

# Holographic patterning of acrylamide-based photopolymer surface

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**Abstract:** The patterning of an acrylamide-based photopolymer surface by holographic recording is studied. The patterns are induced by light alone and no post-processing is required. Periodic surface modulation is observed in addition to a volume phase grating. An investigation has been carried out using white light interferometry into the dependence of the amplitude of the photoinduced surface relief modulation on the spatial frequency, intensity of recording and sample thickness. The observed dependencies indicate that the diffusion of material during the holographic recording plays a major role in surface relief formation. The possibility for inscription of surface relief patterns opens the door to at least two new applications for this photopolymer: fabrication of diffractive optical elements and biosensors.

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## Reference and links

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## 1. Introduction

Photoinduced single step inscription of a surface relief modulation in photopolymer systems opens attractive perspectives for applications such as diffractive optical elements [1,2], recording of computer generated holograms [3], and biosensors. Important characteristics of the material in relation to these applications are its spatial resolution, the amplitude of photoinduced surface relief, the required recording time for achievement of maximum surface modulation and its long-term stability. For optimisation of all these characteristics a thorough understanding of the mechanism of surface relief formation under light illumination is necessary. Although photopolymer systems have been a subject of constantly increasing interest, especially after the development of self-processing photopolymer systems, only a few investigations dedicated to the surface relief formation have been reported in the literature [1,2,4,5].

In the present paper, for the first time to our knowledge, we report an investigation of the photoinduced surface relief modulation in an acrylamide-based photopolymer system [6,7], developed at the Centre for Industrial and Engineering Optics, Dublin Institute of Technology. A maximum surface relief modulation in the order of 3  $\mu\text{m}$  at spatial frequency of 4 l/mm was achieved using recording times of 30 s and intensity of 2.5mW/cm<sup>2</sup>. The dependencies of the photoinduced relief modulation on the spatial frequency, recording intensity and sample thickness are presented. These characteristics are compared with the reported data for other photopolymer systems.

## 2. Theory

### 2.1 Photopolymers for holographic recording

The general composition of a photopolymer layer contains a monomer, electron donor, photosensitizing dye and, optionally, a polymer binder. Upon illumination of the photopolymer with a non-uniform light field of appropriate wavelength the sensitizing dye absorbs a photon and reacts with an electron donor to produce free radicals. These initiate polymerisation where the light was absorbed. The changes in the density and the molecular polarizability, which accompany the polymerisation, lead to change in the local photopolymer refractive index and a hologram is recorded. According to recently developed theoretical models [8-11] describing holographic recording in photopolymer systems, an important process in hologram formation is monomer diffusion from the dark to bright fringe areas. Our recent investigations of the post recording behaviour of low diffraction efficiency gratings reveal that monomer diffusion in the acrylamide-based photopolymer system is comparatively fast [12] and is characterised by diffusion constant of  $D_{01}=1.6 \cdot 10^{-7}$  cm<sup>2</sup>/s in absence of crosslinker. If, as believed, monomer diffusion is involved in the process of photoinduced surface relief formation then relatively short exposure times and high surface relief amplitude would be expected in this photopolymer system even at low spatial frequencies. In the same study it was observed that in addition to the monomer diffusion from dark to bright fringe areas a second diffusion process in the opposite direction –from bright to dark fringe areas occurs on a much slower time scale  $D_{02}=6.25 \cdot 10^{-10}$  cm<sup>2</sup>/s. We ascribe this second process to diffusion of terminated or non terminated short polymer chains. The influence of this second process on the amplitude of the holographically inscribed surface modulation has not been investigated so far.

### 2.2 Models describing the process of holographic patterning in self-developing photopolymers

The models describing surface relief formation in photopolymers can be divided into two main groups. The first group explains the relief formation by shrinkage of the photopolymer depending on the intensity of light. This mechanism is applicable to the systems where the

peaks of the surface relief appear in the non-illuminated areas [5]. The models of the second group are based on the assumption that redistribution of system components by diffusion is responsible for the relief formation. This fits well with experimental data showing that the surface relief peaks appear in the illuminated areas. The models from the second group differ mainly in the suggested cause of the diffusion of the components – a monomer chemical potential [1] gradient, concentration gradient of monomer [2] and local shrinkage of a polymer layer [4]. All of the proposed models consider the change in the surface free energy during the relief formation as the main reason for the restricted resolution of the inscribed relief structures [1-5].

