Comparison of peristrophic multiplexing and a combination of angular and peristrophic holographic multiplexing in a thick PVA/acrylamide photopolymer for data storage

Elena Fernández,^{1,3} Manuel Ortuño,² Sergi Gallego,² Celia García,¹ Augusto Beléndez,² and Immaculada Pascual^{1,*}

¹Dep. Interuniversitario de Óptica, Universidad de Alicante, Apartado 99, E-03080 Alicante, Spain

²Dep. Física, Ingeniería de Sistemas y Teoría de la Señal, Universidad de Alicante, Apartado 99, E-03080 Alicante, Spain

³E-mail: elena.fernandez@ua.es

*Corresponding author: pascual@ua.es

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Two different types of multiplexing are used to store 90 holograms at the same location in a polyvinyl alcoholacrylamide photopolymer material. In the first, the 90 holograms are stored using only peristrophic multiplexing, whereas in the second a combination of angular and peristrophic multiplexing is used. The results (diffraction efficiency and dynamic range, $M\#$) obtained with these two multiplexing techniques are compared. With the first, the dynamic range was $M\# = 13$ and with the second $M\# = 8$. An exposure schedule method is used to calculate the exposure time necessary to store the holograms with a more uniform, higher diffraction efficiency. © 2007 Optical Society of America

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1. Introduction

Due to the importance that new technologies (computers and Internet) are acquiring, there is an increased demand for more capacity, more density and faster readout rates in the new computers. Conventional memory technologies like CD-ROM's, with a capacity to store $0.7 \text{ bits}/\mu\text{m}^2$, and DVD's, with a capacity to store $4.5 \text{ bits}/\mu\text{m}^2$, have managed to keep pace with the demand for bigger, faster memories. But these memories are two-dimensional surface-storage technologies, and they have arrived at their limits of capacity.

Because of this, a new field of research has been opened up in three-dimensional holographic discs [1–5]. Research is focused on the characterisation of new holographic recording materials [6] where many holograms may be superimposed with new multiplexing schedules in order to store information with holographic techniques. The aim of new techniques is to enable the maximum number of $\text{bits}/\mu\text{m}^2$ to be stored.

Anderson et al. [7] managed to store $150 \text{ bits}/\mu\text{m}^2$ and Steckman et al. [8] $100 \text{ bits}/\mu\text{m}^2$. For this reason, different methods for multiplexing holograms, such as angular [4,9–12], peristrophic [1,9,13], or shift-multiplexing [8,14–15] are being made use of to store multiple holograms at the same location.

In this respect, photopolymers are considered interesting materials for recording holographic memories because they have excellent holographic characteristics, such as large dynamic range, $M\#$, of about 13 for a material thickness of $700 \mu\text{m}$ [4,6,9], good light sensitivity, real time image development, high optical quality and low cost. In addition, their properties like energetic sensitivity or spectral sensitivity can be easily changed by modifying the composition [6,10,16].

Dynamic range is the number of holograms with a diffraction efficiency of 100% which can be stored in a material with a specific thickness. It is the storage capacity of a holographic material and is characterized by the parameter $M\#$:

$$M\# = \sum_{i=1}^N \eta_i^{1/2}, \quad (1)$$

where η is the diffraction efficiency of each multiplexed hologram and i 'th is the number of holograms multiplexed.

With the dynamic range, it is possible to know how many holograms can be stored in the material with a specific diffraction efficiency, or what diffraction efficiency the holograms would have if a specific number of holograms were recorded in the material. It may be calculated from Eq. (2):

$$\eta_{AVR} = \left(\frac{M\#}{N} \right)^2, \quad (2)$$

where N is the number of stored holograms.

In order to fully exploit the dynamic range of the material used as many holograms as possible need to be stored. In this study, 90 holograms are stored at the same location using only peristrophic multiplexing or a combination of angular and peristrophic multiplexing [9]. Furthermore, we compare the results obtained when holograms are multiplexed with these two different types of multiplexing.

