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Effects in reconstruction of diffraction gratings multiplexed in acrylamide photopolymers

M. Ortuño^{*b}, S. Gallego^b, A. Márquez^b, C. Neipp^b, A. Beléndez^b and I. Pascual^a

^aDepartamento Interuniversitario de Óptica, Universidad de Alicante

^bDepartamento de Física, Ingeniería de Sistemas y Teoría de la Señal, Universidad de Alicante

^{*}mos@ua.es

ABSTRACT

In this study we focus on the properties of acrylamide photopolymer layers about 1 mm thick with hydrophilic binder of polyvinyl alcohol and yellowish eosin as dye. We consider peristrophic multiplexing. Note the differences in the reconstruction step between the next two peristrophic multiplexing schemes: rotation axis parallel and rotation axis perpendicular to the hologram media. In the first case (parallel) a very thick media allows for a high storage density and a highly selective individual hologram reconstruction thanks to the Bragg angular selectivity. In the second case (perpendicular), an additional optical system is necessary to separate the various simultaneous holograms reconstructed. We show experimental evidence for the two schemes. We analyze the behaviour of the material when multiplexing diffraction gratings using peristrophic multiplexing. In the reconstruction step we observe differences, between the various multiplexed holograms, in their angular response shape. We consider that this is due to polymer diffusion in exposed areas or to the dye concentration variation with registered holograms number, because xantene dyes are bleached at the initiation step of radical polymerization reaction. The photopolymer composition is an important factor to be taken into account when single or multiple holograms are registered. The differences in both cases are considered in this work.

Keywords: photopolymer, peristrophic multiplexing, holography, acrylamide

1. INTRODUCTION

Acrylamide (AA) photopolymers with a polyvinyl alcohol (PVA) binder are very versatile holographic recording materials¹. Their most important characteristics include high diffraction efficiency, ease of preparation and the fact that they do not need post-processing after the hologram has been recorded². These properties make them very suitable for studying factors relating to the process of hologram recording, changes in the photopolymer composition, etc. Recently acrylamide based photopolymers have been developed in 1 mm thick layers, and these are ideal for use as the recording medium in studies relating to holographic information storage³.

In order to take full advantage of the storage capacity of the recording medium, various multiplexing⁴ techniques are used enabling multiple holograms to be recorded in a given volume of photopolymer. Depending on its thickness and composition, each photopolymer is capable of reaching a particular dynamic range⁵ ($M/\#$), that is, it enables a certain number of holograms to be recorded with constant maximum diffraction efficiency for each hologram. Generally a large number of holograms are recorded in photopolymers that have a high dynamic range, which means that the individual diffraction efficiency for each of these holograms is very low⁶. This enables good use to be made of the recording medium, but does not allow a perfectly defined angular response curve to be obtained in the reconstruction. Therefore, certain effects regarding the influence recording each new hologram has on the photopolymer and on the decrease in its dynamic range may go unnoticed.

In this study we use the peristrophic multiplexing technique with the axis parallel to the recording media to store a small number of holograms, with a high individual diffraction efficiency (DE), in a previously developed acrylamide photopolymer approximately 1mm thick³. In this way a perfectly defined angular response curve is obtained for each of the holograms. The maximum diffraction efficiency (DE_{max}) of each hologram is seen to decrease as the number of holograms recorded increases, due to consumption of the available dynamic range. Furthermore, a progressive decrease

^{*}mos@ua.es; phone +34-965902886; fax: +34-965909750

in the width of the angular response curve is seen as the number of holograms recorded increases. Two hypotheses are proposed in order to explain this effect, which is clearly seen when various holograms are recorded with a significant individual DE (which consume the available dynamic range). The first hypothesis is based on the diffusion processes that take place after the hologram is recorded, and compares the effect on the angular response curves with what happens in a typical case of diffusion. The second is related to the progressive decrease that takes place in the dye content of the photopolymer as more holograms are recorded.

Holograms were also recorded by means of peristrophic multiplexing with the axis of rotation perpendicular to the recording medium, and the results in terms of decrease in width of the angular response curve compared with those obtained using multiplexing with the axis of rotation parallel to the medium.

2. EXPERIMENTAL

2.1 Preparation of the material

In recording materials based on PVA/AA, a solution of PVA in water forms the matrix and this is used to prepare the solution of monomer (AA) and photopolymerization initiator system: yellowish eosin (YE), triethanolamine (TEA). The PVA was supplied by Fluka, AA and TEA by Sigma and YE by Panreac.

