Optik 114, No. 8 (2003) ■■■–■■■
© 2003 Urban & Fischer Verlag
http://www.urbanfischer.de/journals/optik

Optik Optics
International Journal for Light and Electron Optics

Influence of the fringe visibility on the characteristics of holograms recorded in photopolymer material

Sergi Gallego[2], Manuel Ortuño[2], Cristian Neipp[1], Augusto Beléndez[1], Inmaculada Pascual[2]

[1] Departamento de Física, Ingeniería de Sistemas y Teoría de la Señal,
Universidad de Alicante, Apartado 99, E-03080 Alicante, Spain
[2] Departamento Interuniversitario de Óptica, Universidad de Alicante, Apartado 99, E-03080 Alicante, Spain

Abstract: In the last years many diffusion models have been proposed to explain the hologram formation in photopolymers. This material has a lot of attractive features as self-developing, high storage capacity (ease process to obtain thick dry layers), high diffractions efficiencies etc. In this work the availability of a first harmonic diffusion model proposed by C. Neipp et al. to describe the recording process for different visibilities is demonstrated. The problems to achieve high diffraction efficiencies at low spatial frequencies for PVA/acrylamide based photopolymer are also solved, and a possible description of this phenomenon is obtained using the diffusion model proposed.

Key words: Holography – volume holographic gratings – holographic recording materials

1. Introduction

Photopolymers are promising materials for use in holography. This material presents some well-known advantages, such as the possibility of obtaining high efficiencies (up to 100%), stability, high thickness (to use as holographic data storage medium) and relativity low cost. During the last years many diffusion models have been used to predict the photopolymer behaviour during the recording process. In most of these models two phenomena are supposed that create the hologram: the monomer polymerization and the monomer diffusion.

The search to describe the hologram formation using simple and intuitive models is not finished. Although the mechanism of photo-polymerization is somewhat complex, if some reasonable assumptions are made the formation of the hologram can be understood in the basis on simple considerations. For instance, the model proposed by C. Neipp et al. [1] has been used to predict the behaviour of a PVA/acrylamide based dry photopolymer. Although the model is simple, it is able to describe the material behaviour when there are changes in some parameters as monomer concentration, exposure intensity, thickness and when a monomer acting as a cross-linker is introduced. The model also predicts the existence of residual monomer after the grating has been recorded [2]. It has been demonstrated that this residual monomer plays an important role in the stabilization of the hologram by bleaching, so controlling the amount of monomer remaining in the hologram after exposure is important. Although this model is able to explain some interesting features of photopolymer materials, it cannot explain the cut-off of diffraction efficiencies for high (up to 1500 lines/mm) and low (down to 800 lines/mm) spatial frequencies observed in acrylamide-based photopolymers [3]. This effect for high spatial frequencies is due to polymer chains growing away from their initiation point and causing the record of a smeared profile [4]. A more accurate and complex theory, such as that proposed by J. T. Sheridan et al. [5] is needed to explain these questions.

In this paper, we firstly evaluate the experimental results obtained when the material is exposed to a sinusoidal interference pattern with different beam intensity modulations (see eqs. 1 and 2) and is recorded with a spatial frequency of 1125 lines/mm. The diffraction efficiency versus exposure curves have been fitted by the first harmonic diffusion model proposed in reference [1] in order to complete the description of the material behaviour by this model.

We supposed that the material is illuminated to an interference pattern of the form:

$$I(x) = I_T[1 + m\cos(K_g x)] \tag{1}$$

where m is the beam intensity modulation defined as:

$$m = \frac{2\sqrt{I_R I_O}}{I_R + I_O}\,. \tag{2}$$

In eq. (1), K_g is the grating constant and $I_T = I_R + I_O$, where I_R and I_O are the reference and object wave intensities, respectively.

Received 2 July 2003; accepted 20 September 2003.

Correspondence to: S. Gallego
Fax: ++34-96-5903464
E-mail: pascual@ua.es

0030-4026/03/114/08-000 $ 15.00/0

In second place we compare the results at 1125 lines/mm with the results at 545 lines/mm. The possibility of obtaining maximum diffraction efficiencies is discussed with a series of experimental results. In order to achieve high index modulations at low spatial frequencies the composition and thickness must be optimised. Finally, in the basis of the first harmonic diffusion based model, possible causes of the problem to record low spatial frequencies in photopolymers are explained.

