

## Special Multi-Part Issue on: Holographic Materials for Data Storage

Guest Editor **Dr. John Sheridan**University College, Dublin

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# HOLOGRAPHY

## Polarization-holographic data storage

We are developing holographic data-storage technology based on polarization holography in thinfilm photo-anisotropic polymer storage materials. Unlike in traditional holography, where the interfering waves have the same polarization, in polarization holography object and reference waves have mutually orthogonal polarization (e.g. left and right circular in our case). These waves do not create interference fringes in the sense of spatial intensity variation, but they give rise to a constant intensity field with spatially variable polarization (Figure 1). These 'polarization fringes' can be stored in a material that presents optically-induced anisotropy under the effect of the local electric field. When the hologram is illuminated by the reference beam (of the same circular polarization) the object wave is reconstructed. Polarization holography has the advantage of high diffraction efficiency even for thin storage layers, and the lack of all diffracted orders except the 0th and the +1st. This means that high-luminosity holographic images can be reconstructed by Gabor type (in line) holography without any noise arising from unwanted diffraction orders.

We use azobenzene side-chain polymers as our polarization holographic storage material.<sup>1,2</sup> In azobenzene polyesters, irradiation with polarized light induces optical anisotropy<sup>3</sup> by changing the orientational distribution of the azobenzene side chains. The mechanism starts with trans-cis and cis-trans isomerizations induced by the absorbed photons. This results in more molecules with transition dipole moment perpendicular to the electric field vector than parallel to it, thus creating birefringence following the spatial variation of the lo-

cal electric-field vector. Photo-anisotropy induced in a thin film of some polyesters is found to be stable up to a temperature of 160°C: the material also has very good optical quality and a high diffraction efficiency. Rewritable storage is possible because the microscopic orientation of the storage material is reversible.

We have developed two types of holographic memory card demonstrator systems. The first system we call the read&write unit. It uses a green laser (532nm) and is able to record and reconstruct holograms on our holographic storage material embedded into a standard credit-card-sized carrier. As the storage material is only sensitive in the bluegreen region, non-destructive readout can be realized using a reconstructing beam that has the same polarization and wavefront but different wavelength (red). This is the basis of the read-only demonstrator that has been built in a second step. Our systems operate in reflection mode, allowing writing and reading to be accomplished from the same side of the card with a small optical head.

A typical reconstructed image is shown on Figure 2. We use a coding method called constant-weight sparse-modulation encoding<sup>4</sup> that presents some loss in data density but has proved to be tolerant to noise and illumination inhomogeneity. With this code we achieved a raw symbol error rate of 10<sup>-4</sup> that is further improved by the standard RS code to a user bit-error rate of 10<sup>-12</sup>. With proper adjustment of the read/write demonstrator optical system we achieved a raw data density as high as 2.77bit/μm<sup>2</sup>.

Page-organized holographic data storage in itself

Continues on page 10.

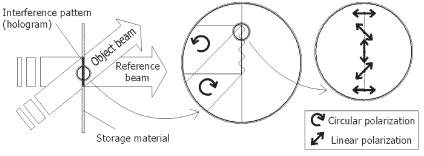


Figure 1. Schematic view of polarization-holographic recording.

## **Editorial**

What must be acknowledged at the very start of this special double issue dealing with holographic materials is that we live in very exciting times. It is probable that within 1-2 years of reading this you will buy a holographic-disk data-storage system, HDS, for research purposes. It is also likely that within 10 years many families will own a home HDS system.

Why so much emphasis on holographic data storage? And surely I am exaggerating, after all the data storage potential of holography has been known for years? Several things have recently happened, or are currently happening, in this area that will lead to major advances.

On the commercial side, after years of work, several companies are currently selling data-storage-quality materials. One company, InPhase Technologies, has demonstrated a working photopolymer-based WORM (write-once/read-many) system, which will be shipped next year. This is a big step forward, both in terms of engineering technology and in terms of psychology. We are about to get a system against which all research claims and future

industrial prototypes must be compared. Holography may be about to become an integral PC and data storage technology, equivalent to CD or magnetic tape. The potential markets and the possibility of owning a device standard explain the very-real current commercial interest.

On the research, side there continues to be steady progress. Importantly, as this multi-part special issue demonstrates, an ever-increasing number of organic and inorganic materialswith a variety of characteristics—are being examined. Several innovative optical read/write systems are being explored and a technology road map is gradually emerging. With a working WORM system available, the impetus to produce commercial rewritable holographic media will increase. Furthermore, researchers in laboratories around the world are developing the industrial design tools necessary to proceed. Sophisticated electromagnetic models exist describing holographic replay. These are being combined with increasingly complex models describing material performance and

linking it to chemical composition.

This work is of great significance to the general optics research community in that this is an enabling technology. The possibility that materials and desktop systems, which are capable of fabricating a patchwork of complex holographic elements, has major implications for optical signal processing. These implications may be as significant as those associated with the development of the printed circuit board

I hope to justify the need for such large data storage capacity and the implication of using holographic media in the next issue. For now I thank the many contributors and I hope you have as much fun reading this special issue as I had putting it together. I thank Ray Kostuk and Sunny Bains for their thoughtfulness and assistance.

