode laser and it was from 0.5 to 10 µm.

The surface relief formation experiments were performed using a holographic recording system (see scheme in Fig. 1a.) where *Verdi6* laser 532nm wavelength light was used for recording. Diffraction efficiency (η) was controlled in real time by measuring the 1st order intensity of the reflected and transmitted recording beams. During the holographic recording by measuring the transmission diffraction efficiency (DE) η_T the changes in volume were controlled (like those in absorption and refractive index) and by measuring the reflection DE η_R – the changes in surface relief modulation. The volume gratings also affect the reflection DE, however their contribution is insignificant (see [2]) and will not be discussed here.

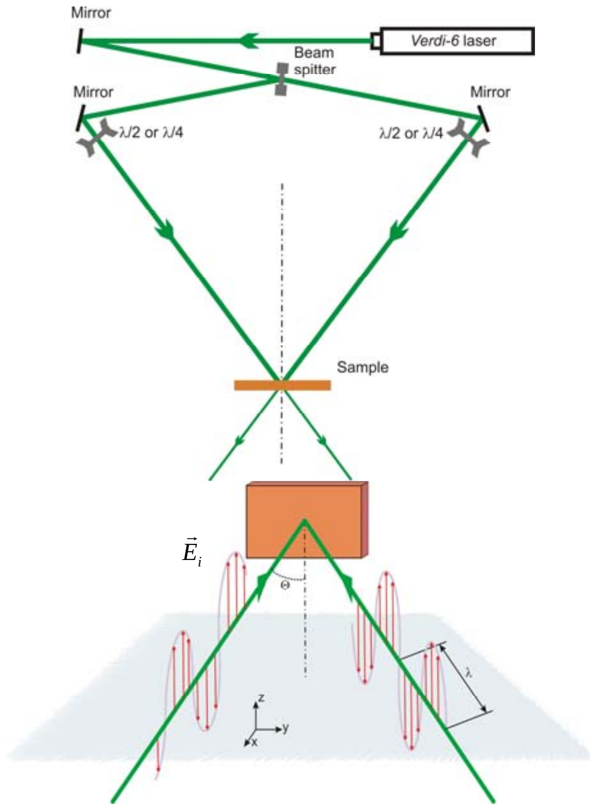


Fig. 1. (a) Experimental setup for the holographic recording experiments, recording was performed by YAG Verdi-6 532nm laser, (b) polarization is defined by x-y plane where both recording beams are located- in this case both laser beams are vertically polarized i.e. are in S polarization state.

The recording efficiency is defined as the slope of the 1st order polynomial equation $\eta_R=c_0{\cdot}t$, i.e. $c_0=\Delta\eta_R/\Delta t$, here t is time of exposure. The enhancement of relief formation is defined as the efficiency of recording with extra illumination divided by that without extra illumination:

$$(\eta_R)_i/(\eta_R)_0$$

The polarization state is defined by the plane where the electric field oscillates *vs.* the x-y plane where both recording beams are located (see Fig. 1b). Therefore, if both planes have the same orientation this means that the light has a P-polarization state; otherwise, if both the planes are at an angle to each other (e.g. 10^o) we say that the light has a 10^o polarization state; if this angle is 90^0, there is S polarization of the light.

3. Results and discussion

Fig. 2 shows typical diffraction efficiency changes with time (up to dashed vertical line). Like the absorption and refractive index photoinduced changes in resist material are comparatively fast process, transmission diffraction efficiency (DE) η_T reach their maximum (~55%) very fast. At the same time with η_T decreasing, reflection DE begin grown linearly- the light starts forming surface relief structures. And at the time with surface relief amplitude increasing (η_R is growing up), because of the extra way of length what disturb transmission DE- η_T continue decrease. Does this process is reversible? Yes, it is! By shifting local light distribution by half period (in Fig.2, after dashed vertical line), reflection DE falls down (erasing process) and then again increasing with time (corresponding to new relief formation). This process is repeatable many times and indicate that the surface relief is product of the local light induced mass transport.

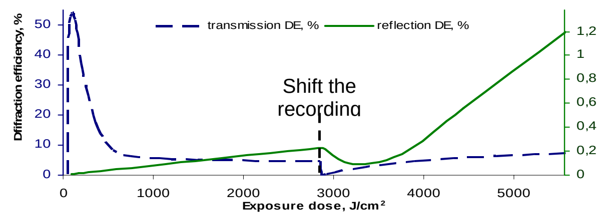


Fig. 2. Diffraction efficiency (η_T values on the left side ordinate axis, η_R- on the right side) changes during holographic recording process, recording kinetic after light distribution shift (dashed vertical line) by half period..