The surface relief formation in a photopolymer containing acrylamide is reported by Boiko et al. [1]. The main difference between the system described in [1] and the one described here is the sensitising dye and the presence of a second monomer that plays a role of crosslinker in our photopolymer. The authors of [1] introduce a thermodynamic model that considers the gradients of the chemical potentials of the components to be responsible for the diffusion of monomer from dark to bright fringe areas. The formation of surface relief under illumination is described by the initial formation of a surface wave due to monomer mass transport. The need for compensation of the gradient of the monomer chemical potential leads to the spreading of this surface wave and the ultimate formation of the surface profile.

A detailed study of the surface relief formation under UV illumination of another photopolymer system is presented by C. Croutxe- Barghorn et al. [3]. The effect is explained by the existence of two different processes. The first process is the transport of reactive molecules from the dark to the illuminated regions resulting from the local change of the monomer concentration during spatially non-homogeneous illumination through a mask. The second process considered by the authors is the appearance of a surface free-energy gradient and a resulting force that tends to stabilise the system. The positions of the surface relief peaks in this system coincide with the illuminated areas. A photopolymer system containing a neutral component in addition to the standard photopolymer composition is investigated by Smirnova et al. [4]. The authors propose a model of relief formation based on the concept of polymer local shrinkage and the formation and redistribution of an additional free volume due to phase separation between the neutral component and polymerised material.

Neither of the revised models takes into account the possibility of counter diffusion of material from bright to dark fringe areas and its influence on the surface relief formation.

### **3. Experiment**

#### *3.1 Materials*

The photopolymer layer was prepared as described elsewhere [6]. The sample consisted of a polyvinyl alcohol binder, two monomers – acrylamide and N,N'-methylenebisacrylamide, Erythrosin B sensitising dye and triethanolamine as an initiator. This photopolymer system is water-soluble. After mixing the components the solution was spread on glass plates with dimensions of 5x5 cm<sup>2</sup>. The films were ready for use after drying for 24 hours at constant humidity of 70%. The optical density of the layers was 0.3 OD.

In order to investigate the dependence of the surface relief amplitude on the photopolymer film thickness, different amounts of photopolymer solution were deposited on the same area of 5x5 cm<sup>2</sup>. Layers of thickness from 50 to 250 μm were obtained following this procedure.

#### *3.2 Experimental set-up*

The photoinduced surface relief was inscribed by holographic recording of transmission diffraction gratings with spatial frequency in the range 4 - 300 l/mm. The second harmonic of a Verdi 05 NdYVO<sub>4</sub> laser at 532 nm was used to record the gratings. Two different holographic set-ups were used for recording gratings with spatial frequencies above 100 l/mm (Fig. 1(a)) and below 100 l/mm (Fig. 1(b)). The first set-up is a standard holographic optical

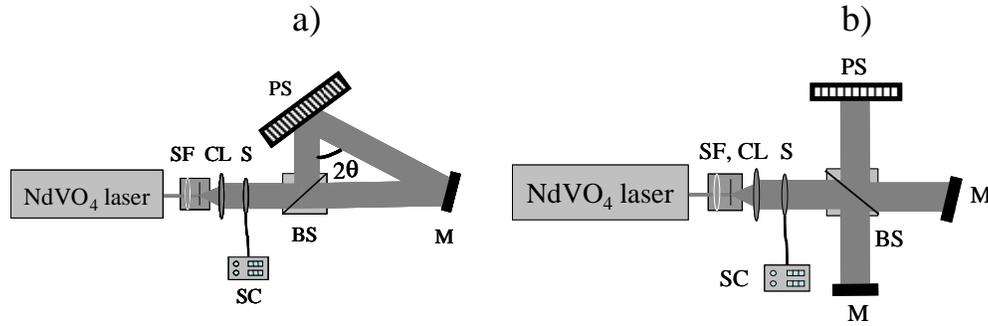


Fig. 1. Holographic set-up of recording at spatial frequencies above 100 l/mm a) and below 100 l/mm b), SF-spatial filter, CL- collimating lens, PS – photopolymer sample, M- mirror, S – shutter, SC – shutter controller.

set-up. The spatial frequency of recording was altered by changing the position of the mirror M in Fig. 1(a) and so changing the angle between the two recording beams. The second set-up is a Michelson type interferometer and the change in the spatial frequency was achieved by rotation of one of the mirrors. For recording more complicated 2D patterns the sample orientation was changed between two consecutive recordings (crossed grating) as shown in Fig. 2. Symmetric patterns were obtained when the spatial frequency was kept constant for both recordings Fig. 2(b) and asymmetric when the spatial frequency was altered between the two recordings Fig. 2(c).