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## Secure holographic memory system with multi-dimensional keys in photorefractive crystal

Continued from page 12.

degrees of freedom in the spatial coordinates.

We present experimental results of a secure holographic memory by use of double random-phase encryption. We use a 10×10×10mm3 LiNbO3 crystal doped with 0.03 mol% Fe. The Fourier transform of the encrypted data is stored holographically in the 90° geometry. The crystal was mounted on a rotary stage for angular multiplexing. The c-axis is on the paper and is at 45° with respect to the crystal faces. A light beam emitted from an Ar<sup>+</sup> laser at a wavelength of 514.5nm was divided into object, reference, and readout beams. The readout is the conjugate of the reference. Four binary images were recorded after the encryption. Figures 2(a) and (b) show

an example of the original binary data and its encrypted image. Figure 2(c) and (d) show the reconstructed images obtained by using correct and incorrect phase keys, respectively: stored images were reconstructed successfully using the correct key only. After binarization, we confirmed that reconstruction is error-free in the four reconstructed images. The average biterror rate with the incorrect keys was 0.502. This value shows the random search.

Photorefractive materials are attractive for rewritable holographic media, however the erasure during readout process is one of impor-

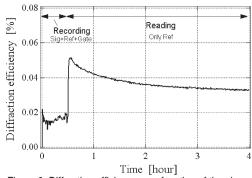


Figure 3. Diffraction efficiency as a function of time in Ru, Pr:SBN. Sig, Ref, and Gate denote He-Ne signal and reference beams, and the Art gate beam, respectively.

tant problems. LiNbO<sub>3</sub> and  $Sr_xBa_{1.x}Nb_2O_6$  are promising materials for volume holographic media.<sup>7</sup> Buse *et al.* presented one solution for the nonvolatile storage in a LiNbO<sub>3</sub> crystal doped with Fe and Mn.<sup>8</sup> We have been investigating  $Sr_xBa_{1.x}Nb_2O_6$ -doped with Ru and Pr for a high-speed holographic recording. Figure 3 shows a temporal behavior of the holographic grating. We can see that the stored information is retained throughout the readout process. This preliminary result encourages us to develop nonvolatile photorefractive media with high speed and a large diffraction efficiency.

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## Photo-anisotropic response in dyed polymeric matrices

Dye sensitization of photopolymer films has long been used to increase absorbance over a limited spectral range to improve microstructure formation through illumination with a specific light source. Recently, characterization of optically-induced mechanisms in dyed polymeric films has been a growing field. These mechnisms have extensive applications: they play a role in photorefractive performance, <sup>1,2</sup> laser emission, <sup>3</sup> data storage, <sup>4,5</sup> information processing <sup>6</sup> and display applications. <sup>6</sup> Gelatine, polyvinyl alcohol, polymethylmethacrylate, acrilamide and polyesters are just some of the materials used frequently as matrices to insert dye molecules.

Our research aims to study characteristics and recording effects when intensity and polarization patterns induce a periodic index change in gelatines and polyvinyl alcohols of differing molecular weights. These patterns are obtained by varying the polarization angles shown in Figure 1. To sensitize the films, two different dyes from the tiazine and triphenilmetane groups are used. In the case of PVA materials,7 the characteristics of the polymer determine the recording efficiency when a specific chromophore is introduced in the matrix. In addition, we have developed a theoretical analysis of the two possible mechanisms in our materials: induced and form birefringence.

The studied materials show photo-induced birefringence in the exposed (bleached) region due to photo-anisotropy: i.e. change in the characteristics of the photosensitive medium-either the refraction index or transmission coefficient—when irradiated with polarized light. It is possible that the dye molecules within a gelatine matrix react to the electric field, allowing the formation of polarization gratings. Based on the measured diffraction efficiency, a theoretical model has been developed to find the photoanisotropy coefficient when the angle between the incident beams is small, when the grating period is greater than the illumination wavelength. In this case it is observed that the intensity of the -1 diffracted beam is a sinusoidal function of the incident polarization angle,8 see Figure 2.

When the grating period is smaller than the illumination wavelength, it is evident from our results that form birefringence (the result of a high spatial frequency in a stratified medium) becomes significant, thus modifying the optically-induced birefringence. We did a theoretical analysis to alter form birefringence, oconsidering the propagation of a plane electromagnetic wave through a grating using modal

theory. We established that the modes that propagate through the structure are the diffraction orders. The eigenproblem is solved as a function of the ratio of the illumination wavelength to grating period. When this ratio is much greater than one (quasistatic limit), the grating shows a response similar to a uniaxial film. With a first order approximation, the modal theory is reduced to a first order theory, as this is the only propagation mode. It is possible to approximate the eigenfunction in order to calculate the effective refractive indices, ordinary and extraordinary, of the equivalent birefringent medium. In this way, an element that varies polarization can be designed. One of the advantages of such elements is that their spectral responses can be made independent of wavelength.