The diffusion constant, D, is a characteristic of each material. This parameter does not change with the spatial frequencies. And the variation of concentrations in the initial solution does not affect this value too much. But the diffusion time constant, τ_D, defined by eq. (3) is a function of the spatial frequencies (K_g):

$$\tau_D = \frac{1}{DK_g^2}\,. \tag{3}$$

When eq. 3 is analysed, it can be seen that when the spatial frequency decreases, whenever the diffusion constant is maintained constant, diffusion time is increased. Therefore, monomer diffusion losses importance in the hologram formation process for low spatial frequencies.

2. Experimental set up

The holograms were recorded by using an Argon laser of wavelength 514 nm (fig. 1). The intensity was of 6 mW/cm^2. The beams impinged on the material symmetric to the normal to the recording material, with a beam ratio of 1 : 1, so unslanted transmission gratings were recorded. The gratings were recorded with two different spatial frequencies: 545 lines/mm and 1125 lines/mm.

To prepare the material a method similar to that described in other papers was used. A polymerization solution was prepared using polyvinylalcohol 18–88 (PVA) provided by Fluka as a binder, acrylamide (AA) as monomer and yellowish eosin as dye. For this type of photopolymer we used n,n′ methylene-bis-acrilamida (BMAA), which is the more common crosslinker utilized in the bibliography [6, 7, 8]. It has been demonstrated that the inclusion of this component in the solution a better conservation of the holograms and a higher refraction index modulation is achieved.

It should be noticed that the particular compositions of the layers must be modified taking into account the spatial frequency at which the hologram will be recorded. For the gratings recorded with 545 lines/mm the monomer concentration and the thickness of the layer must be considerably increased, with respect to those of a 1125 lines/mm grating. This is due to the worse response of the material at 545 lines/mm than at 1125 lines/mm.

The thickness of the layer was obtained by method proposed by A. Beléndez et al. [9] and the optical thickness is obtained analyzing the angular response by fitting based in Kogelnik's theory to unslanted transmission gratings (see eq. 4)

$$\eta = \exp\left(-\alpha d/\cos\theta'\right)\sin^2\left(\frac{\pi n_1 d}{\lambda\cos\theta'}\right), \tag{4}$$

where α is the coefficient of absorption and scattering, d is the thickness, θ' is the reconstruction angle inside of the material, λ is the wavelength of reconstruction and n_1 is the refraction index modulation.

3. Experimental results

Information of the recording process in photopolymer materials has usually been obtained by studying the temporal evolution of the diffraction and transmission efficiency replayed on Bragg for the diffraction grat-

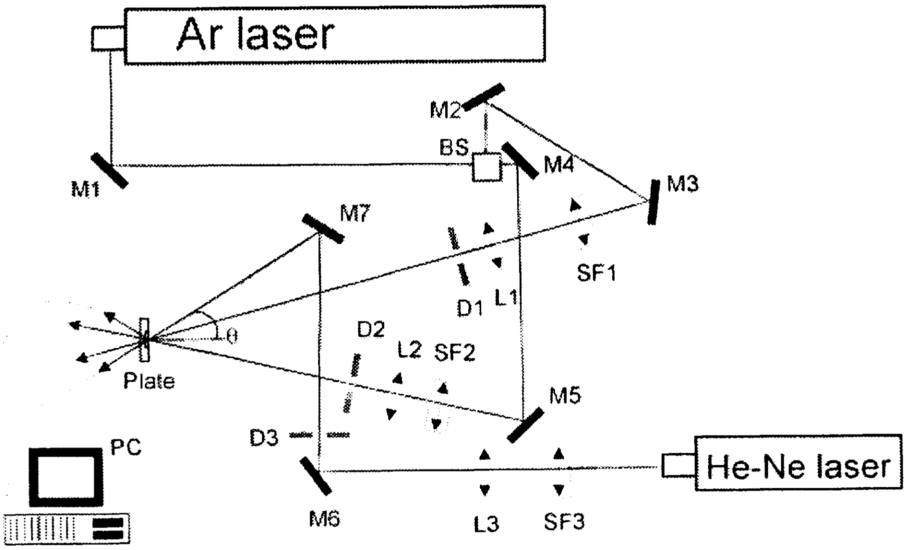


Fig. 1. Experimental set: BS, beam splitter, Mi, mirror, SFi, spatial filter, Li, lens, Di, diaphragm, PC, personal computer.