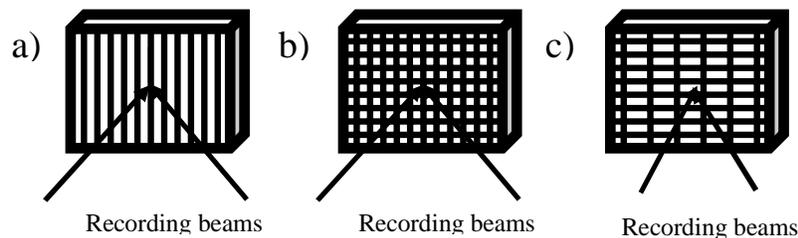


Fig. 2. Patterns recording. After recording of the first grating a) the sample is rotated at 90° and a second grating with the same b) or different spatial frequency c) is recorded.

The recording intensity was varied in the range 1-25 mW/cm<sup>2</sup>. In order to observe the photoinduced surface modulation at different stages of formation the exposure time was varied in the range 0.5 s to 250 s.

The photopolymer surface after recording was scanned using a white light interferometric (WLI) surface profiler *MicroXAM S/N 8038*.

The overall film thickness was characterised by cutting the photopolymer layer using a sharp scalpel and measuring the dimensions of the cut with the WLI profiler.

## 4. Results and discussion

### 4.1 Determination of the position of the surface relief peaks

In order to localise the position of the surface relief peaks a simple holographic grating with very low spatial frequency of 5 l/mm was recorded. Immediately after the recording the alternating stripes of bleached (where the light was absorbed) and unbleached regions were easily observed. The Dektak profiler is equipped with a video camera and the position of the measuring tip with respect to the bright or dark fringes can be easily correlated to the actual

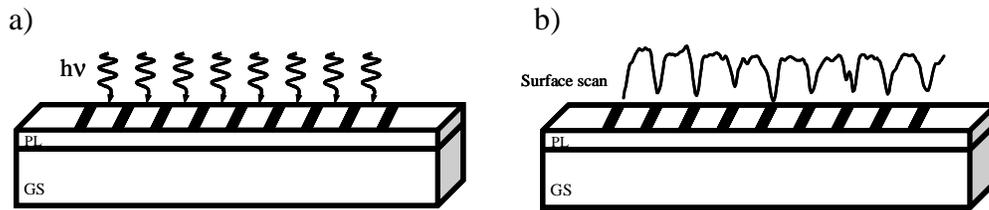


Fig. 3. Position of the surface relief peaks when a simple diffraction grating is recorded. Light illumination leads to well distinguished consecutive bleached and unbleached stripes. a) Surface scan with Dektak\_3 profiler equipped with video camera correlates the position of the peaks with the bleached regions b). PL- photopolymer, GS – glass substrate.

measurement in a vertical direction. Using this method we were able to observe that the surface relief peaks appear in the light illuminated areas as shown in Fig. 3. Such an observation supports the hypothesis that the diffusion plays an important role in the surface relief formation. At the same time shrinkage of polymer during the recording may have a role. It is known [14] that the bulk free radical polymerisation is usually accompanied by substantial shrinkage of the polymer. The degree of shrinkage in some studies is used for characterisation of the rate of polymerisation in photopolymers [15]. If the formation of the relief was caused only by the shrinkage of the polymer the peaks of the relief should appear in the dark regions. It was, however observed here that the peaks coincide with the illuminated regions. This reveals that if there is an initial shrinkage of the polymerised regions in the beginning of recording it must be very quickly followed by diffusion of molecules from regions outside the illuminated areas.

#### 4.2 Photoinduced surface relief at different stages of recording

In order to trace the surface relief formation, the recording process was interrupted at different stages. The profile was measured after exposing the sample to a sinusoidal modulated intensity with a spatial frequency of 10 l/mm for various time intervals between 0.5 s and 90 s. The recording intensity was 2.5 mW/cm<sup>2</sup>. The surface profiles after recording for 0.5 s, 10 s, 30 s and 90 s are presented in Fig. 4. As can be seen from the figure a significant surface relief modulation of 250 nm is inscribed already in only 0.5 s recording time. The surface profile modulation increases up to 1.6 μm when the recording time is 20 s and then decreases to 1.1 μm when the recording time is 90 s. The change in the amplitude of photoinduced surface relief is accompanied by pronounced change in the shape of the surface profile. It supports the hypothesis that diffusion of material is involved in the mechanism of formation of the observed periodical structure. It is noteworthy that relatively short exposure times are required for inscription of surface relief modulation above 1 μm in this photopolymer system. For

