Light induced morphological processing of chalcogenide films

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The study of direct recording of the surface relief gratings on amorphous chalcogenide thin films is presented. Direct patterning was performed on As_2S_3 by 532 nm wavelength laser light using a 10 □m optical slit. The evolution of a surface relief in dependence from the recording time and polarization has been investigated in detail. It has been shown that the formation of surface relief grating closely depends on the superposition of electric field of the gradient beam and supplemental beam, thus the mass transfer can be directed both ways – towards or away from the electric field intensity gradient.

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1. Introduction

Each material is characterized by chemical properties, fluidity, viscosity, absorption, refraction etc [1]. It is possible to slightly change or adjust these properties by using light irradiation. These changes in a light sensitive material enable a selective removal of regions on a thin film by developing and etching [2, 3]. This chemical process is rather complicated, requiring various developing solutions (etchants) for different resist materials. Nowadays it is possible to produce patterned elements directly by light without any kind of chemical processing. Direct patterning is the key technique for fabricating one- and more dimensional surface structures on a wide variety of components. It offers significant advantages such as enabling precise modification of the surface without contamination, remote and contact-less operation, flexibility and precise energy deposition.

The direct surface-relief formation during the holographic recording process confirms the polarization-dependent mass transport [4-6]. A number of models have been proposed to explain the origin of the driving force on the molecular level responsible for the formation of surface-relief gratings under the light illumination. These models include mean-field theory [7], permittivity gradient theory [8], gradient electric force model [9] and others. Still there is no general agreement on the origin of the driving force for this process and no complete mechanism has been yet obtained. Therefore the aim of this work is to study the changes of amorphous semiconductor thin films mechanical and optical properties in the influence of optical irradiation. To achieve it, a new and simplified approach for the morphological processing of surface relief has been used.

2. Experimental

The studied As_2S_3 films with 2.1 μm thickness were vacuum thermal evaporated onto quartz substrates. To investigate direct recording possibilities, a single-beam recording setup was introduced. The main object of this setup is a narrow optical slit with 10 μm width. Through this slit, the sample was exposed by a uniformly distributed and unfocused monochromatic light therefore crating light intensity gradient. Such recording system allows to produce one line gratings and opens a possibility to investigate the relief formation's dependency on the intensity of light and its polarization. Previous direct holographic recording experiments with a linearly polarized light showed that the relief forming efficiency can be significantly improved by an additional incoherent supplemental beam [9]. The same approach was utilized for this setup and supplemental exposure was used from the other side of the sample. For the gradient beam 532 nm Nd:YAG CW laser and for the supplemental beam other incoherent 532 nm CW laser diode was used. The obtained surface relief was mapped by atomic force microscopy (AFM) using *Veeco CPII*.

Polarization of gradient and assisting beams was varied by halfwave and quarterwave plates. Since the sample surface is perpendicular to the gradient beam and supplemental beam, polarization direction needs to be defined. Thereby, if polarization direction of the light coincides with the slit direction (*i.e.* grating vector) then polarization state can be written as 0 degree or *s* polarization. If polarization direction of the light is perpendicular to the slit direction, polarization state can be written as 90 degree or *p* polarization.

3. Results and discussion

In order to verify the influence of polarization state of the recording beams on the direct surface patterning, we performed experiments by illuminating the amorphous As_2S_3 films through an optical slit. This single-line grating recording is one of the simplest ways to investigate light induced molecular migration- it is possible to control exposure dose, polarization, intensity and as well as to monitor the direction of mass transfer.

The first experiments, not taking into account the polarization of the gradient light, resulted with negligible outcome- the height of the obtained surface relief reached only few nanometers and could be attributed to photo-induced expansion effect [10]. From the previous work it is known that it is possible to improve the recording efficiency by introducing additional perpendicularly polarized homogeneous illumination [9]. It has been determined that the polarization of the supplemental beam causes isotropic or anisotropic softening which in conjunction with polarization of the gradient beam might show some peculiarities. As it turns out, the same approach worked for the optical slit.

Fig. 1 shows the outcome of the two recording experiments performed with *s* or *p* polarized gradient light in a combination with the orthogonally polarized supplemental beam. It can be seen that the direction of the photoinduced mass transfer in both cases is with an opposite sign. Consequently, the *s* polarized gradient beam with the *p* polarized supplemental light (*s*/*p* setup) creates a contraction of the film or formation of a valley, but the *p* polarized gradient beam with the *s* polarized supplemental light (*p*/*s* setup) creates an expansion of the film or formation of a ridge.