The material used to carry out this experiment is a photopolymer based on PVA-acrylamide [10,16]. Layers about $700 \pm 10 \mu\text{m}$ thick were made and when 90 holograms are stored, a dynamic range of between 8 and 13 is obtained in them.

To obtain the maximum dynamic range and a high uniform diffraction efficiency, an iterative method is used to determine the exposure schedule for multiplexing holograms [1,9,17]. In order to calculate the exposure times necessary for all the holograms to reach the same diffraction efficiency (uniform diffraction efficiency), this method makes use of the dynamic range, the number of holograms stored in the material, the diffraction efficiency of each of the stored holograms, and the exposure energy used to record them.

2. Experimental Setup

The photopolymer used to register the holograms is composed of acrylamide (AA) as the polymerizable monomer, triethanolamine (TEA) as radical generator, N,N' methylene-bis-acrylamide (BMA) as crosslinker, yellowish eosin (YE) as sensitizer and a binder of polyvinyl alcohol (PVA).

Table 1 shows the component concentrations of the photopolymer composition.

A solution of PVA in water forms the matrix and this is used to prepare the mixture of AA, BMA, and photopolymerization initiator system composed of TEA

Table 1. Concentrations of the Photopolymer Composition

	Composition
Polyvinylalcohol	13.50% w/v
Acrylamide	0.31M
Triethanolamine	0.12M
Yellowish eosin	$9 \cdot 10^{-5}\text{M}$
N,N' methylene-bis-acrylamide	0.04M

Fig. 1. Experimental setup: BS, beam splitter; Mi, mirror; Li, lens; Di, diaphragm; SFi, microscope objective lens and pinhole; Ri, radiometers.

and YE. The PVA is supplied by Fluka, AA, TEA and BMA by Sigma, and YE by Panreac. The mixture is made under red light, deposited by gravity in circular polystyrene molds and left in the dark for six days to allow the water to evaporate in conditions of temperature, T , between 20°C and 25°C , and relative humidity, RH , 40%–60%. These conditions of drying time, temperature and relative humidity are optimized to obtain the maximum diffraction efficiency in $700 \mu\text{m}$ thick plates. Once dry, they are removed from the mold and placed on a $5.5 \times 5.5 \text{ cm}$ glass support to be used for recording.

Holographic gratings were recorded using the output from a diode-pumped frequency-doubled Nd:YVO_4 laser (Coherent Verdi V2) which was split into two beams and then spatially filtered, using a microscope objective lens and a pinhole, and collimated to yield a plane-wave source of light at 532 nm. The diameters of these beams are 1.5 cm and the intensity 5 mW/cm^2 with an intensity ratio of 1:1. The Gaussian light beams were spatially overlapped at the recording medium intersection at an angle of 17.2° (measured in air), resulting in an interference grating with a spatial frequency of 1125 lines/mm. The diffracted intensity is monitored in real time with the He-Ne laser positioned at Bragg's angle ($\theta' = 20.6^\circ$). In order to obtain the diffraction efficiency as a function of the angle at reconstruction, the plates are placed on a rotating stage. The diffraction efficiency was calculated as the ratio of the diffracted beam to the incident power (Fig. 1).

3. Results and Discussion

As mentioned in the introduction, 90 holograms are stored at the same location in the material using two different types of multiplexing. In the first, the 90 holograms are stored using only peristrophic multiplexing, whereas in the second the holograms are stored using a combination of angular and peristrophic multiplexing.

In both cases a method is applied to calculate the exposure times to be used to store the holograms. In order to apply this method an initial iteration is made,

from which the number of holograms stored in the material, dynamic range, and diffraction efficiencies are obtained. These data are then used to calculate the exposure times necessary to store the holograms in subsequent iterations so as to obtain uniform diffraction efficiencies [1].