We prepare the solutions using a conventional magnetic stirrer, under red light and in laboratory conditions ($T = 21\text{ }^{\circ}\text{C}$, relative humidity $RH = 48\%$). In Table 1 are the concentrations of the components of optimized³ photopolymer solution which is deposited in order to obtain solid layers of recording media. The solutions are deposited by gravity, in polystyrene or polymethylmethacrylate circular molds to avoid strain and distortion in the material during the water evaporation process, and left in the dark to allow the water to evaporate, while recording the laboratory conditions (T , relative humidity) during the process. When part of the water has evaporated, the “dry” material is removed from the mold, cut into squares and adhered, without the need for adhesive, to the surface of glass plates measuring $6.5 \times 6.5\text{ cm}^2$. The plates are then ready for exposure, which takes place immediately. The thickness of the material is measured using a conventional micrometer.

Components	Concentration
Polyvinyl alcohol $M_w = 130000$	13.30% w/v
Triethanolamine	0.15 M
Acrylamide	0.34 M
Yellowish eosin	9.00×10^{-5} M

Table 1. concentrations of the components of optimized photopolymer solution

2.2 Holographic set-up

To study the behavior of the photopolymer as a holographic recording material, we obtained slanted and unslanted diffraction gratings using a holographic set-up. The experimental device is shown in Figure 1. An Argon laser at a wavelength of 514 nm (peristrophic multiplexing with rotation axis parallel to hologram media) or a Nd:YAG laser with 532 nm (peristrophic multiplexing with rotation axis perpendicular to hologram media) was used to store diffraction gratings by means of continuous laser exposure. The laser beam was split into two secondary beams with an intensity ratio of 1:1. The diameters of these beams were increased to 1.5 cm with an expander, while spatial filtering was ensured. The object and reference beams were recombined at the sample at an angle θ (16.8° to the normal in argon laser and 17.4° in Nd:YAG laser) with an appropriate set of mirrors, and the spatial frequency obtained was 1125 lines/mm. The working intensity at 514 nm and 532 nm was 5 mW/cm^2 . The diffracted and transmitted intensity were monitored in real time with a He-Ne laser positioned at Bragg's angle ($\theta' = 20.8^{\circ}$) tuned to 633 nm, where the material is not sensitive. In order to obtain transmission and diffraction efficiency as a function of the angle at reconstruction we placed the plates on a rotating stage. The transmission and diffraction efficiency (TE and DE respectively) were calculated as the ratio of the transmitted and diffracted beam, respectively, to the incident power.

Fig. 1. Experimental set-up: BS, beamsplitter, Mi, mirror, SFi, spatial filter, Li, lens, Di, diaphragm, PC, data recorder.

2.3 Peristrophic multiplexing

In order to analyze the photopolymer characteristics on several holograms recording, we have used the peristrophic multiplexing technique, with rotation axis parallel to media layer and with perpendicular axis too. In the first case we put the layer on automatic rotation stage to record diffraction gratings changing layer and recording beams angle. In the second case we use an manual rotation stage additionally with the automatic rotation stage. At the end of register, we are able to reconstruc the holograms by angular scan.

3. RESULTS

3.1 Consumption of the photopolymer's dynamic range

16 holograms with 5 s of exposure time are registered in 960 μm thick photopolymer layer at 5 mW/cm^2 of recording beams total intensity. Figure 2 shows the reconstruction carried out immediately after recording; the x axis has an arbitrary origin.

Hologram 1 (the only one whose diffraction grating is unslanted) is found at the origin of the axis of rotation of the rotation stage. From this position the stage is rotated in one direction, and holograms 2 to 9 are recorded. Then the stage is brought back to its initial position and rotated in the opposite direction, when holograms 10 to 16 are recorded. Between each recording the stage is rotated 0.5° . A reduction in the DE_{max} of each hologram is seen as the number of holograms recorded increases. Holograms 10 to 16 have a much lower DE_{max} than holograms 1 to 9 due to consumption of the dynamic range. In this type of recording, with a considerable number of holograms, and in the cases in which a large number of holograms with low DE_{max} are stored, it is not easy to see changes in the width of the angular response of each hologram.

Fig. 2. Recording of 16 holograms by peristrophic multiplexing.

3.2 Decrease in width of the angular response curve when a small number of holograms with significant DE_{max} are recorded

Figure 3 shows the angular response obtained after recording two diffraction gratings in a photopolymer layer $720\text{ }\mu\text{m}$ thick. The first recording was done rotating the photopolymer layer 0.5° to the left of its original position (peak 1). The second recording was then done 0.5° to the right of the original position (peak 2). In both cases the exposure time was 10 s and the total intensity 5 mW/cm^2 . The reconstruction was carried out after both recordings had been made. The origin of the x axis is arbitrary.

Fig. 3. Recording of 2 holograms by peristrophic multiplexing.