ings recorded on the material [1, 10]. By doing this, the different models proposed in the literature can be checked. These models should accomplish to explain the effects that the variation of different experimental conditions produce in the theoretical functions. In particular, the model proposed in ref. 1, explains the influence on the diffraction versus time curves that the following parameters have: variation of the initial monomer concentration, variation of the thickness or variation of the recording intensity. In this work, the model will be used to predict the influence of the beam intensity modulation on the temporal evolution of the diffraction efficiency and to analyse the recording process at a low spatial frequency ($\sim$550 lines/mm).

3.1. Different beam modulation

Figure 2 shows the diffraction efficiency as a function of the exposure for a transmission diffraction grating recorded on a PVA/acrylamide photopolymer at a spatial frequency of 1125 lines/mm. For the grating studied the composition was optimised in order to achieve maximum diffraction efficiency at Bragg angle for an intensity modulation, m, of 1. The dotted points represent the experimental data whereas the continuous line described the theoretical function obtained with the proposed model. The energy sensitivity is of $\sim$240 mJ/cm^2. For the dry photopolymer layer the optic thickness was of 53 µm, valour obtained by fitting the theoretical function of the angular response of the diffraction efficiency (continuous line in fig. 2b), given by Kogelnik's theory, to the experimental data (dots in fig. 2b). The experimental data in fig. 2b correspond to the diffraction grating considered in fig. 2a, once the recording process is stopped (corresponding to the last point of fig. 2a).

In order to study the influence of the beam intensity modulation, m, in the recording process a series of transmission diffraction gratings were recorded by using different values of m: 0.94, 0.8, 0.7, 0.55 and 0.43. Figures 3–7 show the temporal (figures with subscript (a)) and angular responses (figures with subscript (b)) of the diffraction efficiency for the different gratings recorded. For the gratings considered in figs. 3–7, the thickness of the layer were measured as 47, 41, 48, 55 and 45 µm, respectively. The theoretical curves presented in figs. 3–7 where obtained by fixing all of the parameters of the model proposed, with exception of the beam intensity modulation, and the thickness of

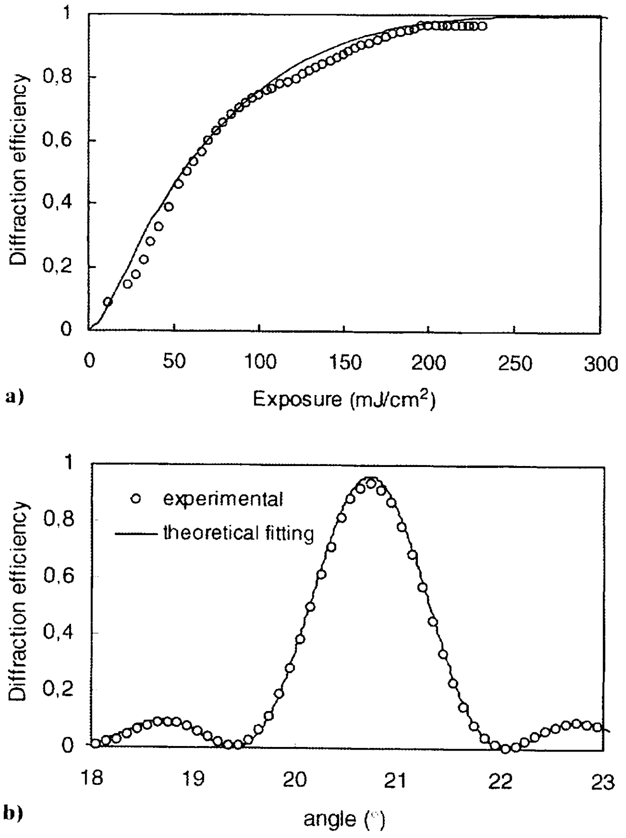


Fig. 2. a) Diffraction efficiency as function of exposure, for $m = 1$ and 53 ± 2 µm of thickness. b) Experimental diffraction efficiency versus reconstruction angle fitting by Kogelnik's coupled wave theory ($n_1 = 0.0053 \pm 0.0001$, $d = 53 \pm 2$ µm).

