PROCEEDINGS OF SPIE
SPIE—The International Society for Optical Engineering

Photon Management

Frank Wyrowski
Chair/Editor

27–28 April 2004
Strasbourg, France

Sponsored and Published by
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Volume 5456

Please use the following format to cite material from this book:

Author(s), "Title of Paper," in *Photon Management*, edited by Frank Wyrowski, Proceedings of SPIE Vol. 5456 (SPIE, Bellingham, WA, 2004) page numbers.

ISSN 0277-786X
ISBN 0-8194-5383-8

Published by
SPIE—The International Society for Optical Engineering
P.O. Box 10, Bellingham, Washington 98227-0010 USA
Telephone 1 360/676-3290 (Pacific Time) · Fax 1 360/647-1445
http://www.spie.org

Printed in the United States of America.

High-efficiency volume holograms recording on Acrylamide And N,N'methylene-bis-acrylamide photopolymer with pulsed Laser

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ABSTRACT

In order to achieve higher diffraction efficiencies of the volume gratings stored in acrylamide based photopolymer, we introduce in the photopolymer a crosslinker (N,N'methylene-bis-acrylamide). The presence of this component increase the rate polymerization and the modulation of refraction index. The recording was performed using a holographic copying process. The original was a grating of 1000 lines/mm processed using silver halide sensitized gelatine, with diffraction efficiency around 50 % for a reconstruction wavelength of 532 nm. The main beam was split in two secondary beams by the original grating, with an intensity ratio 1:1. The results obtained using the new composition of material are compared with the composition without crosslinker. In the other hand the no linearity of the material's response is also studied comparing the energetic sensitivity, diffraction efficiencies and index modulation of gratings recorded with pulsed and continuous laser. This study is realized fitting the angular scan of each grating using Kogelnik's theory. The gratings are recorded with wavelength of 532 nm when pulsed exposure is used and with wavelength of 514 nm when continues exposure is used. Using pulsed laser at 532 nm the photopolymer without crosslinker presents the diffraction efficiencies lightly smaller than 60%. In the other hand when the crosslinker has been introduced in photopolymer composition, the diffraction efficiencies achieves are higher than 85 %.

Keywords: Holography, holographic recording materials, data storage, photopolymers

1. INTRODUCTION

The study of photopolymer materials based in polyvinyl alcohol/acrylamide (PVA/AA) has spurred a great deal of interest because their many attractive features to be used as holographic memories [1,2]. The behaviour of this type of holographic recording materials is well know using continuous-wave [3] and pulsed-laser irradiation [4]. The main problem of used continuous exposure are the especial requirements on the setups (conditions of recording and laboratory premises). The pulsed exposure eliminates this disadvantages and enables one to obtain new results in the fields of graphic technique, holographic interferometry... This substantial simplification of the recording processes and the decrease in the labour of producing holograms is very important in order to optimize the industrial production of holograms. In this work the pulsed recording is done by copying process [4-6]. The results obtained recording volume holograms generated by means of a copying process using pulsed exposure are employed in the investigation of photochemical an photophysical processes and in the study of the influence of the pulse energy and number of pulses in the diffraction efficiency and energetic sensitivity [7-10]. Although the reached diffraction efficiencies, using this method, they have not been optimal, are only around 60%.

For this reason, in order to achieve higher diffraction efficiencies of the volume gratings stored in acrylamide based photopolymer, we introduce in the photopolymer composition N,N'methylene-bis-acrylamide (BMA) as crosslinker. The presence of the monomer crosslinker has been studied by many authors [11-13] and the importance of this component in the polymerization rate constant, in the polymer index refraction and in the hologram conservation have been evaluated. In reference [11] is demonstrated that the presence of BMA increase the index modulation and the energetic sensitivity achieve in the recording process using continuous exposure.

In this paper the recording was performed using a holographic copying process (method 1). The original was a grating of 1000 lines/mm processed using silver halide sensitized gelatine, with diffraction efficiency around 50% for a reconstruction wavelength of 532 nm. The main beam was split in two secondary beams by the original grating, with an intensity ratio 1:1. The samples were exposed and holograms recorded with a collimated beam from a frequency-doubled Nd:YAG (532 nm) Q-switched laser. The energy of each pulse was 0.6 mJ/cm^2, the pulsed duration ~8 ns and the frequency was of 10 Hz, with this frequency high diffraction efficiencies was achieve as can be seen in reference [4]. When the gratings were recorded with continues laser (method 2), the laser employed was an Argon laser tuned at 514 nm.