A. Peristrophic Multiplexing

Firstly, 90 holograms are stored using only peristrophic multiplexing, with an angular separation of 2° . In earlier studies it was determined that the angular selectivity for this material with this thickness is 0.5° [9]. Therefore, an angular separation of 2° is more than sufficient to prevent the holograms from overlapping.

For the first hologram to be formed, a specific exposure must be used, because below this exposure the material does not respond. For this reason, the first hologram is stored with an exposure time of 2 seconds. In order to store the other holograms different configurations were tested previously. First, we stored all the holograms with the same exposure time, but the first holograms had high diffraction efficiencies and the last holograms had diffraction efficiencies close to 0%. When the holograms are stored, the monomer and the dye are being consumed and therefore the material is less sensitive. For this reason it is necessary to increase the exposure time for the last holograms so that they reach the same diffraction efficiency as the first holograms. And instead of storing all the holograms with the same exposure time, we decided to increase this time as more holograms were stored. These times that are used to store initially the holograms and that are chosen based on the material are called “initial iteration times” in our study.

Then, the exposure time used to store the holograms at the initial iteration is as follows: 2 s for the first hologram (time necessary for the material to respond), 0.5 s for hologram 2 to hologram 6, and then 0.5 s is added for every five holograms stored. Figure 2 shows exposure times of the initial iteration versus hologram number.

Once the holograms with the exposure times of the initial iteration have been stored, the diffraction

Fig. 2. Exposure times of initial iteration versus hologram number for peristrophic multiplexing and for angular and peristrophic multiplexing.

Fig. 3. Diffraction efficiency versus hologram number after initial iteration for peristrophic multiplexing only.

tion efficiency of each one of them is measured. Figure 3 shows diffraction efficiency versus number of holograms obtained when the holograms are recorded with the above exposure times. Ninety holograms are stored with a mean diffraction efficiency of 2.9%. As can be seen, the diffraction efficiency of the first forty holograms (between 8% and 4%) is much higher than the diffraction efficiency of the last holograms (around 0.02%). Therefore, for the holograms to be stored with uniform diffraction efficiency (all the holograms with the same diffraction efficiency), it is necessary to decrease the exposure times for the first holograms and increase the exposure times for the last ones. An exposure schedule method is used to calculate these exposure times [1]. These exposure times that we calculated are called “exposure times of first iteration”.

With the diffraction efficiencies from Fig. 3 we calculated the cumulative grating strength, $\sum_{i=1}^N \eta_i^{1/2}$, where η is the diffraction efficiency and N the number of holograms stored so far, and represented as a function of exposure energy in Fig. 4. When the curve is saturated, we can obtain the dynamic range, which in this case is $M\# = 13.5$.

Then, in order to store the 90 holograms with uniform diffraction efficiency, first the data obtained from Fig. 4 are fitted in the following theoretical Eq. (3):

$$A = a_0 + a_1 E + a_2 E^2 + a_3 E^3 + a_4 E^4 + a_5 E^5 + a_6 E^6, \quad (3)$$

Fig. 4. Cumulative grating strength as a function of exposure energy.

Fig. 5. Exposure times of first iteration versus hologram number. White circles represent the times for peristrophic multiplexing and black circles the times for angular and peristrophic multiplexing.

where A is the cumulative grating strength and E the exposure energy. Once the coefficients a_i have been calculated, the time needed to record the holograms may be calculated from Eq. (4):

$$t_n = A_{\text{sat}} / N \cdot I \left[a_1 + 2a_2 \sum_{i=1}^{n-1} E_i + 3a_3 \left(\sum_{i=1}^{n-1} E_i \right)^2 + 4a_4 \left(\sum_{i=1}^{n-1} E_i \right)^3 + 5a_5 \left(\sum_{i=1}^{n-1} E_i \right)^4 + 6a_6 \left(\sum_{i=1}^{n-1} E_i \right)^5 \right], \quad (4)$$

where A_{sat} is the dynamic range obtained, N is the number of holograms to be stored, I is the recording intensity, and E_i the energy used to record up to the i 'th hologram.