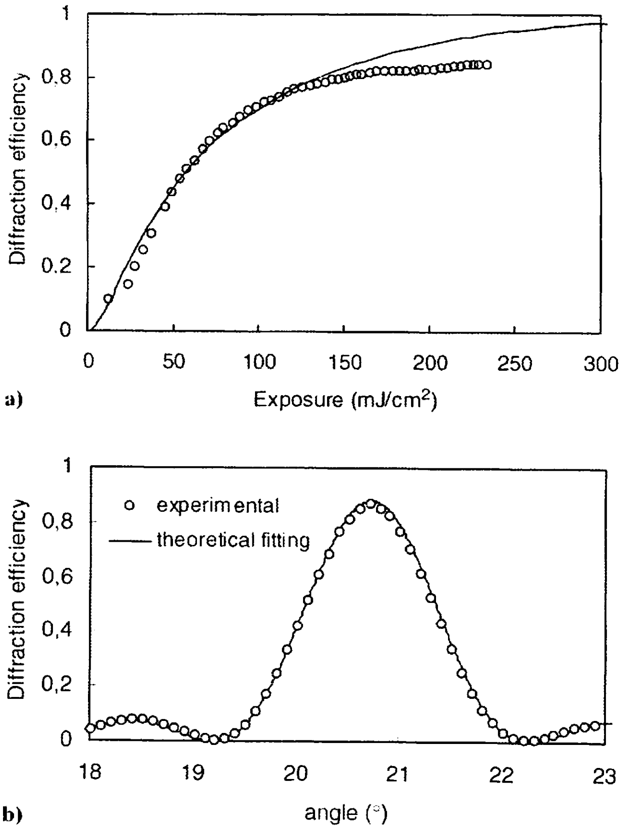
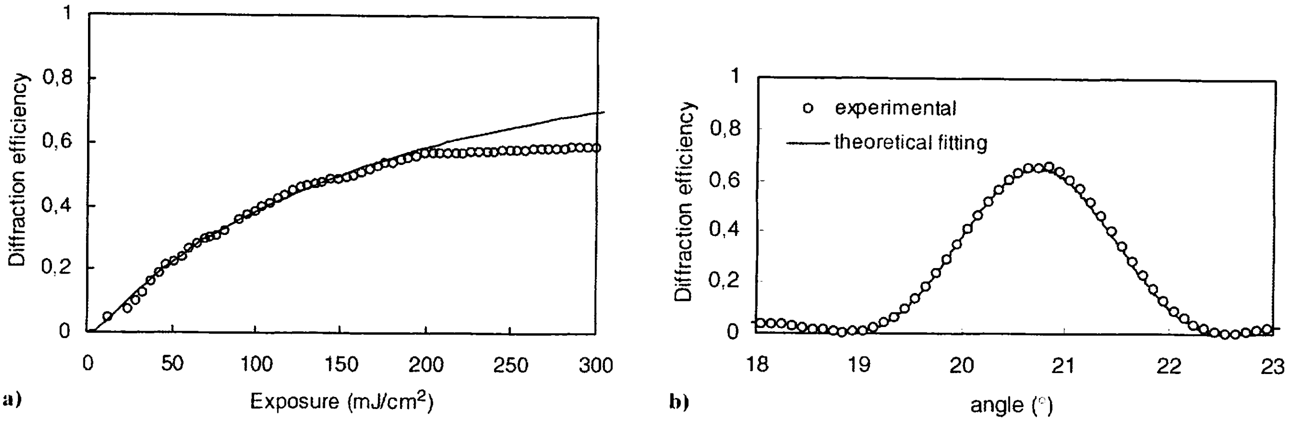
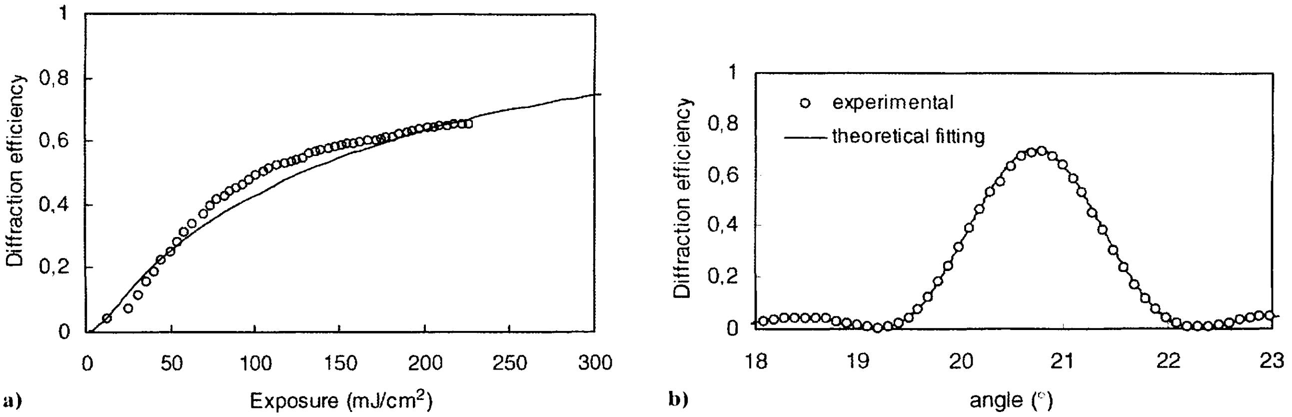