The experimental results obtained in this work show that the presence of the monomer crosslinker increase the diffraction efficiency to values higher than 85%. In the other hand the results obtained using the composition of material with crosslinker are compared with the composition without crosslinker using pulsed exposure. The no linearity of the material's response is also studied comparing the energetic sensitivity, diffraction efficiencies and index modulation of gratings recorded with pulsed and continues laser.

2. EXPERIMENTAL SETUP

Figure 1 shows the geometry of the irradiation arrangement, used to store the holograms by means of pulsed beam (method 1). The process is a copying one, in which the master is placed in direct contract with the photopolymer in such a way that the transmitted and diffracted beams interfere in the photopolymer film, provided that the paths of the beams differ sufficiently to produce interference.

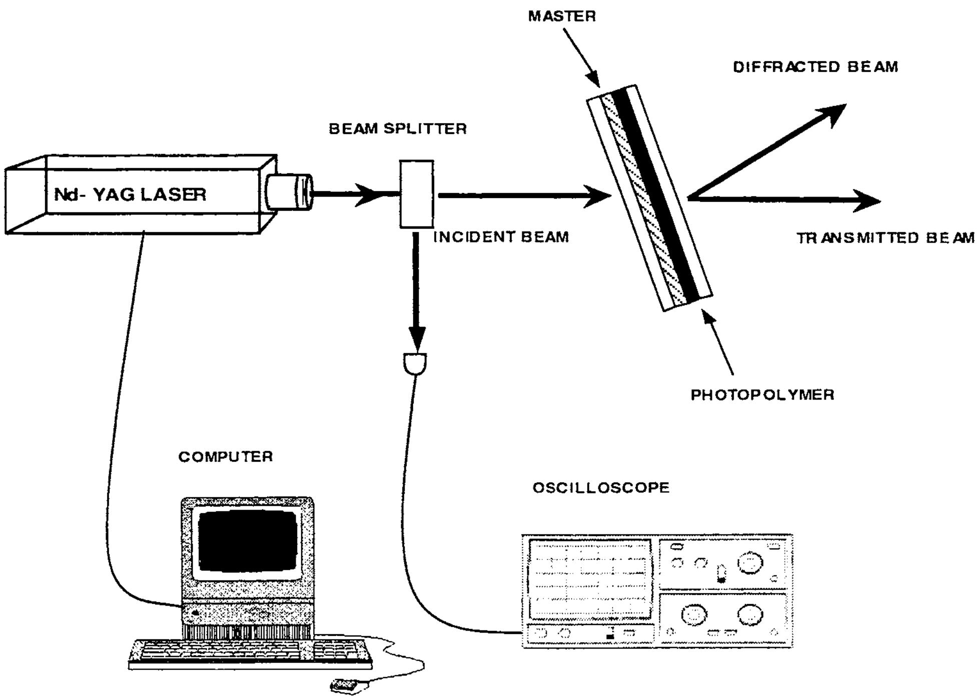

Fig 1. Experimental setup to record holograms using pulsed laser (method 1).

A beam splitter placed in the way of the pump beam directs about 10% of the laser beam energy toward a detector. The generated signal was recorded with an oscilloscope. The remaining 90% of the original pattern is direct to the master so that the diffracted and transmitted beams impinge onto the photopolymer film. A computer controls both the number and repetition of the pump pulses. The diffracted intensity and the angular scan was monitored with a He-Ne laser tuned at 633 nm, at which wavelength the material does not absorb.

The photopolymer employed to store the gratings, has acrylamide (AA) as monomer, using polyvinylalcohol 18-88 (PVA) provided by Fluka as a binder, triethanolamine (TEA) as coinitiator, yellowish eosin as dye and BMA as crosslinker in the material type 1. The exact composition of the two type the materials employed can be seen in table 1.