It has been also shown<sup>10</sup> that, in spite of its high viscosity, a relief grating is formed when orthogonal linear polarized beams are superimposed upon the plates. In this case, the frequency of the gelatine relief gratings does not depend on the direction of the electric field of the orthogonal recording beams. When orthogonal, circularly-polarized beams are used to form polarization gratings, no relief grating is produced.

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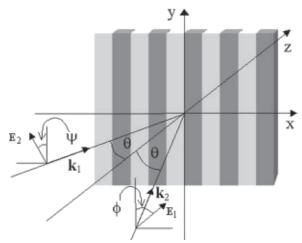


Figure 1. Schematic representation of two linearlly polarized beams of light  $\mathbf{E}_1$  and  $\mathbf{E}_2$  with a polarization angle  $\psi_1$  and  $\psi_2$ . The incidence angles  $\theta_1$  and  $\theta_2$  are with respect with the normal of the plane of incidence.

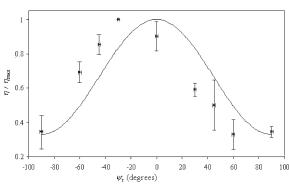


Figure 2. Intensity in the –1 diffraction order as a function of the azimuth angle of the incident linear polarization.

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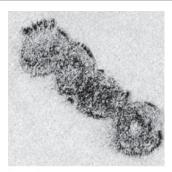
# Temporary holographic data storage using bacteriorhodopsin

Bacteriorhodopsin (bR) is a photochromic protein with interesting photophysical properties. These include: a large spectral shift, more than 150nm between the two key states (B, M) in the bR photocycle; a very high spatial resolution of several thousand line pairs per millimeter; a high quantum yield of 64% for its primary photoreaction: and 100% reversibility.1 These characteristics, plus the fact that they are commercially available,2 make bR films highly attractive as a rewritable holographic medium (see Figure 1).

The ground state, B, has an absorption spectrum that peaks at 570nm. The absorption spectrum is broad enough so holograms can be recorded using a frequency-doubled Nd:YAG (532nm) laser. However, since the material cannot be fixed or gated, performing a reconstruction using a wavelength within the

absorption band will lead to erasure. The material also exhibits thermal erasure. In wildtype bR, this occurs on the millisecond timescale: the genetic variant D96N exhibits a thermal relaxation time of 40-80s. The blue variant D85N can store information for several hours or even days, but this is at the expense of the material's sensitivity. Thermal erasure can be slowed down significantly by cooling the film, as has been demonstrated for both reflection and transmission geometries.<sup>3</sup>

Holograms are generally recorded in D96N bR using a fluence of 3-5mJ/cm<sup>2</sup>, and continuous-wave recordings are more efficient than pulsed. Since bR has constant sensitivity on sub-microsecond timescales, it can be used for the recording of ultra-fast events. However, due to photo-induced and thermal erasure, bR can only be used for temporary storage applications. Currently we use bR for Holographic Particle Image Velocimetry (HPIV, see Figure 2). For this application, two holograms of particles in water are recorded using a small time delay. From the displacement of the particles, the volumetric flow field can be determined. Using bR's polarization-dependent sensitivity, we were able to develop a highly-efficient polar-



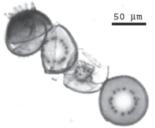


Figure 1. Holographic reconstruction (top) and microscopic image (bottom) of diatoms. Since no shrinkage occurs, holograms in bR can deliver high-resolution images.<sup>5</sup>

ization-multiplexing technique for this application.<sup>4</sup>

In the previous example, bR was used as a temporary holographic medium, from which the image itself was extracted. Alternatively, bR can be used as a holographic buffer. A holographic buffer may be very useful in digital holography. When a wavefront of a fast event con-

tains too much information to be digitized directly on a single CCD, it may be recorded in bR at first. Afterwards, the reconstructed (conjugate) wavefront may be scanned and digitized by using many CCD exposures.<sup>5</sup> For applications where large amounts of data are stored in bR, it is important to maximize the read-out time and speed. Besides already-mentioned solutions such as cooling and polarization multiplexing, it is also possible to perform a wavelength-shifted reconstruction.<sup>3</sup> Furthermore, the formation and decay of a grating in bR can be accurately modelled: insight gained from such simulations can be used to further optimize the recording and reconstruction parameters.<sup>6</sup>

In conclusion it can be said that bR has some very interesting characteristics that makes it well-suited for a variety of applications. Its reversibility is both a strong and a weak point, since it allows for easy experiments with high turn-around time, yet leads to erasure of the stored information. To extend the range of applicability of bR, a number of techniques have been demonstrated that aim at an extended and brighter reconstruction.

We thank Donald Barnhart from the University of Illinois at Urbana-Champaign, as



Figure 2. From double-exposed particles (70µm) to flow field; bR offers several advantages for HPIV.