Fig. 3. a) Diffraction efficiency as function of exposure, for $m = 1$ and 47 ± 2 µm of thickness. b) Experimental diffraction efficiency versus reconstruction angle fitting by Kogelnik's coupled wave theory ($n_1 = 0.0052 \pm 0.0001$, $d = 47 \pm 2$ µm).



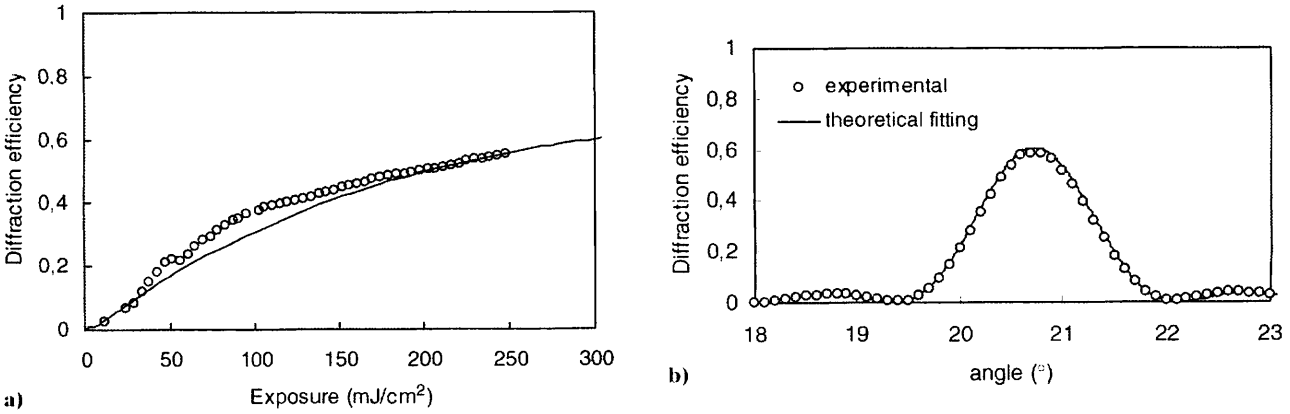
a) b)

Fig. 4. a) Diffraction efficiency as function of exposure, for $m = 0.8$ and 41 ± 2 μm of thickness. b) Experimental diffraction efficiency versus reconstruction angle fitting by Kogelnik's coupled wave theory ($n_1 = 0.0046 \pm 0.0001$, $d = 41 \pm 2$ μm).



a) b)

Fig. 5. a) Diffraction efficiency as function of exposure, for $m = 0.7$ and 48 ± 2 μm of thickness. b) Experimental diffraction efficiency versus reconstruction angle fitting by Kogelnik's coupled wave theory ($n_1 = 0.0041 \pm 0.0001$, $d = 48 \pm 2$ μm).



a) b)

Fig. 6. a) Diffraction efficiency as function of exposure, for $m = 0.55$ and 55 ± 2 μm of thickness. b) Experimental diffraction efficiency versus reconstruction angle fitting by Kogelnik's coupled wave theory ($n_1 = 0.0033 \pm 0.0001$, $d = 55 \pm 2$ μm).