The impact of the supplemental beam polarization on the relief formations efficiency or obtained relief height is shown in Fig. 2a. Intensity of the gradient beam and the supplemental beam was kept constant at 1.5W/cm^2 and 1W/cm^2 respectively. In this case *p* polarized 532 nm light has been used as the gradient beam and polarization for the supplemental beam has been changed from *s* (0 degree) to *p* (90 degree) polarization by a 10 degree step. In the case of cross-polarized gradient and supplemental beam (the 0 degree point) the obtained profile is with the highest amplitude. By decreasing the angle between those two polarization states the obtained profile heights also decreases. When the polarization of the gradient beam and supplemental beam coincides (the 90 degree point) the obtained surface relief reaches only few nanometers. This case shows the same outcome as the setup without the supplemental beam, *i.e.*, the obtained profile height is negligible and can be related to photo-induced contraction or expansion [11]. All obtained measurements are linearly arranged therefore the line approximation has been used. 0.99 R-squared value means that this setup is very stable and with high reliability. Five of the obtained profiles are shown in Fig. 2b. The minimum positions of the obtained profiles (-5 and 5 μm) correspond to the location of the slit edges. It is shown that any noticeable changes of the sample surface starts from the middle of the slit thus confirming that this recording is a bulk process.

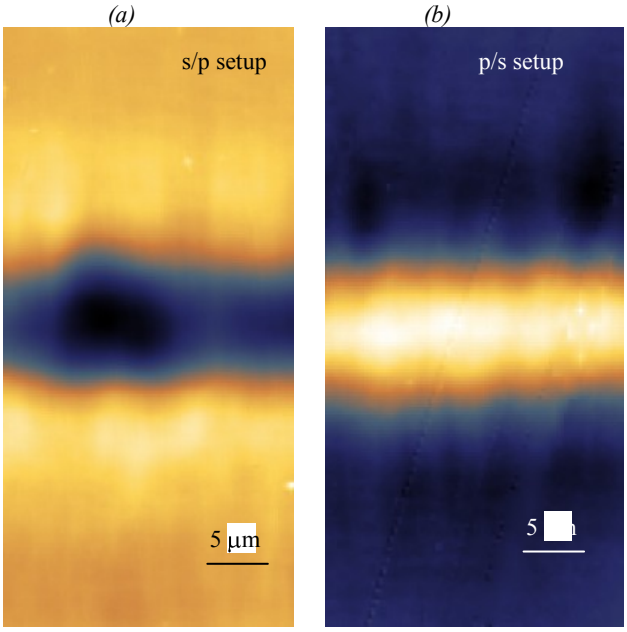
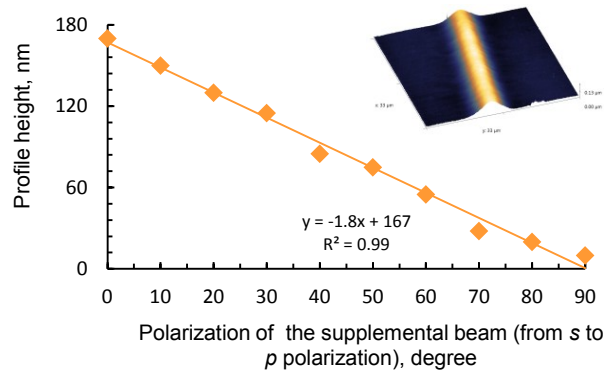
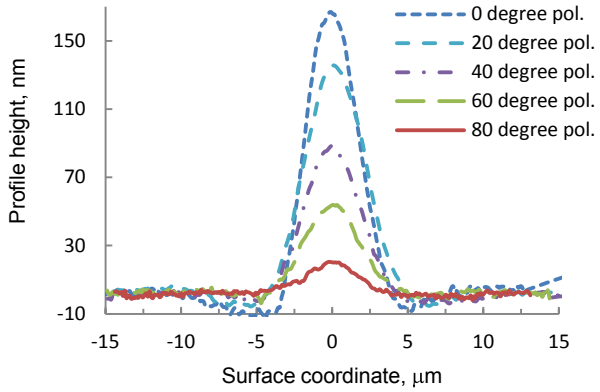


Fig. 1. Surface relief 2D topography of 10 μm wide optical slit experiment showing different mass transfer directions with respect to the gradient beam and supplemental beam polarization state (mentioned as a polarization for the gradient/supplemental beam)



(a)



(b)

Fig.2. (a) Obtained relief heights in dependency from polarization direction of the supplemental beam and (b) its profiles; 10 μm optical slit experiment was performed with 532nm p polarized light for the electric field intensity gradient, exposure time t=1h.