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## Photoreactive properties of manganese-doped lithium

Photorefractive lithium-niobate crystals have been extensively investigated and used as holographic recording materials. Transition metal dopants, such as Fe, Cu, Ce and Mn, are added to the melt as oxides to improve the photorefractive effect. Among all dopants, Fe has been investigated extensively. For holographic storage, two important system parameters are dynamic range (M/#, or M number) and sensitivity. The larger the M/#, the higher the storage density and the better the signal-to-noise-ratio (SNR). Sensitivity determines the recording speed. Also, it is found that the capacity of a holographic storage system is inversely proportional to the cube of the working wavelength, which means that higher storage density can be achieved by using shorter wavelength. In general, a recording material with large M/# and high sensitivity is desirable: preferably working in the blue spectral region, where the wavelength is relatively short. In addition to the above desired properties, the high sensitivity of the material must not be accompanied by fanning.

One approach to boosting the M/# and sensitivity is to increase the doping level. For example, in the widely used LiNbO<sub>3</sub>:Fe crystals, the highest practical doping level is about 0.1wt% Fe<sub>2</sub>O<sub>2</sub> and is found to be limited by a fast, dark decay due to electron tunneling.1 We have found that the effect of dark decay is less in LiNbO3:Mn than in LiNbO3:Fe, and higher doping levels can be used in the former. The highest practical doping level in LiNbO<sub>3</sub>:Mn has been found to be around 0.5wt% MnCO<sub>2</sub>, and refractive index changes and sensitivities up to 1.5×10<sup>-3</sup> and 1.3cm/J are measured for extraordinarily polarized light at 458nm. Moreover, this high sensitivity is measured in crystals where the fanning is very weak.

Figure 1 shows the measured data-decay time constants of gratings in LiNbO<sub>3</sub>:Fe and LiNbO<sub>3</sub>:Mn. Usually, in crystals with low doping levels, the dominant dark-decay mechanism is proton compensation with characteristic activation energy— $E_a$ — around 1eV. In crystals with high doping levels, the dominant dark decay mechanism is electron tunneling. This limits the highest practical doping levels.<sup>2</sup> From Figure 1 we can see that, although the doping levels in these two crystals are comparable, the dark decay in the Mn-doped crystal is still

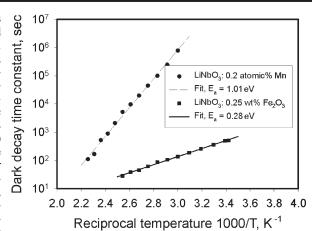


Figure 1. Measured dark-decay time constant of non-fixed holograms in LiNbO,—doped with 0.2atomic% Mn—and in LiNbO, doped with 0.25wt% Fe,O,..

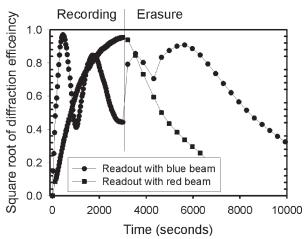


Figure 2. Recording and erasure curves for a 0.84-mm-thick LiNbO<sub>3</sub> crystal doped with 0.5wt% MnCO<sub>3</sub>.

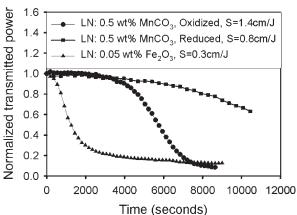


Figure 3. Light power transmitted through the crystal during the fanning measurements.

dominated by proton compensation. In the iron-doped crystal, the dark decay is dominated by electron tunneling. This fact allows us to use manganese-doped lithium-niobate crystals with higher doping levels to get larger M/# and sensitivity. We found, experimentally, that the highest practical doping level in manganese-doped lithium niobate for holographic storage is about 0.5wt% MnCO<sub>2</sub>.

Figure 2 shows a recording and erasure curve for a 0.84-mm-thick crystal doped with 0.5wt% MnCO<sub>3</sub>. The recording and erasure beams were plane waves at 458nm and with extraordinary polarization. The recording intensity was 11.5mW/cm² per beam. During recording and erasure, a readout beam at 633nm and ordinary polarization was added to monitor the temporal evolution of the refractive index changes. Using coupled-wave theory, we estimated the maximum refractive index change as Δn=1.5×10<sup>-3</sup>.

We performed an experiment to quantitatively assess the fanning behavior of several LiNbO<sub>2</sub>:Mn and LiNbO<sub>2</sub>:Fe crystals with approximately the same thickness. One plane wave with extraordinary polarization was split into two beams: one was directed to the crystal with normal incidence, the other to a detector and as the reference beam. The power of the transmitted beam after the crystal was measured from time to time. We used 458nm for LiNbO<sub>2</sub>:Mn and 488nm for LiNbO<sub>a</sub>:Fe, and the average intensities in all the cases were the same: 7mW/cm<sup>2</sup>. In Figure 3 we plot the normalized transmitted power as a function of time. Clearly, from the very beginning. the holographic scattering in LiNbO<sub>3</sub>:Fe builds up very quickly (and therefore the transmitted power drops). LiNbO3:Mn crystals, on the other hand, remain almost unaffected after one hour. Note that the sensitivities of the LiNbO<sub>3</sub>:Mn crystals we used in this experiment are much higher than those of the LiNbO<sub>3</sub>:Fe crystal while the buildup of fanning in LiNbO3:Mn crystals is much slower. From these measurements we see that, indeed fanning is dramatically reduced in LiNbO<sub>3</sub>:Mn crystals for recording with blue light.