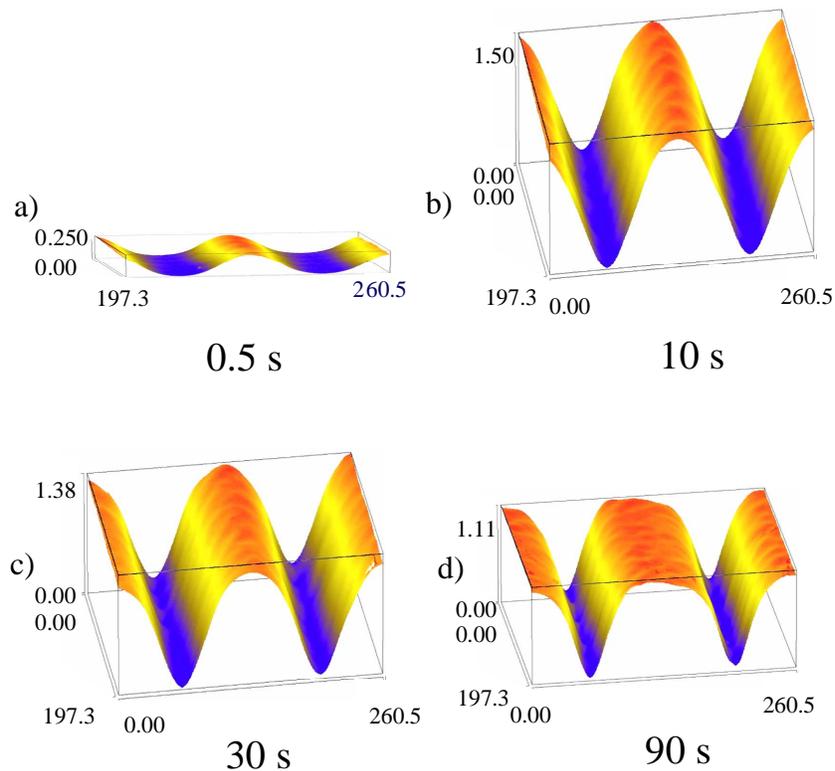


Fig. 4. Surface relief modulation in  $\mu\text{m}$  at spatial frequency of 10 l/mm after recording for 0.5 s (a), 10 s (b), 30 s (c) and 90 s (d). The recording intensity is  $2.5\text{mW}/\text{cm}^2$ .

comparison, the required time for recording in other photopolymers systems [1, 2] using similar intensities is in the order of several minutes. For instance, in order to achieve surface relief modulation above  $1\mu\text{m}$  in our system an exposure level of  $25\text{mJ}/\text{cm}^2$  is required. For the photopolymer system reported in [2] an exposure level of approximately  $500\text{mJ}/\text{cm}^2$  is necessary to achieve such surface relief modulation.

As seen in Fig.4 a decrease of the surface relief modulation was observed with increase of the exposure time. The effect of saturation of the material around the bright peaks can be one of the reasons for this effect.

#### 4.3 Dependence on the intensity of recording

The dependence of the photoinduced surface modulation on the recording intensity was studied for the simplest case – illumination with the interference pattern of two plane waves and recording of a diffraction grating. A number of different exposure levels were chosen and the intensity and recording times were simultaneously varied. This approach assures that the same number of photons is delivered in different time intervals. The surface relief amplitude was measured as the difference in height of the middle of the bright and middle of the dark fringe. The surface relief profiles were similar to the ones presented in Fig. 4. A deviation from this shape of the profile was observed only at intensity of  $20\text{mJ}/\text{cm}^2$  and recording times above 25 s. In these cases a crater shape with a dip in the middle was observed and the

amplitude was measured as the difference of the height in the middle of the dark fringe and the top of the crater edges (the maximum surface relief amplitude). Pronounced intensity