	Type 1	Type 2
ACRYLAMIDE (AA)	0.44 M	0.44 M
N,N' METHYLENE-BIS-ACRYLAMIDE (BMA)	0.04 M	0 M
TRIETHANOLAMINE (TEA)	0.20 M	0.20 M
YELLOWISH EOSIN (YE)	2.5×10^{-4} M	2.5×10^{-4} M
POLYVINYLALCOHOL 18-88 FLUKA (PVA)	7.5% w/v	7.5% w/v

Table 1. The photopolymer solution composition.

The composition type 1 is used to record holograms by continuous exposure too, and after analyze the differences between the behaviour of the same material composition for the two different methods of recording diffraction gratings. For recording holograms using continuous exposure a typical experimental setup is used as can be seen in figure 2. An Argon laser at a wavelength of 514 nm 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 of 16.8° to the normal with an appropriate set of mirrors, and the spatial frequency obtained was 1125 lines/mm. The working intensity at 514 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 (20.8°) tuned to 633 nm, where the material does 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 having into account Fresnel losses.

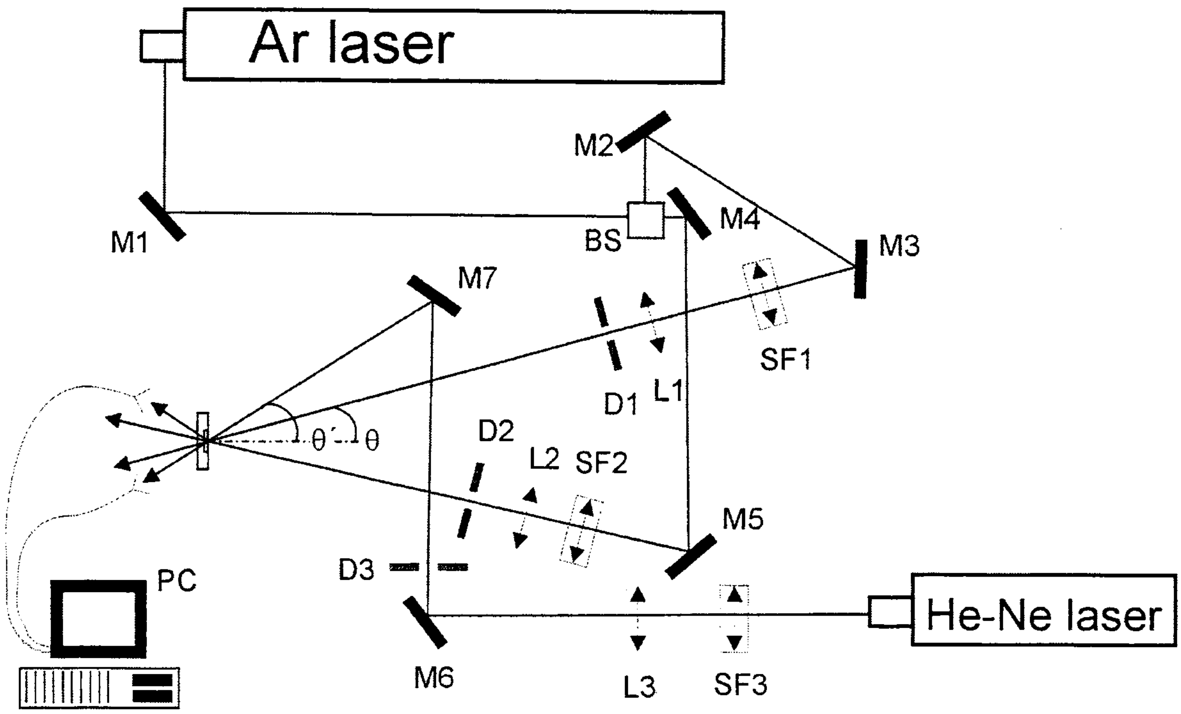

Fig 2. Experimental setup to record holograms using continuous laser (method 2): BS, beamsplitter, Mi, mirror, SFi, spatial filter, Li, lens, Di, diaphragm, PC, data recorder.