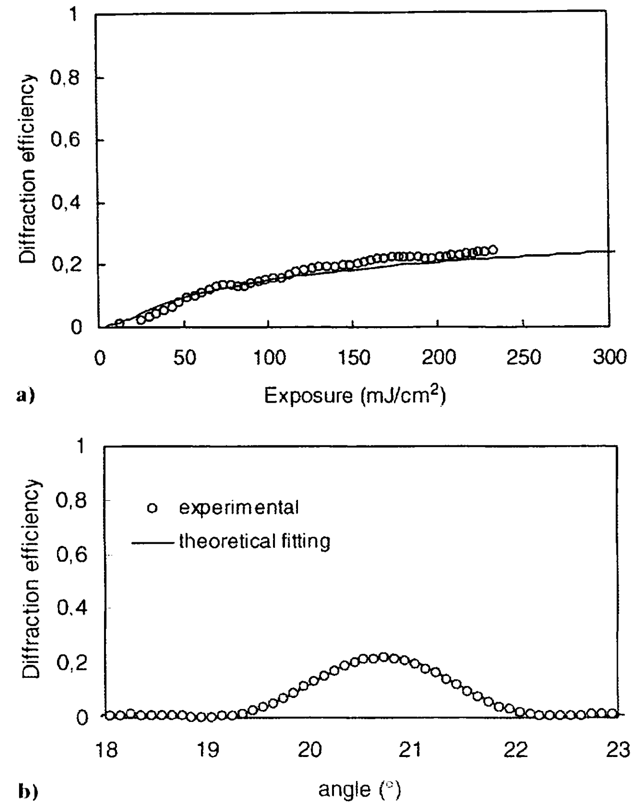


Fig. 7. a) Diffraction efficiency as function of exposure, for $m = 0.43$ and 45 ± 2 µm of thickness. b) Experimental diffraction efficiency versus reconstruction angle fitting by Kogelnik's coupled wave theory ($n_1 = 0.0022 \pm 0.0001$, $d = 45 \pm 2$ µm).

the layer (obtained after the fitting of the angular response of the diffraction efficiency). As can be seen the dependence of the temporal evolution of the diffraction efficiency with the beam intensity modulation is well described by the theoretical model.

3.2. Low spatial frequency (545 lines/mm)

It is well known that the PVA/Acrylamide based photopolymers have problems to record low (under 700 lines/mm) and high frequencies (over 2500 lines/mm). In order to obtain maximum diffraction efficiency, the monomer concentration of the layer must be optimised. For instance, in the particular case of a grating recorded with 545 lines/mm the monomer concentration must be increased to the double with respect to that recorded with a spatial frequency of 1125 lines/mm. In fig. 8a, the experimentally measured diffraction efficiency as a function of the exposure is shown for a grating with 70 µm of thickness. The energy sensitivity was of 200 mJ/cm². On the other hand fig. 8b shows the angular response of the diffraction efficiency, the continuous line corresponds to the theoretical fit using Kogelnik's theory. It can be seen that the diffraction efficiency at Bragg angle was maximum, near 97%.

