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Hologram multiplexing in a highly photosensitive photopolymerizable material in a sol-gel matrix

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ABSTRACT In this paper, we present a photopolymerizable material in a sol-gel matrix suitable for recording a large number of multiplexed diffraction gratings with low total exposure energy. The matrix of this material is an inorganic porous silica glass, a material that does not shrink when radical photopolymerization is initiated. The photosensitive component is based on acrylamide, *N*,*N*′-methylenebisacrylamide, triethanolamine and yellow eosine as a photoinitiator. In the reported study, 50 holograms were angularly multiplexed with diffraction efficiencies between 0.1 and 1.9% resulting in a dynamic range of M/# = 3.9. The total exposure was 4.5 mJ/cm^2 and the energy used in recording each hologram was 95 μJ/cm^2. This indicates a very high sensitivity for this material in the range of 3.3 to 15.5 cm/mJ. Due to this good holographic performance, the material is suitable for holographic data storage applications.

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Photopolymers have attracted great interest as holographic recording media because they have several interesting properties such as high sensitivity, high resolution, real-time dry processing and low cost. These properties are very important for practical applications, especially in the field of holographic data storage. These materials exhibit, however, substantial dimensional changes during recording or when the temperature changes and are available only as thin films, which limits the number of holograms that can be multiplexed in one location. Changes in dimension can lead to rotations of the Bragg angles of the recorded holograms which degrade the fidelity when the holograms are reconstructed and finally limit the density of data that the material can support [1–3].

In recent years, the sol-gel approach has emerged as a promising way of preparing and manufacturing holographic recording media. Several groups have reported hybrid sol-gel materials, but they present problems of dimensional change, low dynamic range, scatter light from inhomogeneities and mainly low energetic sensitivity [4]. In this letter we report results for an inorganic silica glass which represents an advance in the use of this class of materials as holographic recording media. We illustrate the capacity of sol-gel based material for recording multiplexed holograms that exhibit stable angular selectivity with low exposure energy and short exposure time.

Holograms of low diffraction efficiency, such as $\eta = 0.1\%$, are of particular interest for holographic data storage, since it is usually desirable to multiplex as many holograms as possible at one location to maximize the storage density and hence the capacity. In photopolymerizable materials it is generally difficult to record holograms of low diffraction efficiency that exhibit stable angular selectivity [5]. In this letter, we demonstrate the capacity of a photopolymerizable filler in a porous silica glass to record multiplexed holograms that exhibit stable angular selectivity of a composition that is very different from those previously published [6]. Another important property of this sol-gel-based material is the low background noise in the diffracted order, an effect that has been reported in both classical photopolymers and sol-gel materials as a result of the scattering coming from inhomogeneities in the recording material (noise gratings). The very small diffracted intensity between Bragg peaks illustrates the high homogeneity obtained in this sol-gel material.

The method of making this material is based on the bulk sol-gel method developed by Wang et al. [7]. The porous silica glass was prepared using a typical sol-gel technique, which proceeds through the steps of hydrolisis and polycondensation of tetraethyl orthosilicate (TEOS) in an alcohol solution containing an acid catalyst, molding and gelation of the sol, followed by aging and drying of the gel under normal laboratory conditions (20° C and 60% RH), resulting in a material with a thickness of the order of 1 mm. Taking advantage of the porosity of this kind of material, we photosensitized the matrix by introducing the silica glass into a photosensitive solution. The composition of this photosensitive solution was, acrylamide (7 g), *N*,*N*′-methylenebisacrylamide (0.3 g), triethanolamine (4 ml), yellowish eosine (3×10^{-3} g) and 2-propanol

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(30 ml) as solvent. These components were used without any purification and were purchased from Aldrich.

Holographic gratings were recorded using the output from a diode-pumped, frequency-doubled Nd:YAG laser (Coherent Verdi V5) which was spatially filtered, using a microscope objective lens and a pinhole, and collimated to yield a plane-wave source of light at 532 nm. The Gaussian light beam with a diameter of 1 cm was then split into two beams. The two beams were spatially overlapped at the recording medium, the power of the light in the two beams was 10 and 9 mW/cm^2, respectively, and the two beams intersected at an angle of 32° (measured in air) which resulted in a interference grating with a spatial frequency of 1100 lines/mm. The sample was mounted on a motorized rotation stage and the sample was rotated by a computer controlled DC motor around an axis that was perpendicular to the plane defined by the two incident laser beams and is located at the intersection of the beam. The exposure time was 0.5 ms and it was controlled using a shutter. Once all the holograms were recorded, angular response measurements were made using a probe beam from the He-Ne laser, at a wavelength of 632.8 nm where the photopolymer does not absorb.