3. EXPERIMENTAL RESULTS

3.1 Comparison of two photochemical compositions with and without crosslinker using pulsed laser.

The importance of introduce the monomer crosslinker when the gratings was stored by pulsed laser can be seen in figure 3. Where the diffraction efficiencies versus energy of exposure, for layers around 60 µm of thickness, for polymer with crosslinker (circles) and without crosslinker (squares) have been plotted. This figure show the better behaviour of the material with crosslinker (type1), than the material without crosslinker (type 2). The energy need to store the maximum of the diffraction efficiency is similar in two cases around 250 mJ/cm^2, but with material type 1 the diffraction efficiency achieved was around 88 % and with material type 2 only was 60 %. After of this maximum of the diffraction efficiency the recording process presents a decrease of the index diffraction efficiency, this effect can bee seen in other works when the pulsed laser is used [4,10]. The slope of the curve of material using crosslinker monomer (type 1) is higher than the material without crosslinker (type 2). That different behaviour is the same effect observed using continuous exposure, the presence of crosslinker increase the polymerization rate and the refraction index of the polymer, this effects can be seen with details in reference [13]. The last aspect that can be analyzed is the comparison of the energy needed for store gratings with significant diffraction efficiency (5%). The gratings stored in material without crosslinker need around 80 mJ/cm^2 for achieve a significant diffraction efficiency, whereas the gratings stored in material with crosslinker achieve a diffraction efficiency of 5% with only 12 mJ/cm^2. This result confirm the idea studied in other works [12-13], that the presence of monomer crosslinker is the origin of the higher values of polymerization in the material.

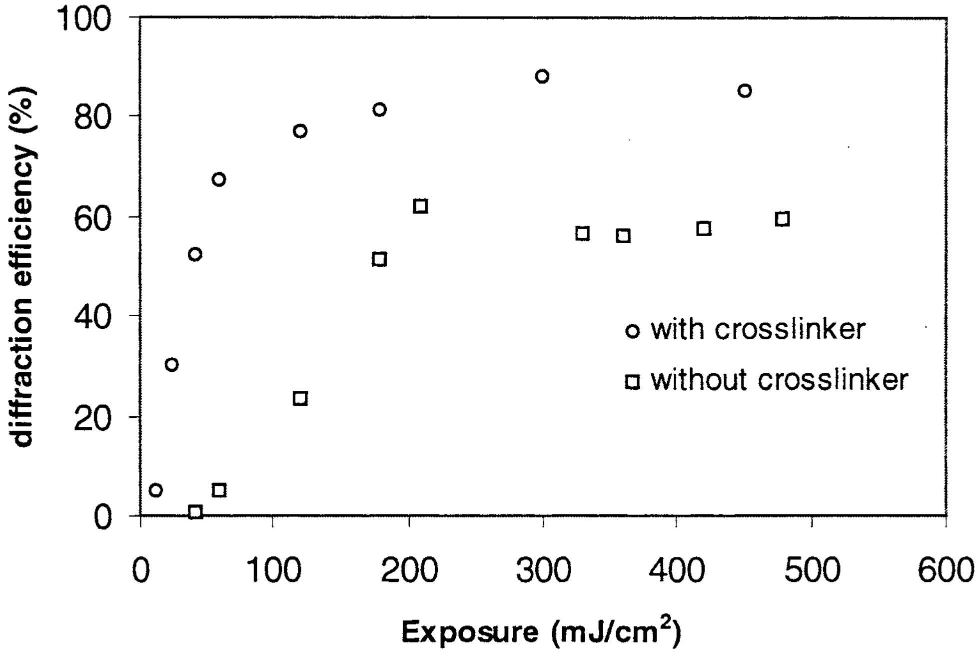

Fig 3. Diffraction efficiency as a function of the energy exposure for the material type 1with crosslinker and the material type 2 without crosslinker.

The thickness of the layers have obtained fitting the angular scan using Kogelnik's coupled wave theory [14]. By this fit the values of the index modulation and the coefficient of absorption and scattering can be also quantified. Figure 4 show the angular response of a grating type 1. The values obtained by the fitting were, the refraction index modulation (n_1), was 0.0045, the thickness was 60 μm and the coefficient of absorption and scattering (α) was 0.0012 μm^{-1}.