One drawback of manganese-doped lithium niobate crystals is the small thermal fixing efficiency. Although thermal fixing has been successfully demonstrated, the efficiency is very low, around

Continues on page 9.

# **Expanding the spatial resolution** of self-processing acrylic films

Photopolymerizable systems have several characteristics that make them attractive as holographic materials for data storage. Interference patterns can be stored in acrylic materials due to the polymerization reaction that causes a structuring of the material at the microscopic scale. The proposed emulsion involves a triacrylic monomer base, a xanthenic dye (sensitive from 450 to 550nm) and a photoinitiator. The layer is embedded between two glass slides. The thickness of the recording layer is defined by a spacer to obtain a value ranging from 30-500µm. The incident light distribution induces an inhomogeneous polymerization that in turn leads to mass diffusion processes (due to concentration gradients in the monomer and dye). The coupling between photochemical conversion and mass transport results in regions with various densities. Volume, thick, phase holograms with high diffraction efficiencies and low scattering noise are then created as spatial variations of refractive index. No chemical or thermal post-treatment is required.

Transmission and reflection gratings were recorded for fringe spacings bigger or smaller than the incident wavelength (514nm). Table 1 shows the experimental results. A constant dose of 650mJ/cm² was applied in order to reach a dye consumption of 85%, allowing nondestructive readout at an active wavelength (green light). This energy density corresponded to a monomer conversion rate of about 35%. The recording of high-spatial-frequency gratings (one beam in Table 1) was performed using an argon-ion laser at 514.5nm and adequate prism set-ups.<sup>2,3</sup> Total internal reflections inside the holographic plate or coupled prism then produced an inter-

ference pattern from just one incoming beam.

During conditioning, incoherent pre-irradiation allowed adjustment of the initial viscosity of the emulsion. The standard pre-illumination corresponded to gelification of the formulation (changeover from liquid state to gel form). Its duration could be increased in order to reduce both the polymer chain propagation from bright to dark regions and the diffusion contribution. A trade off was then necessary to avoid a decrease in chemical reactivity caused by pre-polymerization.

The optimal response was obtained in the case of the unslanted transmission sinusoidal gratings with a fringe spacing around 1µm. Diffraction efficiency (diffracted intensity to incident intensity ratio) was then 80%, corresponding to a refractive index modulation amplitude of 0.006. The material's response decreased at elevated fringe inclinations for transmission gratings, as in the case of reflection holograms with fringe planes parallel to the layer (Table 1). This was partly due to material shrinkage effects occurring during polymerization. Though it did not exceed 7%, it introduced a Bragg-mismatch during holographic exposure.3 Reducing the shrinkage via stronger pre-exposures led to better diffraction efficiencies.

All these experiments were motivated by applications such as :

- optical data storage
- fabrication of optical devices using the photostructuring properties of a polymerizable system doped by NLO molecules<sup>1</sup>
- realization of edge-extraction holographic filters<sup>2</sup>

 fabrication of planar substrate-guided-wave components, benefiting from the two glass slides embedding the layer. The grating fringe inclinations and diffraction angles corresponded to coupling or polarizing proper-

cal switching.<sup>2,3</sup>
The Région Alsace is gratefully acknowledged for its financial support.

ties related to expected applications in opti-

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## Table 1. Diffraction efficiency and refractive index modulation (according to H. Kogelnik's Theory) versus fringe spacing for different recording conditions (thickness =50mm).

- Pre-irradiation: 1.1 means that the duration of the pre-irradiation is 1.1 times longer than the duration needed for just obtaining the gelation of the formulation.
- Set-up: one or two incoming beams (one beam provided better stability in the incident fringe patterns).
- Fringe inclination: the angle of the interference fringes from the normal to the photosensitive plate.

| Pre-irradiation                    | gelation 1.1 |                  |     |     |     |      |      | 1.1  | gelation |      | 1.7  |
|------------------------------------|--------------|------------------|-----|-----|-----|------|------|------|----------|------|------|
| Set-up                             |              | 2 incident beams |     |     |     |      | 1    | 1    | 2        | 1    | 1    |
| Fringe inclination (°)             |              | 0                |     |     |     |      |      |      | 22.5     | 90   | 90   |
| Fringe spacing (µm)                | 5            | 2.5              | 1.5 | 1   | 0.5 | 0.24 | 0.24 | 0.24 | 0.45     | 0.24 | 0.24 |
| Diffraction<br>efficiency (%)      | 50           | 60               | 80  | 80  | 65  | 23   | 49   | 65   | 10       | 10   | 19   |
| Index modulation x 10 <sup>3</sup> | 3,2          | 3.6              | 4.3 | 5.3 | 3.5 | 1.2  | 1.8  | 2.4  | 1.0      | 1.0  | 1.1  |

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**20 February 2004:** Ideas for articles you'd like to write (or read).