Fifty holograms were angularly multiplexed by incrementing the sample angle (θ_s) in steps of 0.7° from −17° to 17°, where the sample angle is the angle between the normal to the sample and the bisector of the angle between the recording beams. Figure 1 shows the diffraction efficiencies of a set of 50 angle-multiplexed gratings as a function of the sample angle. As can be seen, the diffraction efficiency η varies between 0.1 and 1.9%, showing the ability of this photopolymerizable material in a silica glass matrix to support multiple volume holograms. Figure 1, shows also the low level of noise that is observed in the diffracted intensity, which is between 0.05% for the first hologram recorded and 0.2% for the last hologram recorded (lower than the values reported in our previous paper [6]). Considering the fact that photopolymers in a porous silica glass typically suffer from high values of scattering [4], especially if the silica matrix is inorganic, these are promising results.

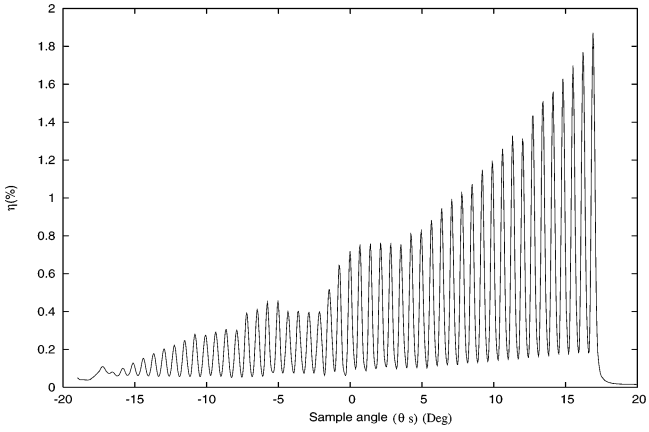

FIGURE 1 Angular scan of the diffraction efficiencies of 50 angle-multiplexed volume holograms. The holograms were recorded with an angular separation of 0.7° from a sample angle between −17 to 17