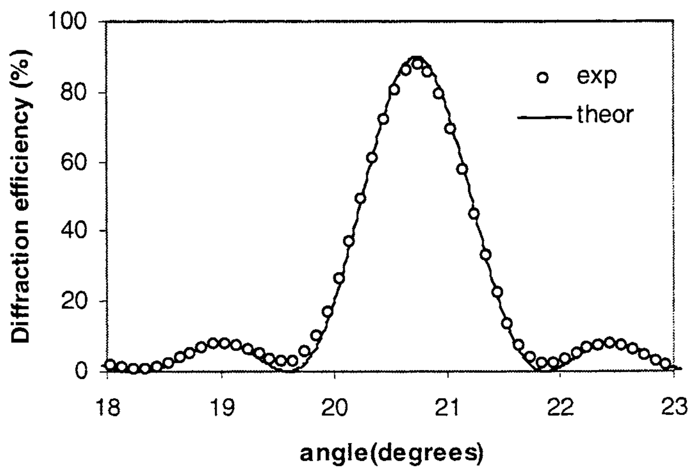

Fig 4. Diffraction efficiency versus the angle of incidence for a grating of material type 1 recorded using pulsed laser. The experimental point (circles) and the fitting by Kogelnik's theory (continuous lines).

3.2 Comparison of pulsed and continuous exposure.

The figure 5 represent de different behaviour of diffraction efficiency as function of exposure for the two recordings methods for two layers with similar composition (type 1) and thickness (around 60 µm). This figure shows a similar maximum diffraction efficiency but very different exposures. Only 70 mJ/cm^2 for continuous exposure and 250 mJ/cm^2 for pulsed exposure. The continuous laser was working with intensity of 5 mW/cm^2. In this figure the no linearity of the material's response is shown. The decrease of the diffraction efficiency after the maximum (around 93%) is typical effect observed in photopolymers when the refraction index modulation is high, this effect is called "overmodulation of refraction index modulation" predicted by Kogelnik couple wave theory and is usually observed in acrylamide based photopolymers [15]. For this reason is interesting analyze the behaviour of the index modulation stored in the material as function of the exposure energy.

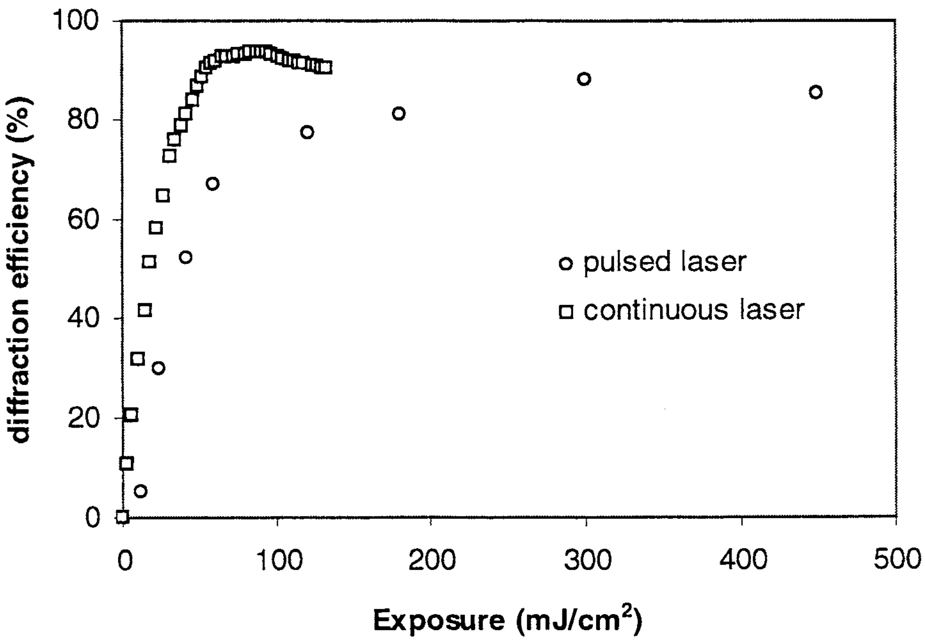

Fig 5. Diffraction efficiency versus exposure for the same material (type 1) for two different methods of recording: pulsed exposure (circles) and continuous exposure (squares).