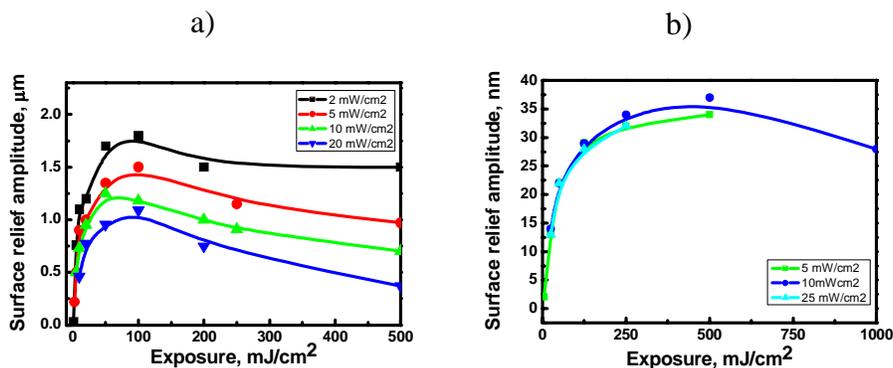


Fig. 5. Intensity dependence at spatial frequency of recording of 10 l/mm a) and 100 l/mm b).

dependence was observed when the recording was carried out at a spatial frequency of 10 l/mm. As is seen from Fig. 5(a) when the photons are delivered at slower rate (low intensity and longer time of exposure) the ultimate surface relief modulation is higher. This could be explained by the fact that at lower intensity the individual polymer chains are expected to grow longer as the radical concentration will be lower and termination will occur at a lower rate. In this case more monomer molecules will be consumed per photon in the process of polymerisation and the final gradient in monomer concentration will be higher. This will lead to more monomer molecules diffusing from bright to dark fringe areas in order to restore the equilibrium in the monomer concentration. In addition to the actual number of diffusing molecules the diffusion time might also be important for the final surface relief amplitude. At lower intensity of recording the polymerisation rate will be slower and there will be more time for the molecules from outside the illuminated areas to diffuse into the bright fringes before the polymerised region is cross linked and not so permeable anymore. Using the same conditions of recording and going to higher spatial frequency will not change the expected number of diffusing molecules but will decrease the time required for diffusion between the fringe areas. If the diffusion time is important, one could expect that at higher spatial frequency the intensity dependence would not be so pronounced. Indeed, when the spatial frequency was increased to 100 l/mm no such dependence was observed (Fig. 5(b)). At the same time an overall decrease in the surface relief modulation was observed at 100 l/mm and this implied that additional factors may influence the final surface relief amplitude.

#### 4.4 Dependence on the spatial frequency of recording

In order to reveal what other factors influence the surface relief formation an investigation of the dependence of the surface relief amplitude on the spatial frequency of recording was necessary. Keeping the rate of delivery of photons constant, intensity of 2  $\text{mW}/\text{cm}^2$ , we measured the surface relief amplitude at up to 300 l/mm using a WLI. Above this spatial frequency the grating's fringe spacing becomes too small to be resolved by the WLI profiler. The results are presented in Fig. 6. They clearly indicate that with increase of the spatial frequency the photoinduced surface relief amplitude decreases. If only the diffusion of material into the bright fringe areas was involved in the surface relief formation, an increase of the amplitude might be expected with the increase of the spatial frequency as the diffusion time will be shorter at higher spatial frequency. The fact that the overall amplitude decreases implies the existence of an additional factor and we believe that it is the increased surface tension that prevents the formation of higher surface relief amplitude at higher spatial

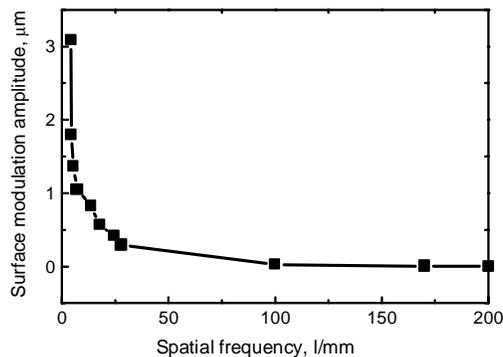


Fig. 6. Dependence of the surface relief amplitude on the spatial frequency of recording. The surface is scanned after recording for 30 s at 532 nm with intensity of  $2\text{mW}/\text{cm}^2$ . The error is within the size of the symbols.

frequencies. In other words, it is easier for the polymer film surface to deform when the fringe spacing is larger. It should be borne in mind that this is not an etching type process where material is dissolved away in order to get the relief pattern. In this material the relief develops as a result of local forces and the film surface must deform in order to create the relief pattern. It is therefore reasonable that at higher spatial frequencies we might observe a decrease in the modulation depth.