In Table 1 the summary of obtained results has been presented for all other polarization combinations. Certain cells are carrying a value $\Delta h \rightarrow 0$ – this means that with such polarization configuration the obtained relief is insignificant and basically represents a smooth surface. The remaining cells which do carry values different from zero are represented by profile height distributions in the 300 nm height and 25 μm wide frames. As can be seen from combinations that produced an observable surface relief modification, the mass transfer can manifest in different ways also for other than *s* and *p* polarization combination. If the polarization of the gradient beam coincides with the direction of optical slit, then independently from the polarization of the supplemental beam (the 1st row: *CP*- circular, *p*- perpendicular to the slit or *45* degree) a mass transfer away from the recording region or formation of a groove can be observed. The efficiency of recording or the depth of groove changes from the best case, when the polarization of supplemental beam is perpendicular to that of the recording beam, then follows the 45 degree and finally *CP* polarization.

The same applies when the polarization of gradient beam is perpendicular to the slit (*s* gradient beam; the 2nd row in Table 1.), only in this case mass transfer is directed towards recording region and formation of a ridge can be observed. For the other two gradient beam polarization cases (*CP* and *45* degree polarization from the 3rd and 4th row) mass transfer direction has been determined by polarization direction of the supplemental beam. The *s* polarized supplemental beam determines mass transfer towards the 10 μm recording region thus forming a ridge while the *p* polarized supplemental beam determines mass transfer away from the recording region thus forming a groove. All recording combinations are symmetrically aligned in respect to the main diagonal of the table. This observation means that the polarization of both – gradient beam as well as supplemental beam play a significant role in the process of mass transfer.

Table 1. The correlation between polarization and light induced mass transfer processes in 2.1 μm thin film of As_2S_3 sample for different polarization combinations of the electric field intensity gradient and supplemental beam (1.5 W/cm² and 1 W/cm² for 1 h respectively) in optical slit experiments; the inset contains profiles in the 300 nm height and 25 μm wide frames

	s suppl.	*p* suppl.	*CP* suppl.	*45°* suppl.
s grad.	Δh → 0	Δh = -220 nm	Δh = -65 nm	Δh = -96 nm
p grad.	Δh = 200 nm	Δh → 0	Δh = 78 nm	Δh = 113 nm
CP grad.	Δh = 95 nm	Δh = -80 nm	Δh → 0	Δh → 0
45° grad.	Δh = 115 nm	Δh = -60 nm	Δh → 0	Δh → 0

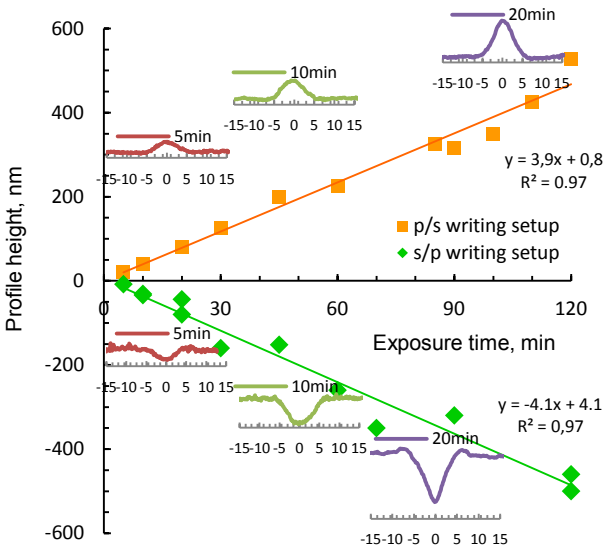


Fig. 3. Obtained relief heights in dependency from the exposure time and its profiles form the first 20 minutes, 532 nm s or p polarized light was used for the electric field intensity gradient and for the supplemental beam (polarization for each setup is accordingly denoted with the slash sign)

The time dependent formation of ridges and grooves for the 10 μm wide slit are shown in Fig. 3. Intensity of the gradient beam and the supplemental beam was kept constant at 4.24W/cm² and 0.37W/cm² respectively. Profiles from the first 20 minutes are shown for both of the recording cases. It can be seen that the formation of ridges as well as grooves are strongly linear therefore a line approximation has been used. As the slope coefficients show, the recording efficiency for this setup is ±4nm/min ("+" for the p/s and "-" for the s/p setup). After a 120 min exposure the amplitude will reach 550 nm (measured from the initial surface). Note that the relief obtained without supplemental beam was unnoticeable for all recording parameters and exposure lengths.

4. Conclusion

We have investigated that the light-induced mass transfer process for amorphous As_2S_3 thin films strongly depends on the polarization of the light. The behavior of mass transfer and thus the resulting recording could be related to interaction between the polar photo-induced defects and the polarized electric field of the gradient beam and the supplemental beam. It has been shown that the formation of surface relief grating in amorphous As_2S_3 films closely depends on the superposition of electric field of the gradient and supplemental beams, thus the mass transfer can be directed both ways – towards or away from the electric field intensity gradient.

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

Acknowledgments

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