In figure 6 the growing of the index modulation as a function of the exposure is shown for the material type 1 (with crosslinker) when is recorded by continuous laser (squares) and by pulsed laser (circles). With the continuous laser the index modulation achieved is 0.0058, 30% higher than the obtained by pulsed laser and the maximum diffraction efficiency is obtained faster (less than 70 mJ/cm^2). For this composition the index modulation of the gratings stabilizes for values around 0.006 [3]. Nevertheless a maximum diffraction efficiency can be achieve too. In the other hand the copy method is cheapest method to copy holograms, because the lenses, mirrors and spatial filters are necessary and the pulsed exposure is very interesting for analyze the different behaviour of material using the two types of exposure (pulsed and continuous) and understand the mechanism of hologram recording [4].

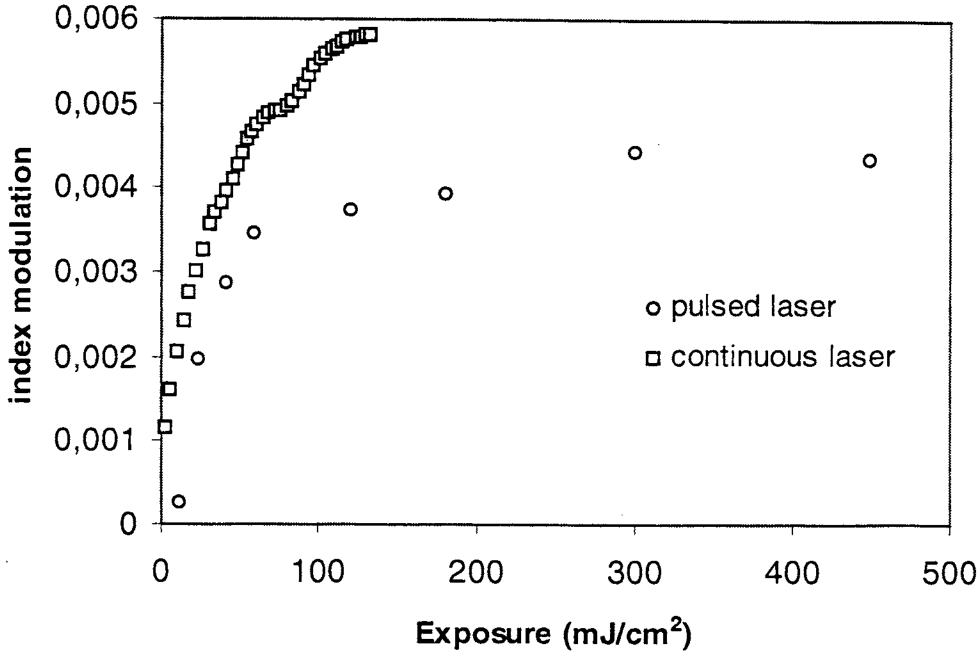

Fig 6. Refraction index modulation versus exposure for the same material (type 1) for two different methods of recoding: pulsed exposure (circles) and continuous exposure (squares).

4. CONCLUSION

In the first place the importance of the presence of monomer crosslinker to recorded holograms with pulsed laser have been studied in this work. The high diffraction efficiencies (87%) achieve with this composition using pulsed laser have been shown. The main advantages are the high index modulation achieved in the recorded gratings and the conservation of this type of gratings as can be seen in references 3 and 11. For this reason using the same thickness of the reference 4, the efficiencies achieved in this paper were 87 %, whereas with the composition of the reference 4 only the 60 % is achieved. Other important aspect that can be observed when the crosslinker is used to pulsed recording is the low energy to start the polymerization and achieve significant diffraction efficiency. This aspect can be used to have a better knowing of the phenomena that governs the formation of the gratings in photopolymers [8].

In the second place the comparison of the behaviour for continuous and pulsed exposure for the same photochemical composition (with crosslinker) have been done. The maximum diffraction efficiencies achieves are similar in two cases. The energetic sensitivity are better when the continues exposure is used and the values of index modulation stored are higher using continues laser exposure. Nevertheless the pulsed recording using copying process is an easy method to obtain industrial number of holograms using simple and cheap setup.

ACKNOWLEDGEMENTS

This work was partially financed by the "Oficina de Ciencia y Tecnología" (Generalitat Valenciana, Spain) under project nº GV01-130 and the CICYT ("Comisión Interministerial de Ciencia y Tecnología", Spain) under project nº MAT2000-1361-C04-04.

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