#### 4.5 Counter propagating waves

In order to investigate the role of the diffusion in the process of surface relief formation we carried out the following experiment. The diaphragm of the WLI (Fig. 7(a)) was imaged onto the photopolymer layer for time intervals varied between 5 and 1800 s. The experiment was carried out in the dark in order to avoid any additional polymerisation

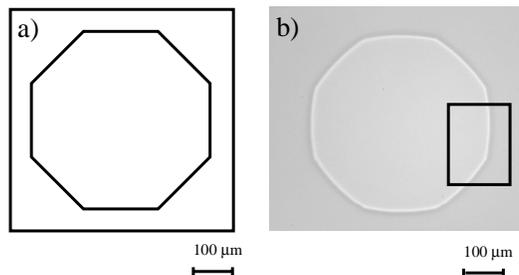


Fig. 7. The diaphragm of the WLI profiler a) was imaged on the photopolymer film surface and the examined with phase contrast microscope b).

due to absorption of ambient light. The wavelength of light used was 532 nm and as the unexposed photopolymer layer is sensitive to this wavelength the illumination was causing polymerisation in the area where the diaphragm was imaged. The change in the refractive index due to the polymerisation of the photopolymer layer was observed with the help of a phase contrast microscope (Fig. 7(b)). It is clearly seen in Fig. 7(b) that there is an area with different refractive index than that of the background and that it has the shape of the WLI diaphragm.

After exposure of the layer for different intervals of time the boundary of the illuminated area (marked with the black rectangle in Fig. 7(b)) was examined. The intensity of the light used for the scan was decreased in order to avoid additional influence of the measuring beam.