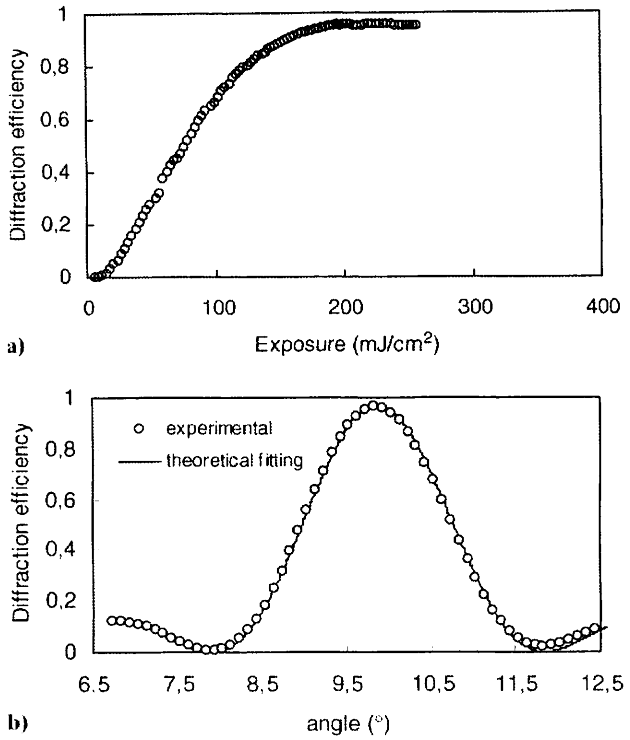


Fig. 8. a) Experimental diffraction efficiency as function of exposure, for spatial frequency of 545 lines/mm and 70 ± 2 µm of thickness. b) Experimental diffraction efficiency versus reconstruction angle fitting by Kogelnik's coupled wave theory ($n_1 = 0.0046 \pm 0.0001$, $d = 70 \pm 2$ µm).

In figs. 9a and 9b the results corresponding to a 140 µm thickness transmission grating are shown. In this case, the thickness is higher than that of the diffraction grating considered in fig. 8, therefore overmodulation effects can be seen. Since the monomer and dye concentrations were the same as those for the previous grating [11], higher noise can also be observed in this case. The diffraction efficiency reaches a maximum at an exposure of 175 mJ/cm², an 89% of light is diffracted.

In fig. 10, three theoretical curves are represented. The first one (the continues line) represents the behaviour of the material in the recording process for a layer of 70 µm if the parameters are the same than the "standard" gratings (around 1100 lines/mm). But when we work at low spatial frequencies the monomer diffusion loss importance in the hologram formation. This effect can be seen in second curve (triangles) where diffusion time constant considered is around four times higher than the standard gratings, because the distance between two consecutive fringes is double. The index modulation achieve for a same energy is now smaller than the first curve. Nevertheless the slope of the curve is similar than the first curve, because the slope is related to the speed of the reaction represented by polymerization rate constant. This constant depends of many factors (the final monomer concentration in dry

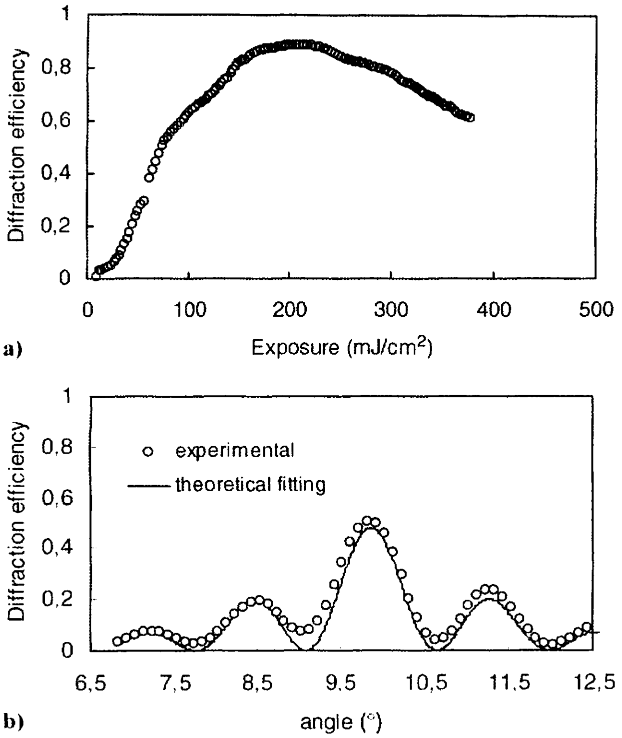


Fig. 9. **a) Experimental diffraction efficiency as function of exposure, for spatial frequency of 545 lines/mm and 140 ± 2 μm of thickness. b) Experimental diffraction efficiency versus reconstruction angle fitting by Kogelnik's coupled wave theory ($n_1 = 0.0046 \pm 0.0001$, $d = 140 \pm 2$ μm).**

layer, the presence of monomer crosslinker, dye concentration ...). Therefore this loss of importance of monomer diffusion causes a reduction in the polymerization constant. This last effect in the hologram formation can be observed in third curve (crossings) where the considered polymerization rate constant is half than in the rest of curves. This third curve is similar than experimental data in two mainly characteristics: slope and energy sensitivity (around 200 mJ/cm²) and confirm our hypothesis.