**2** April 2004: Calendar items for the twelve months starting June 2004.

# A dual-cure photopolymerizable system with applications in visible and mid-infrared holography

The first recording material to make holograms was the photographic emulsion. Later, other photosensitive materials were used. However, there is no universal film that can be used in all the recording situations. The reason is that the principle of holography (interference pattern recording) encompasses different variables like wavelength, power, and polarization of the recording and reading beams. Besides, the resolution required of the recording material is related to the angle between the object and reference beams. We have used an innovative photopolymer mixture (dual-cure system) to record holograms with visible light. The mixture has also been used in the fabrication of holograms that work with mid-infrared light (10.6µm)

## The photosensitive mixture

The dual-cure polymer system is based on an ultraviolet (UV)-curing vinylether monomer and an acrylate olygomer. This mixture undergoes polymerization mainly by radicals. Acrylate monomers polymerize first and faster than vinyl liquid monomers which act as plasticizing agents. The resulting polymer consists of a three-dimensional structure of two interpenetrated networks. The dual-cure system self-develops during the recording of the hologram. It can be fixed in its original recording position.

#### Holography with visible light1

Optical characterization of the film was done by recording interference gratings. The light source was an argon laser ( $\lambda$ =0.5145 $\mu$ m). Readout of the gratings, performed simulta-

neously with the recording, was done with a He-Ne laser ( $\lambda$ = 0.6328 $\mu$ m). The angular position of the reading beam was set at the Bragg angle because gratings were of the thick type. Diffraction-efficiency behavior—as a function of power density of the recording beams, the thickness of the films, and the spatial frequency of the interference pattern, was studied. A maximum diffraction efficiency of about 60% was obtained. Applications of the dual-cure mixture comprised the recording of single- and double-exposure holograms. Also, the relief depth of a conical surface was obtained by a contour-generation method (see Figure 1).

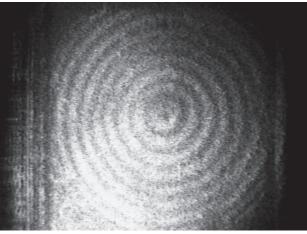


Figure 1. Interferogram of a conical surface obtained by the contour/ generation method.

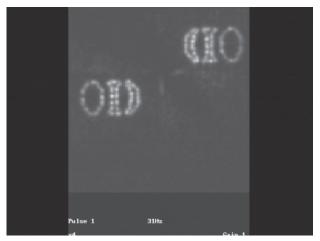


Figure 2. Thermal image given by a hologram taken with a pyrocamera.

## Mid-infrared holography<sup>2</sup>

In the past, efforts have been made to apply the holographic principle when light, other than visible, is used. Wavelengths in the UV, near-infrared, and mid-infrared regions of the spectrum have been used to record holograms. A drawback with mid-infrared light is poor recording resolution due to heat conduction between interference fringes. To avoid this problem we have made a hologram based on a computer generated hologram (CGH). The final diffractive element was made on a polymeric, infrared-transmissive material: polyethylene.

The steps to fabricate the diffractive element were as follows: a) a phase–transmission func-

tion was calculated by the computer; b) this function was printed on a high-contrast film to get a binary amplitude mask; c) by UV photolithography, and using the mask, the information was transferred to a dual-cure polymer film. At the end of the process, the film had a surface relief which was used as a master. Finally, d) thermal embossing was used to make the polyethylene diffractive element. The CGH was calculated with the letters 'CIO' as the object. After fabrication, the polyethylene CGH was illuminated with a CO<sub>2</sub> beam and a germanium lens was placed after the hologram to obtain the inverse Fourier transform. Thermal intensity distribution was analyzed using a pyroelectric camera. The thermal image formed is shown in Figure 2. In the center, no zero order is present: it was blocked to avoid damage to the sensitive element of the camera. Diffraction efficiency of this hologram was about 21%.

To study the response of the polyethylene material to the spatial frequency of a pattern, a series of gratings were made. Ronchi gratings with spatial frequencies of about 4lp/mm, 6lp/mm and 12lp/mm were used as masks. A parameter in the experiment was the thickness of the polymer layer. It was found that the maximum diffraction efficiency (for IR light) was about 29% for a grating with 4lp/mm, with a surface modulation of about 10.5μm.

S. Calixto acknowledges fruitful discussions with Reyna A. Duarte-Quiroga and Manuel Ornelas-Rodriguez.

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# Comparison of photosensitive holographic recording materials

Research in our group has mainly centered on the development, characterization and optimization of holographic recording materials. Here we present a brief review of the most important results of the four materials we have been working with..

## BB640 Holographic emulsions

These have been studied with different processing schemes. With energetic developers (D8) we have obtained anomalous D-Log E curves reaching densities of 13. Some interesting new features have been found. First, low scattering and high-diffraction efficiencies and have been achieved with direct-rehalogenating and reversal bleaching: better than with classical fixation-free rehalogenating bleaching. Also, recently, reflection ho-

lograms have been realized with high efficiency and nonlinear recording effects.<sup>3</sup>

### Purple-membrane polyacrylamide films

Recently, bacteriorhodopsin has been used for many applications in optical image processing: optical memory, optical phase conjugation, real-time holography, and spatial light modulation, for example. This photochromic retinal protein is contained within the purple membrane of halobacterium halobium, which is encountered in extreme conditions.4 We have prepared wild-type purple-membrane polyacrylamide films of 50-300µm thickness and recorded read-write holographic gratings in the green zone, reconstructingd with 100mW/cm<sup>2</sup> of a wavelength where the material does not absorb. This resulted in diffraction efficiencies near 1.5% and an energetic sensitivity of 200mJ/cm<sup>2</sup>. These preliminary results are similar to those reported in the bibliography, so this genetic variant of the purple membrane should be good once optimized.