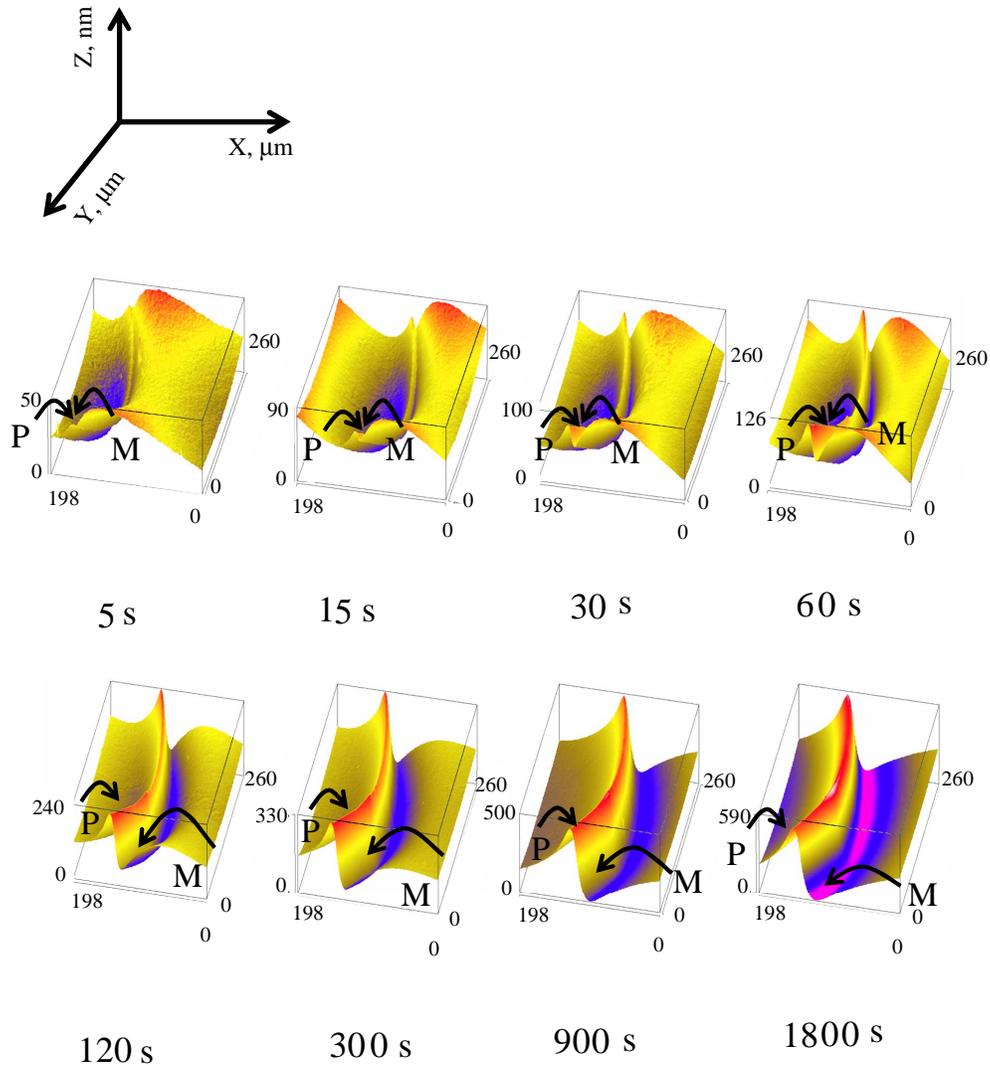


Fig. 8. The boundary between the illuminated and dark region, marked with the black rectangle in Fig.7b) was studied after different times of exposure from 5 to 1800 s c). The formation and the development of the two counter propagating waves at the edge of illuminated and non illuminated areas are observed.

The time for a single scan procedure in the most sensitive mode of the WLI, when only 1  $\mu\text{m}$  in the vertical direction is scanned, is 5s. We assume that during the scan the measuring beam does not substantially change the observed picture. The photopolymer layer was translated after every scan and the subsequent pictures corresponding to different exposure times were taken at new spots on the photopolymer surface. A calculation of the expected

position of the characteristic rings due to the diffraction from a circular aperture reveal that the observed picture is not a result from intensity variation due to diffraction. The smallest size aperture in our optical system was 5 mm (the objective), the wavelength used was 500 nm and the distance between the aperture and the sample was around 5 mm.

It is seen in Fig. 8 that two waves that propagate in opposite directions, appear at the boundary between the illuminated and dark areas. The first, much larger, wave (marked with M in Fig. 8) which propagates towards the illuminated area could be ascribed to diffusion of monomer molecules. The second wave which has smaller amplitude and propagates out of the illuminated area (marked with P in Fig. 8) could be ascribed to short terminated or non terminated polymer chains (this corresponds to the second diffusion process occurring in the opposite direction which was observed and quantified in previous studies). The ultimate effect is that the illuminated area is elevated above the non-illuminated one. This implies that more molecules flow into the illuminated area than leave it. Such an effect could be expected as the monomer molecules are much smaller and their diffusion rate is expected to be much greater than that of the short polymer chains.

An estimation of the average distance,  $x_d$ , traveled by the diffusing particle in a time  $t$  can be obtained using the formula<sup>18</sup>

$$x_d = 2\sqrt{\frac{Dt}{\pi}}, \quad (1)$$

Where  $D$  is the diffusion coefficient of the medium.

Investigation [13] of the diffusion rates for the two diffusion processes at the beginning of holographic recording in acrylamide- based photopolymer containing monomer and crosslinker reveals that the first, faster process is characterised by diffusion coefficient of  $D_{01}=1.32 \cdot 10^{-8} \text{ cm}^2/\text{s}$ . This process is ascribed to a diffusion of monomer molecules from dark to bright fringe regions. The second, slower process, ascribed to diffusion of short terminated or non terminated polymer chains, is characterised by  $D_{02}=2.5 \cdot 10^{-10} \text{ cm}^2/\text{s}$ . After substitution of the diffusion coefficients in eq.1 one can estimate the corresponding average distances traveled by the monomers and the short polymer chains. The average distance traveled by monomers changes from 5  $\mu\text{m}$  for the first 5s to 97  $\mu\text{m}$  for 1800 s. At the same time the average distance traveled by short polymer chains (corresponding to the wave P in Fig. 8) changes from 700 nm to 13.4  $\mu\text{m}$ . The estimated distances are well within the scale of the observed two waves in Fig. 8. This, of course, is a simplistic picture of the processes at the bright/dark boundary as it uses the “dark” diffusion coefficients to estimate the diffusion distances. In fact the diffusion properties of the photopolymer will change with time in the process of polymerization and the diffusion will slow down as more monomer molecules are converted into polymer molecules.

The significance of this experiment is that it constitutes the first direct evidence, to our knowledge, of the mass transport of material from the bright to dark fringe regions. This mass transport process is more likely to be important in the stage of recording when the polymer chains are below given size. The measurements of the surface relief modulation at 100 l/mm were repeated 24 hours after recording and no significant change was observed.