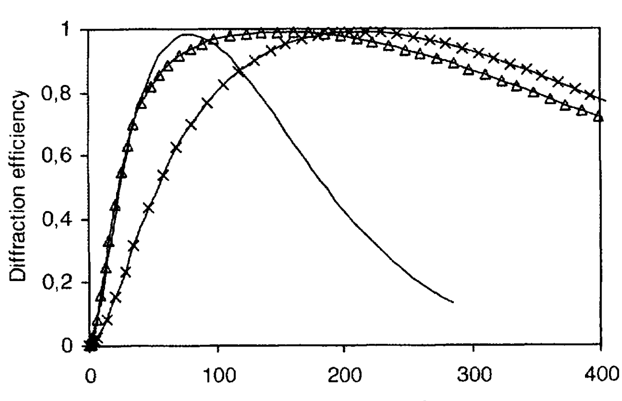


Fig. 10. **Theoretical diffraction efficiencies. Standard parameters (continues line), high diffusion time, around 150 s (triangles), and for high diffusion time and low polymerization rate constant (0.01 cm² mW⁻¹ s⁻¹).**

4. Conclusion

The experimental response of an acrylamide-base polymeric film to different beam intensity modulations fitting by the first harmonic diffusion model has been presented. We found the optimum composition of layers to obtain layers with maximum diffraction efficiency at Bragg's angle or overmodulation. We demonstrate that the first harmonic diffusion model is valid to study spatial frequencies under 1125 lines/mm. For all, if we wont storage holographic optic elements (for example lens) we must calc de median spatial frequencies previously, and then prepare the solution with appropriated concentrations to the diffractions efficiencies (or refraction index modulation) are optimum for these frequencies.

Acknowledgements. This work was supported by the Ministerio de Ciencia y Tecnología, CICYT, Spain, under project MAT2000-1361-C04-04. 04 and by the "Oficina de Ciencia y Tecnología" (Generalitat Valenciana, Spain) under project GV01-130.

References

[1] Neipp C, Gallego S, Ortuño M, Márquez A, Álvarez M, Beléndez A, Pascual I: Fist harmonic diffusion based model applied to PVA/acrilamida based photopolymer. J. Opt. Am. B (in press)
[2] Gallego S, Neipp C, Ortuño M, Márquez A, Álvarez M, Beléndez A, Pascual I: Diffusion based model to predict the conservation of holographic gratings recorded in PVA/acrylamide photopolymer. Appl. Opt. (accepted by minor revision)
[3] Martin S, Feely CA, Toal V: Holographic recording characteristics of an acrylamide-based photopolymer. Appl. Opt. **36** (1994) 5757–5768
[4] Lawrence JR, O'Neill FT, Sheridan JT: Photopolymer holographic recording material. Optik **112** (2001) 449–463
[5] Sheridan JT, Downey M, O'Neill FT: Diffusion based model of holographic grating formation in photopolymers: generalized non-local material responses. J. Opt. A.: Pure Appl. Opt. **3** (2001) 477–488
[6] Martin S, Leclere PELG, Renotte YLM, Toal V, Lion YF: Characterization of an acrylamide-based dry photopolymer holographic recording material. Opt. Eng. **33** (1994) 3942–■■■
[7] O'Neill FT, Lawrence JR, Sheridan, JT: Thickness variation of self-processing acrylamide-based photopolymer and reflection holography. Soc. Photo-Opt. Instrument. Eng. **40** (2001) 533–539
[8] Yao H, Huang M, Chen Z, Hou L, Gan F: Optimization of two-monomer-based photopolymer used for holographic recording. Mater. Lett. **56** (2002) 3–8
[9] Beléndez A, Beléndez T, Neipp C, Pascual I: Determination of the refractive index and thickness of holographic silver halide materials by use of polarized reflectances. Appl. Opt. **41** (2002) 6802–6808
[10] Neipp C, Gallego S, Ortuño M, Márquez A, Beléndez A, Pascual I: Characterization of a PVA/Acrylamide photopolymer. Influence of a cross-linking monomer in the final characteristics of the hologram. Opt. Comm. (in press)
[11] Coufal HJ, Psaltis D: Holographic data storage. In Sincerbox GT (ed.): Holographic data storage pp 181. Springer-Verlag, New York 2000