### Photopolymerizable dry polymeric films

Recently we have studied the holographic gratings of acrylamide based on polyvinylalcohol dry films in some detail, and have presented the characterization of the angular responses of these gratings, with and without bi-functional crosslinking agents. As we demonstrated in

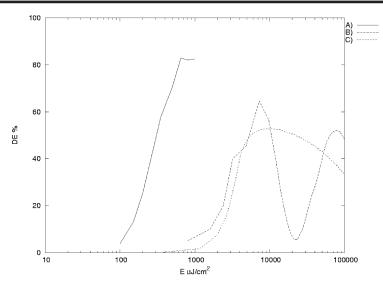


Figure 1. Energetic variation of the diffraction efficiency for different materials: A) BB640 emulsion developed with AAC and bleached with R9. B) A10µm-thick, dry photopolymer film of composition: acrylamide, triethanolamine, N,N'-dihidroxyethylen-bisacrylamide, polyvinylalcohol and methylene blue. Intensity was 4mW/cm².8 C) Solgel photopolymerizable material of composition: acrylamide, yellowish eosin, and triethanolamine, all in a porous glass. Thickness 1.5mm, intensity 4mW/cm².

1998, the addition of this kind of compound increases the energetic sensitivity. Values near to those of commercial compositions are produced, as are over-modulation effects 100µm-thickness layers. 8-9

On the other hand, a theoretical model has been developed for compositions without bifunctional crosslinking agents, using acrylamide as monomer. This model allows us to obtain data on the photopolymerization kinetics and the length of the formed polymeric chains. The results are in accord with those of other analytical techniques.<sup>10</sup>

## Photopolymerizable sol-gel materials

To achieve high storage density, the recording medium must be more than 1mm thick. However, in PVA films the values obtained were around 100µm, or in some cases 200µm. In this sense, an important alternative to polymeric binders are sol-gel glasses. These can be fabricated into thick films or monoliths with the desired thickness and properties such as high optical quality, rigidity, and envinromental stability. We have recorded holographic gratings in a photopolymerizable sol-gel glass using acrylamide as the monomer, triethanolamine as the coinitiator, and yellowish eosin as the photoinitiator. Diffraction efficiencies near 55% were with an exposure of 8mJ/cm<sup>2</sup>. The resulting holographic gratings were not stable,

and this instability has been used to determine the diffusion coefficients of acrylamide and polyacrylamide inside this glass.

In the figure we compare these materials, not including the purple membrane film due to its low diffraction efficiency. As can be deduced, holographic emulsions perform the best results. However, in sol-gel materials—due to the high thickness—it is expected that better sensitivities will be achieved with better composition.

This work was financial-ly supported by the Interministerial Commission for Science and Technology (CICYT) of Spain (projects MAT2000-1361-C04-03 and MAT2002-0169) and CTDIB/2002/134 from the Valencia General Council on Innovation and Competitiveness.

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Continues on page 10.

## Color holography with new BBVPan plates

Color reproduction of the light scattered by a diffusing object recorded in a single hologram has been achieved in the past. The main problem found by holographers is getting the appropriate recording material. Very good color holograms have been recorded with Slavich PFG03C panchromatic emulsion. The main problem when working with these plates is their rather soft gelatine, which means that laboratory conditions must be kept stable to avoid moisture absorption by the plates during recording: something that will affect color reproduction of the final hologram. The new emulsion BBVPan, manufactured by ColourHolographics Ltd., has features that overcome this problem as well as increased sensitivity.

Here we present preliminary results obtained with BBVPan plates used to record color holograms of a diffusing object. The setup used is a conventional Denisyuk configuration with three laser beams, see Figure 1. Wavelengths used were 442nm from a 80mW/cm² He-Cd laser, 532nm from a 5W/cm² cw diode-pumped frequency-doubled Nd:YAG laser, and 632.8nm from a 24mW/cm² He-Ne laser.

Plates were pre-sensitized by soaking in a 3% Triethanolamine (TEA) solution in water for two minutes, followed by soaking the wet plates in de-ionized water for another six minutes.<sup>2</sup> Plates were dried with a photographic roll and warm air, and left to cool down for 30 minutes in normal laboratory conditions. A sequential exposure follows, starting with the blue recording followed by the green and the red. Optimum exposure energies for higher brightness and good color reproduction were the following: He-Cd 225µW/cm<sup>2</sup>, Nd:YAG 350µW/cm<sup>2</sup> and He-Ne 1.5mW/cm<sup>2</sup>. After exposure, plates were developed with standard AAC developer (ascorbic acid 18g/l, sodium carbonate 60g/l) for four minutes at 20°C, washed, bleached

He-Ne Laser

Nd-Yag Laser

Shutter

Beam Expander

Folding Mirrors

Figure 1. Setup for recording color holograms with three lasers.