The curve corresponding to the first diffraction grating recorded (peak 1) is seen to be wider than the second (peak 2). The width of the curve is related to the thickness of the grating stored in the photopolymer (d) by Kogelnik's equation⁷ in volume holograms:

$$\eta(t) = \Gamma \sin^2 \frac{\pi n_1(t) d}{\lambda' \cos \theta_i'} \quad (1)$$

$\eta(t) = DE \times 10^{-2}$, Γ is a factor that takes into account the loss of light due to absorption, dispersion and reflection. θ_i' is the angle between the reconstruction beam and the normal to the recording medium, measured inside the material. $n_1(t)$ is the modulation index in the region of maximum luminous intensity and λ' is the wavelength of the reconstruction laser beam. A decrease in the width of the angular response curve necessarily implies an increase in the thickness of the stored diffraction grating, provided that the other parameters remain constant.

In order to interpret the phenomena observed in Figure 3 two different hypotheses are used. One of them considers that in the type of photopolymer we are dealing with, diffusion of the components inside the material is greatly favoured once the hologram has been recorded, especially in thick layers. The increase in width of the angular response curve is characteristic of diffusion phenomena, as can be seen in Figure 4, corresponding to a layer of photopolymer 950 μm thick, which shows the angular response obtained after recording (0 days) and after storing the hologram for 19 days in the dark.

Fig. 4. Diffusion effects after 0 and 19 days of hologram storage time.

After 19 days the DE_{max} decreases from 72% to 66%, and the angular interval of the DE_{max} increases (from approximately 0.4° to 0.6°). The increase in width of the curve is related to the decrease in thickness of the stored diffraction grating (equation 1) due to diffusion phenomena. Although an increase in width of the angular response curve is characteristic of diffusion phenomena, it takes much longer for this effect to be seen than the interval between two recordings in the case of multiplexing.

Assuming that in the case shown in Figure 3, a similar diffusion process takes place, the short time elapsed between the first recording and the second would hypothetically be sufficient for diffusion of the polyacrylamide chains (when reconstruction is carried out), to be such that it has a greater effect on the results obtained for the first grating than for the second. Therefore, some of these chains, formed in the bright zones during recording diffuse transversally towards the dark zones, where the initial concentration was zero, thereby producing a decrease in refractive index modulation which results in peak 1 becoming wider. For peak 2 the time elapsed would not be sufficient and therefore the thickness of the stored grating and the refractive index modulation are maintained, so the width of the curve is less and its DE_{max} is greater than for peak 1.

Figure 5 shows the angular response after recording 3 multiplexed diffraction gratings in an 890 μm thick photopolymer layer with an individual exposure of 16 s (82 mJ/cm^2).

Fig. 5. Recording of 3 holograms by peristrophic multiplexing.

The 3 holograms reach a high maximum diffraction efficiency ($DE_{max} \approx 40\%$), and the dynamic range is $M/\# = 1.93$. The same phenomenon as regards the width of each curve is seen, although now the difference between the second (peak 2) and third recording (peak 3) is less than that between the first (peak 1) and the second (width of each curve: peak 1 = 0.35° , peak 2 = 0.24° , peak 3 = 0.21° , quotients between the widths: peak1/peak2 = 1.46, peak2/peak3 = 1.14). The peak2/peak3 quotient has a value very close to 1, indicating that the width of peak 2 is not affected by a hypothetical diffusion effect. However, initially progress of diffusion phenomena is directly proportional to the time the molecules have to diffuse (maximum time for the grating corresponding to peak 1 and minimum time for the grating corresponding to peak 3). The fact that the relationship between the widths is not maintained indicates that the effect seen is not related to a diffusion process. It should be remembered that the diffusion processes that take place after the holograms are recorded are only significant if the polymeric matrix allows sufficiently high diffusion coefficients to be reached, and this is not the case with the composition used for this photopolymer.

Figure 6 shows the angular response after recording two diffraction gratings by peristrophic multiplexing with the axis of rotation perpendicular to the media in a $900\text{ }\mu\text{m}$ thick photopolymer layer, with an individual exposure of 15 s and total intensity of 5 mW/cm^2 . The angular separation between the gratings is 90° .

Fig. 6. Angular response after recording of 2 holograms.

The same phenomenon of variation in width of the angular response curve is again seen, although the time elapsed between the two recordings is minimal (typical value of 1 minute). The curve at 0° , recorded first, has an approximate width of 0.7° , whereas the width is 0.5° for the curve at 90° . On the other hand, the two gratings have the same $DE_{\max}$. This value should be lower for the first grating if it were affected by a diffusion phenomenon, in order for the relationship between d and $n_1(t)$ expressed in equation 1 to be fulfilled.