Figure 5 with white pixels shows the exposure times of the first iteration for peristrophic multiplexing. These times are obtained with the fitted values of Fig. 4 to Eq. (3) and with Eq. (4).

With these exposure times (Fig. 5, white circles), 90 holograms are stored again with peristrophic multiplexing. Figure 6 shows the diffraction efficiency of the 90 stored holograms versus the hologram number after the initial iteration (white circles) and after the first iteration (black circles). After the first iteration, the dynamic range obtained is $M\# = 12$, and the mean diffraction efficiency is 2.1%. The diffraction efficiency and dynamic range have diminished compared with the values for the initial iteration (white circles in Fig. 6). But as we can see, the diffraction

Fig. 6. Diffraction efficiency versus hologram number for the initial iteration (white circles) and for the first iteration (black circles) for peristrophic multiplexing only.

Fig. 7. Diffraction efficiency versus hologram number after initial iteration for a combination of peristrophic and angular multiplexing.

efficiencies are much more uniform and closer to the mean diffraction efficiency.

B. Combination of Angular and Peristrophic Multiplexing

Secondly, 90 holograms are now stored using a combination of angular and peristrophic multiplexing. For peristrophic multiplexing an angular separation of 5° and 18 peristrophic positions from 0° to 90° are used. At each peristrophic position 5 holograms are stored using angular multiplexing with an angular separation of 0.5° .

The exposure times used to store the holograms in the initial iteration are the same as in the case shown in Fig. 2, 2 s for the first hologram, 0.5 s for hologram 2 to hologram 6, and then 0.5 s is added for every five holograms stored.

The diffraction efficiencies obtained are shown in Fig. 7. As in the case of Fig. 3, the first holograms have a higher diffraction efficiency (around 2.5%) than the last ones (around 0.02%). Figure 8 shows the cumulative grating strength as a function of exposure energy. The dynamic range obtained in this case is $M\# = 7$.

The data in Fig. 8, together with Eqs. (3) and (4), are used to calculate the exposure times of the first iteration necessary to store the holograms so that the diffraction efficiencies in Fig. 7 are more uniform. This exposure times are shown in Fig. 5 with black circles. As can be seen, the exposure time for the combination of angular and peristrophic multiplex-

Fig. 8. Cumulative grating strength as a function of exposure energy.

Fig. 9. Diffraction efficiency versus hologram number for the initial iteration (white circles) and for the first iteration (black circles) for a combination of peristrophic and angular multiplexing.

ing is slightly greater than for peristrophic multiplexing in the case of holograms 1 to 65. For the rest of the holograms (66 to 90) the exposure time for peristrophic multiplexing is greater.

Diffraction efficiencies obtained after the first iteration cannot be interpreted with the exposure times because the holograms stored with peristrophic multiplexing have diffraction efficiencies greater than those stored with angular and peristrophic multiplexing even though they were stored with shorter exposure times.

With these exposure times (Fig. 5, black circles), 90 holograms are stored once again and the diffraction efficiencies obtained as a function of the hologram number are represented by black circles in Fig. 9.

In order to make it easier to compare the results obtained in Fig. 7 with those, in Fig. 9 diffraction efficiencies obtained after the initial iteration are represented by white circles, and those obtained after the first iteration by black circles. In this case, from the black circles it can be seen that, with the exception of the last 10 holograms that have a diffraction efficiency of around 0.05%, all the holograms have a diffraction efficiency of between 1.5% and 0.5%, which is close to the mean diffraction efficiency of 1%. The dynamic range is $M\# = 8$. In other words, the diffraction efficiencies of the holograms are now more uniform than the values obtained at the initial iteration. The last holograms did not reach a higher diffraction efficiency because the dynamic range has been consumed and there is not sufficient monomer left in the material to make more holograms.