Fig. 3 shows dependence of the recording efficiency on its parameters, i.e. the efficiency of surface relief formation *vs.* intensity of the incoherent light (softening by extra illumination) during holographic recording. Other recording parameters like recording light, its intensity and polarization state (532nm, 0.2W/cm^2 and P-pol.) was held constant.

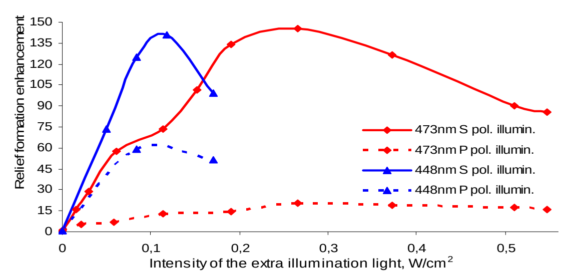


Fig. 3. Surface relief formation enhancement during holographic recording vs. intensity of the extra illumination laser light (dots: experimental data) in the case of extra illumination at different wavelengths and polarization states of incoherent laser light (if $I_{extra}=0$ the relief formation enhancement equals one). Recording was performed by P&P polarization state on As_2S_3 sample by Verdi6 532nm (P polarized) wavelength laser light where $I_1 = I_2 = 0{,}19$ W/cm^2 and period $\Lambda = 1$ um.

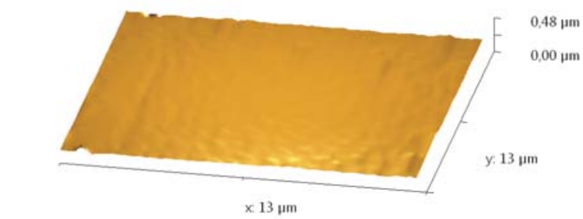
First of all, in Fig. 3 we can see that the efficiency of surface relief formation is strongly changing at increasing intensity of extra illumination. Each curve in the linear part of these curves (up to 0.1W/cm^2 for 448nm extra illumination light and up to 0.2W/cm^2 for 473nm light) we can describe by its slope: for 473nm P polarized extra illumination it is about 75W^{-1}cm^2, for 473nm S polarized- 675W^{-1}cm^2, for 448nm P and S polarized- 64 and 1300W^{-1}cm^2. Here we can easily calculate our gains of the setup parameters compared to the same setup but without extra illumination during holographic recording. For example by using 448nm S polarized 0.08W/cm^2 wavelength light for extra illumination during experiment, we can increase relief formation enhancement (proportional to η_R) from one to about hundred (1300 W^{-1}cm^2 multiplied by 0.08W/cm^2).

These all curves somewhere reach theirs maximum. This means that for every laser light wavelength used for extra illumination there exist conditions for the best recording conditions. By varying the wavelength of extra illumination light we can change its intensity to reach the best performance. As could be seen from Fig. 3, at changing λ from 448nm to 473nm the best performance is reached when I_{473nm}=0.26W/cm^2, which is different from the 448nm case (I_{448nm}=0.11W/cm^2). This can be explained by the fact that 473nm is closer to absorption edge of the sample, i.e. for the same effect we need to illuminate it by the light of higher intensity.

Table 1. Surface relief formation efficiency during holographic recording ($\Delta\eta_R/\Delta t$) vs. polarization state of extra illumination (473nm 0,26W/cm^2 incoherent laser light) and recording laser light. The recording performed on As_2S_3 sample by Verdi6 532nm (P- polarized) laser light ($I_1 = I_2$ =0,25 W/cm^2 , Λ = 1µm)

Extra illumin. pol	Rec. pol.	$\Delta\eta_R/\Delta t$, 10^{-7}/sec	Relief formation enhancement
s		91	260
p	p & p	13,7	39
-		0,35	1
s		0,22	2,2
p	s & s	19	190
-		0,1	1
s		0,17	4,3
p	s & p	0,035	0,88
-		0,04	1
s		23	32
p	45 & 45	0,2	0,28
-		0,72	1
s		102	2,3
p	45 & -45	90,4	2
-		44	1
45		98	2,2
s		58	0,64
p	L & R	62	0,69
-		90	1
s	L & L	57	100
p	or	0,25	0,44
-	R & R	0,57	1

Here (Fig.3., recording by P&P polarized light) we see that the recording efficiency depends not only from the extra illumination wavelength light and its intensity but also from its polarization state. In case of all extra illumination wavelengths (448nm and 473nm) best performance is reached by its S polarization state. In fact, we can initiate constant coefficient a_0 bigger then one what shows how many times S polarized light gives better performance that it is by P polarized light (whatever how big is its intensity). For example, we need just change the extra 473nm P polarized illumination light to S polarized and we get seven times better result (in case of 448nm- about two times). Here we very well can see anisotropic impact to the sample of the linearly polarized light, like the polarization- dependent deflection of As_2S_3 flakes in [7].