When 6 holograms are recorded with this type of multiplexing in a photopolymer of the same thickness, the same behaviour occurs, as can be seen in the reconstruction performed after the recording (Figure 7).

Fig. 7. Angular response after recording of 6 holograms.

The $DE_{\max}$ is reached in an angular interval of from 1.2° in the first recording (hologram 1) to 0.4° in the last recording (hologram 6).

The second explanation of the effects observed considers the change in dye content that takes place during recording of the holograms, rather than the influence of diffusion. Recording holograms by means of multiplexing involves consumption of a proportion of the available dynamic range of the photopolymer (Figure 2), and this is associated with a decrease in the content of polymerizable monomer and dye. The decrease in monomer content affects the value of $DE_{\max}$ reached in each hologram (in multiplexing with constant exposure time). Regarding the dye, in all the cases analyzed, there is a high concentration of dye available during recording of the first grating, with the result that only the most superficial layers of the photopolymer are involved (as a result of attenuation of the light due to absorption of the dye). Consequently, the width of the angular response curve is greater since the grating occupies less thickness (equation 1). During the second recording, the concentration of available dye has decreased, so the light may activate the dye molecules in deeper layers of the media thickness. Thus the width of the angular response curve is less than in the first case, which indicates that there is now a greater thickness for the second diffraction grating, and so on. This hypothesis is in complete agreement with the findings, as opposed to the one based on diffusion processes. Moreover, important conclusions may be drawn from this study, since this explanation is related to the interpretation of the curves obtained in the case of diffusion processes like the one represented in the Figure 4. From this, it may be deduced that diffusion processes have a greater effect on the deepest zones (otherwise the stored grating would disappear completely) where there is least polymerization because of attenuation of the light due to absorption of the dye. Thus, there is a decrease in the thickness occupied by the grating and consequently an increase in the width of the angular response curve. This implies the existence of an effective grating thickness, or fraction of thickness of the grating recorded in the photopolymer, capable of producing diffraction.

CONCLUSION

From the experiments performed it may be concluded that recording a small number of holograms with a high $DE_{\max}$ allows a correct follow-up of the evolution of the shape of the angular response curves. It was found that as the available dynamic range of the photopolymer is consumed, progressively lower values of $DE_{\max}$ and narrower angular response curves are obtained. This effect is very similar to that which occurs as a result of the diffusion processes that take place after recording the hologram. However, the decrease in dye content in the photopolymer, due to the decrease in light absorption, is shown to be the cause of this effect seen in the case of multiple recordings, from which it may also be deduced that the dye concentration (a derivative of xanthene) is a factor that limits the dynamic range of this type of photopolymer. Comparison of a recording of various holograms and a diffusion process in the case of a single hologram makes it possible to distinguish between the thickness of the stored grating and that of the photopolymer. Furthermore, following this reasoning, the concept of effective grating thickness or fraction of grating thickness capable of producing diffraction efficiency may be inferred. Therefore, in every stored diffraction grating, a fraction of its thickness must be located deep down in the thickness of the hologram, where there are polymerized molecules that do not contribute to the diffraction efficiency reached.

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REFERENCES

1. R. A. Lessard, and G. Manivannan, "Holographic Recording Materials: an overview," in *Holographic Materials*, T. J. Trout, ed., Proc. SPIE **2405**, 2 (1995).
2. S. Martin, P. E. L. G. Leclerc, Y. L. M. Renotte, V. Toal, and Y. F. Lion, "Characterization of an acrylamide-based dry photopolymer holographic recording material", *Optical Engineering* **33**, 3942-3946, 1994.
3. M. Ortuño, S. Gallego, C. García, C. Neipp, A. Beléndez, and I. Pascual, "Optimization of a 1 mm thick PVA/acrylamide recording material to obtain holographic memories: method of preparation and holographic properties", *Applied Physics B* **76**, 851-857, 2003.
4. H. J. Coufal, and D. Psaltis: *Holographic Data Storage* (G. T. Sincerbox, Springer-Verlag, New York 2000).
5. F. Mok, G. W. Burr, and D. Psaltis, "A system metric for holographic memory systems", *Optics Letters* **21**, 896-898, 1996.
6. K. Y. Hsu, S. H. Lin, and Y. H. W. T. Whang "Experimental characterization of phenanthrenequinone-doped poly(methyl methacrylate) photopolymer for volume holographic storage", *Optical Engineering* **42**, 1390-1396, 2003.
7. H. Kogelnik "Coupled Wave Theory for Thick Hologram Gratings", *Bell System Technical Journal* **48**, 2909-2947, 1969.