C. Comparison of Peristrophic Multiplexing and a Combination of Angular and Peristrophic Multiplexing

We shall now compare the results of both iterations for the two types of multiplexing used. It should be pointed out that in both cases the diffraction efficiencies were obtained with the same exposure times in the initial iteration. Moreover, the composition of the material in both cases was the same (Table 1), the solutions were deposited at the same time and allowed to dry for the same period of time under the same conditions of temperature and humidity. The only difference between the two was the way in which

the holograms were stored, and this gave rise to different results.

In Figs. 6 and 9, the diffraction efficiencies obtained at the initial and first iteration are compared.

We shall now compare the diffraction efficiencies at the initial iteration in Figs. 6 and 9 and the diffraction efficiencies at the first iteration in the same two figures. For the initial iteration in Fig. 6, it can be seen that the first 30 holograms have a diffraction efficiency higher than 4%, holograms 30 to 50 a diffraction efficiency between 4% and 2%, holograms 50 to 70 between 2% and 1%, and holograms after number 70 have a diffraction efficiency below 1%.

Let us now consider the initial iteration in Fig. 9. Only two holograms have a diffraction efficiency above 4%. In Fig. 6 the first 50 holograms have a diffraction efficiency above 2%, whereas in Fig. 9 only the first 20 holograms reach this value. Between holograms 20 and 40, the diffraction efficiency ranges from 2% to 1%, and the remaining holograms have a diffraction efficiency below 1%. As can be seen, the diffraction efficiencies for the initial iteration in Fig. 9 are lower than those in Fig. 6.

Now we shall consider the diffraction efficiencies for the first iteration in Figs. 6 and 9. Whereas in Fig. 6 the first 70 holograms have a diffraction efficiency above 2%, in Fig. 9 these holograms have a diffraction efficiency of around 1%, and only hologram 1 has a diffraction efficiency of above 2%.

Therefore, from these two figures it can be concluded that with peristrophic multiplexing alone (Fig. 6) higher diffraction efficiencies are obtained than with the combination of angular and peristrophic multiplexing (Fig. 9).

When the gratings are stored with peristrophic multiplexing, the angle with which object and reference beams overlap in the material is the same. Therefore, the beams enter symmetrically with respect to the material. In this case, the interference planes of the grating are perpendicular to the film.

However, when the gratings are stored with angular multiplexing, the angles with which object and reference beams overlap in the material are not equal; therefore, in this case there is an asymmetric geometry and the interference planes are not perpendicular to the material, they are slanted. In the asymmetric case, since the planes of the grating are slanted, the diffraction efficiency is smaller than in the symmetrical case [18]. Therefore, the reason why the slanted gratings have a smaller diffraction efficiency is due to a physical process, not a chemical one.

However, although higher diffraction efficiencies are obtained with peristrophic multiplexing on its own, due to the angular selectivity of the material it is not possible to store more than 200 holograms without overlapping. On the other hand, a combination of two or more types of multiplexing enables a greater number of holograms to be stored. For the material to be used as a holographic memory and the greatest possible number of bits stored in it, over 500 holograms would need to be stored. Hence, two or more types of multiplexing must be combined to in-

crease the number of holograms that can be stored without overlapping.

4. Conclusion

In this study, 90 holograms were stored in a PVAacrylamide photopolymer using two different types of multiplexing—peristrophic multiplexing alone, and a combination of angular and peristrophic multiplexing. An exposure schedule method was applied in both cases to calculate the exposure times necessary to obtain uniform diffraction efficiencies. In the first case, a dynamic range of $M\# = 12$ was obtained with a mean diffraction efficiency of 2.1%. In the second case, the dynamic range was $M\# = 8$ and the mean diffraction efficiency 1%. As can be seen, with the combination of angular and peristrophic multiplexing the mean diffraction efficiency and dynamic range are lower than with peristrophic multiplexing alone. However, with a combination of the two types of multiplexing a greater number of holograms may be stored without overlapping. Therefore, whether it is best to use peristrophic multiplexing alone or a combination of the two types of multiplexing depends on the number of holograms to be stored.

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