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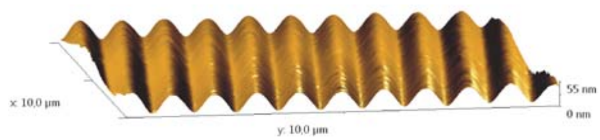
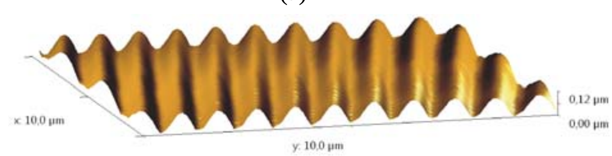


(a)



(b)

Fig. 4. AFM surface pictures of exposed (A and B) and unexposed (*) parts of the sample. (A) and (B) parts were extra illuminated in real time by P-polarized and S-polarized 473nm wavelength light (for A and B see also Figs. 3 and 5). The recording was performed on As_2S_3 sample by Verdi6 532nm (P-polarized) laser light ($I_1 = I_2$= 0.19W/cm^2 I_{extra} = 0.26W/cm^2 and Λ = 1µm).

Table 1 summarizes the data not only on P&P-polarized recording light but also on all other polarization combinations. Here it is important to pay attention to the recording polarization state and the extra illumination polarization state for the maximum performance. As could be seen, the best recording performance without any extra illumination is reached by opposite circularly and 45 and -45 degree polarized light; however, when we start using extra illumination the situation changes. First, we can see that in the case of P&P recording polarization an extra S-polarized illumination is needed for the best performance, whereas in the S&S case – P-polarized one; i.e., in each of the cases cross-polarized extra illumination is required for equal linear recording polarization states. In these cases the enhancement of relief formation is changing from 1 (extra illumination was not used) to ~200. If both recording beams are in 45 degree polarization state, using S-polarized extra illumination we can reach enhancement of surface relief formation up to 30.

Different situation is by using cross polarized recording beams for holographic recording. For -45 and 45 degree polarized recording beams all polarization states of the extra illumination give identical results for the best performance, i.e. whatever polarized light is used for extra illumination we get the same relief formation enhancement. If recording by S&P polarized light, we can get just four times better results by using S polarized extra illumination.

In the case of circularly-polarized recording lights, extra illumination gives an excellent result if both recording beams are polarized in the same direction and the extra illumination light is in S-polarization state (enhancement of relief formation is changing from 1 to 100). If both recording beams are oppositely circular-polarized, the extra illumination just reduces the efficiency of surface relief formation.

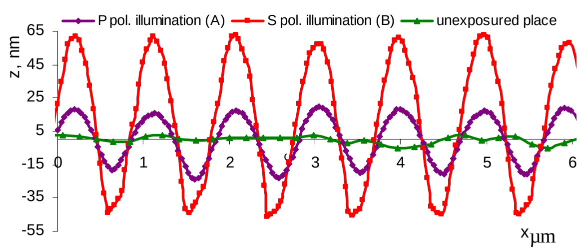


Fig. 5. Surface relief profile after recording at 532nm wavelength P-polarized light ($I_1=I_2=$ 0.19W/cm^2) in the case of extra illumination by 473nm 0.26W/cm^2 wavelength S-polarized (line that represents relief which the largest amplitude changes) and P-polarized (line that represents relief which the middle amplitude changes) light and its comparison with unexposed spot of the sample; (dots: experimental data).

Fig. 4 shows the surface morphology AFM images induced by holographic recording. By (*) is denoted the surface of the amorphous chalcogenide As_2S_3 film before holographic recording is indicated, and by (A), (B) –this surface after recording at extra illumination by P- and S-polarized light. Exact (A) and (B) places (maximum performance setup) are shown in Fig. 3, and its surface relief profile – in Fig. 5. As could be expected from Fig. 3, the surface relief is expressed much stronger in the case of S-polarized extra illumination.

4. Conclusion

The main conclusion that can be drawn from the results of this work is as follows. It is possible to raise the efficiency of holographic recording by extra illumination of the sample by incoherent laser light during the recording. In some cases even more than hundred times better recording efficiency could be reached. We can obtain much stronger diffraction than without extra illumination, or, alternatively, much less time is needed to reach the same diffraction efficiency as without extra illumination.

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