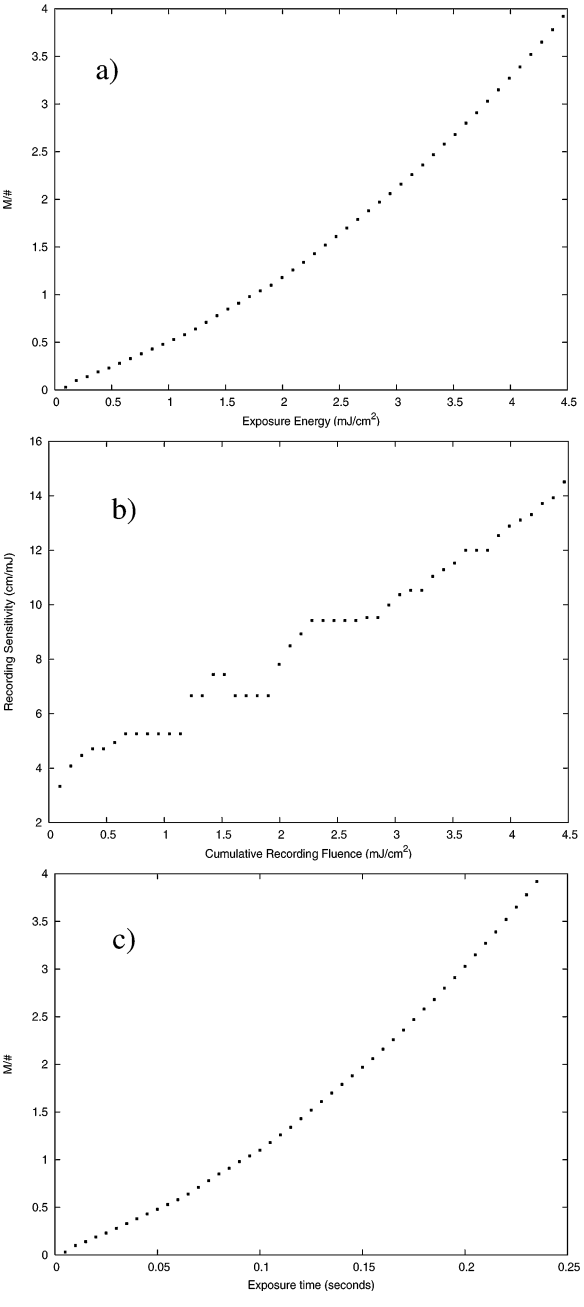

FIGURE 2 **a** Cumulative M/# as a function of total exposure energy. **b** Recording sensitivity in cm/mJ versus cumulative recording fluence in mJ/cm^2 **c** Cumulative M/# as a function of total exposure time. M/# is the sum of the square root of the diffraction efficiencies of the holograms

To record these holographic gratings, the same energy was used for each hologram, obtaining a linear increase of η with increasing number of holograms multiplexed at that location. The energy used to record each hologram was 95 μJ/cm^2 and the total energetic exposure to record all the gratings was 4.5 mJ/cm^2. Figure 2a shows the cumulative dynamic range M/# as a function of total exposure energy where M/# is calculated according to Eq. 1 [1]. As can be seen, a high dynamic range can be obtained with low exposure energies for this material. Figure 2b shows the recording sensitivity (**S**) as a function of the cumulative exposure **S** as defined by Eq. 2 [8]:

$$\mathrm{M/\#} = \sum_{\mathrm{i}=1}^{\mathrm{M}} \sqrt{\eta_{\mathrm{i\,max}}} \tag{1}$$

$$\mathbf{S} = \frac{\sqrt{\eta_{\mathrm{i\,max}}}}{I_{\mathrm{i}} t_{\mathrm{i}} L} \tag{2}$$

where L is the thickness of the recording material (1 mm), t_{i} is the exposure time for the ith event, I_{i} is the intensity for the ith recorded grating and $\eta_{\mathrm{i\,max}}$ is the maximum diffraction efficiency of the ith grating.

The values of sensitivity of the silica glass are between 3.3 for the first recorded grating and 14.5 cm/mJ for the last one, and these values are similar to those published for photopolymerizable materials without the sol-gel matrix material [5, 9]. It is noteworthy that the sensitivity increases in monotonic fashion throughout the recording. This behavior is contrary to the evolution of the sensitivity in photopolymerizable recording material without sol-gel matrix, where it decreases in monotonic fashion [8].

Table 1 shows the sensitivity obtained in the most relevant work reported to date based on either classical photopolymers, photopolymer-filled nanoporous glasses and hybrid sol-gel material. As shown in the table, the silica glass presents a higher sensitivity than the nanoporous materials [4, 10], a sensitivity similar to the commercially available ULSH-500 photopolymer [5] and slightly lower sensitivity than the acrylamide and blue methylene in a polyvinyl alcohol matrix [9]; however, in this last case, the thickness of this material (< 100 μm) and the number of holograms that can be multiplexed in one location are low compared to that the other systems.

Material	S (cm/mJ)
Our Silica glass	(3.3–14.5)
Hybrid sol-gel [4]	0.1
Vycor glass [10]	4.2×10^{-3}
Polyvinyl alcohol matrix [9]	48
ULSH-500 photopolymer [5]	1–10

TABLE 1 Sensitivity normalized to thickness of the recording material, for the most significant materials

On the other hand, Fig. 2c shows the cumulative M/# as a function of total exposure time. The working intensity was 19 mW/cm^2, and this value was used throughout the study. The exposure time is 5 ms for an energy of 95 μ J/cm^2 per hologram at the working intensity of 19 mW/cm^2. These results are very important if the goal for the material is to be used in holographic data storage, because lasers with a power of the order of tens of mW are expected to be used in holographic data storage applications and exposure times of milliseconds are desirable to obtain data rates that are of technical interest. If this sol-gel material is compared to other similar nanoporous glasses, such as those shown in Table 1, it can be seen that this sol-gel material presents a slightly higher dynamic range, a clearly shorter exposure time and lower exposure energy, and a slightly better angular selectivity.

In conclusion, this study reports a material with optimal properties for use in holographic data recording. This material does not exhibit shrinkage, which photopolymerisation typically is associated with, has low thermal expansion, high dynamic range and high sensitivity allowing short exposure times at conveniently low intensity of the recording light. We have also demonstrated the capacity of this material to record gratings with low diffraction efficiency but high angular selectivity.

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