#### 4.6 Dependence on the photopolymer film thickness

The influence of the photopolymer film thickness on the amplitude of the photoinduced surface relief was also studied. Samples with thickness from 50 $\mu\text{m}$  to 250 $\mu\text{m}$  were prepared by depositing different amounts of photopolymer solution on the 50x50mm glass substrates. In this range of thicknesses no pronounced difference in the photoinduced surface relief amplitude was observed. The same conditions of recording were applied for all of the samples.

#### 4.7 Holographic recording of patterns

Using the arrangement described in Fig. 2 different holographic patterns were inscribed on the photopolymer surface. The success in using two sequential recordings can be ascribed to the large dynamic range of the material and the fact that only some of the monomer is consumed during the first recording. Patterns with differently shaped unit cells can be inscribed by simply rotating the photopolymer plate by different angles. The advantage of this technique of recording is that by using a sufficiently powerful laser (such lasers are now available at modest prices) one can obtain large area optical elements with variety of patterns with no need to produce a specially designed mask for every single pattern. Examples of symmetric Fig. 9(a) and asymmetric patterns Fig. 9(b) are presented. We believe that this technique can have a substantial impact on the design of different liquid crystal devices. A separate investigation of this subject is under progress.

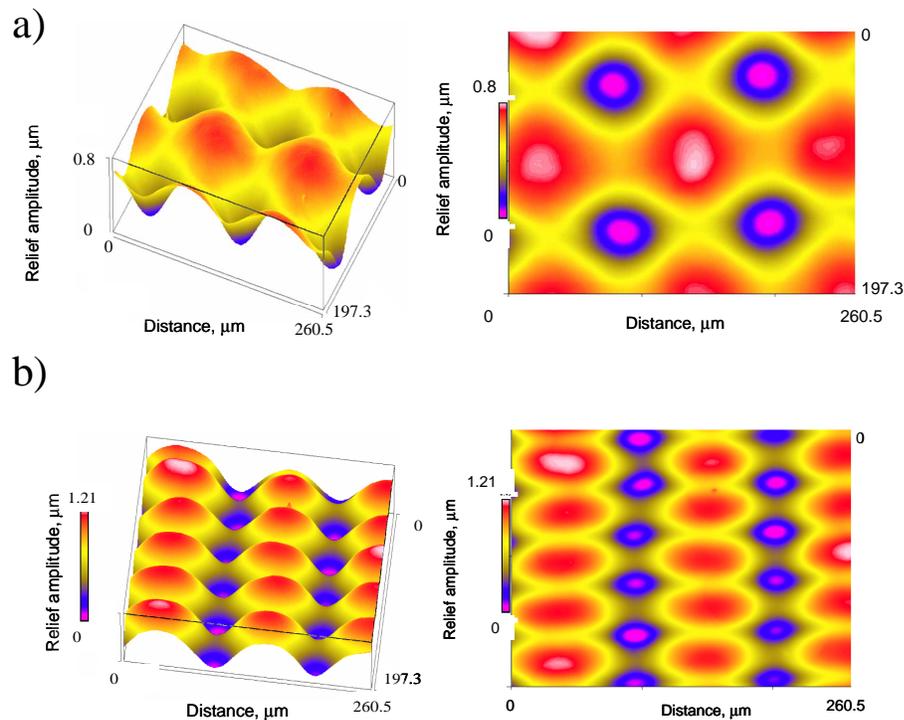


Fig. 9. Patterns inscribed after two consequent recordings using the same spatial frequency a) and changing the spatial frequency between the two recordings b).

#### 5. Conclusions

The process of surface relief formation by holographic recording in acrylamide-based photopolymer was studied. It was established that the surface relief peaks coincide with the areas illuminated by light. This fact and the measured dependencies of the surface relief amplitude on the intensity and time of recording suggest that the diffusion of material plays a substantial role in the surface relief formation.

Two diffusion processes that make opposing contributions to the surface relief formation have been identified here and in previous studies. Diffusion of monomer molecules from dark to bright fringe areas has a positive contribution to the surface relief formation. The second process was ascribed to diffusion of short terminated and non-terminated polymer chains from

bright to dark fringe areas. It probably takes place in an early stage of recording and would have negative effect on the surface relief formation.

At higher spatial frequencies the overall surface relief amplitude decreases. At these spatial frequencies the surface tension possibly plays the major role by preserving the photopolymer layer surface from deformation.

The results presented in this paper should be regarded as a preliminary analysis of the surface relief formation in acrylamide-based photopolymers. Further experiments on the dependence of the surface relief modulation on the visibility of the fringe pattern, the dye concentration and modelling of the surface relief shape are under progress.

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