Figure 2. Photograph of the reconstructed image of the color hologram, shown together with the real object under the same illumination. Actual size of the hologram is 6×10cm.

with fixation free bleaching R-10 (sulphuric acid 10cc/l, ammonium dichromate 2g/l, potassium bromide 35g/l), washed and soaked in de-ionised water with photoflo and a few drops of acetic acid to prevent printout. A color print of the final hologram, replayed with an halogen lamp and with the real object under the same illumination, is shown in Figure 2. Replay wavelengths with a reconstruction angle of 45° are 445nm, 538nm and 639nm: very close to the recording wavelengths.

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- ColourHolographics Ltd. say that, "We consider that, with our modified formula, it now appears unnecessary to pre-sensitize the plates. The only difference is that development time to density is twice as long but the efficiency is the same." The author has not tested this yet.

## Photoreactive properties of manganese-doped lithium

Continued from page 5.

 $10^4$ . One possible explanation this is that most of the Mn traps in LiNbO $_3$ :Mn crystals are Mn $^{2+}$  and the modulation depth is very small during revelation. $^{2,3}$ 

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## Polarization-holographic data storage

Continued from cover.

provides a higher degree of data security than conventional bit-oriented storage in magnetic or optical media. This is because the hologram records information in the form of a complex phase or amplitude distribution describing the whole data page while bit-oriented storage records spatially-separated bits sequentially along a well-defined track. Polarization holography, where information is coded in the spatial variation of birefringence, represents even more efficient coding. Retrieving data is not possible by conventional intensity-sensing optical devices: the only possible way to reconstruct data is to build a holographic setup that reproduces the wavefront and the polarization state of the reference wave to very high precision.

Data security can be further enhanced by phase encoding the recording waves. Random or deterministic phase modulation of the signal or the reference wave during hologram recording is a well-known and widely-investigated technique

of data multiplexing and encryption.<sup>5, 6</sup> We apply this technique to thin polarization holograms. This does not allow high multiplexing factors, but presents an advantage in the security: due to the lack of Bragg selectivity, an imperfect reconstruction reference wave will give rise to the same diffraction efficiency as the correct reference, but the reconstructed signal will contain random noise instead of data.

The operating principle of our system is shown in Figure 3. The data page is optically Fourier transformed onto the storage material, and the hologram is recorded with the phase-modulated recording reference wave in the thin photo-anisotropic film. For reading out, the hologram is illuminated by a reconstructing reference wave, and the resulting beam is Fourier transformed back to produce the output image. If recording and reconstructing reference waves are identical, the input

Reconstruction

data page will be perfectly reconstructed. If the recording and reconstructing reference waves are different, the output image will be noisy. To prove the feasibility of the concept, we have built an experiment demonstrating the phase-coded reference holographic recording and readout. The images reconstructed by the proper reference beam have presented a few deterministic pixel errors. These are due to localized defects in our phase mask used for spreading out the Fourier space. Holograms reconstructed by an improper reference wave provides an image containing 40-60% pixel errors.

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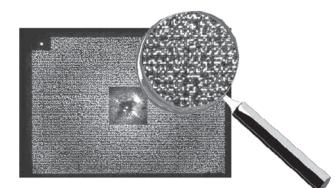


Figure 2. A data page recorded and reconstructed by the read&write unit. The data page contains 16.4Kbyte information recorded in a 220µm by 220µm Fourier hologram, that corresponds to a 2.77bit/µm² (223Mbyte/in²) storage density.

## 

Figure 3. Holography with a phase-coded reference beam. The recorded hologram can only be reconstructed with the properly-coded reference, otherwise the system returns noise instead of data.

## Comparison of photosensitive holographic recording materials

Continued from page 8.

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# Secure holographic memory system with multi-dimensional keys in a photorefractive crystal

Holographic memory is expected to realize a large storage capacity—1Tbyte in a 5inch disk-and an ultrafast data-transfer rate: 1Gbit/s. This makes it potentially useful for digital libraries, high-resolution movies, or in mobile devices. In data storage, security is one of the most important issues. Optics can provide security beyond that of digital encryption techniques because various physical parameters and the two-dimensional

nature of light distribution can be used for the data encryption. Here we present our approach to developing secure holographic memory systems using multidimensional keys. We also present a promising photorefractive material for nonvolatile holographic recording that can be used for rewritable media.

Optical encryption<sup>1</sup> can be used to prevent unauthorized users from accessing information stored in holographic memory systems. A number of ways of encrypting the stored data exist.<sup>2-6</sup> Some are based on encrypting the data and others on preventing memory access by unauthorized users. To implement the first of these, optical encryption techniques such as double-random phase encryption or XOR can be used. The former is one of most powerful ways of protecting data. In double-random phase encryption, two random phase-masks located at the input and Fourier planes can convert an original image into a stationary random-noise image. A phase key at the Fourier plane is required to retrieve the original data. If an incorrect key is used, the reconstructed image will consist of random noise. The number of possible two-dimensional phase keys is extremely large, so it is impossible to decrypt the

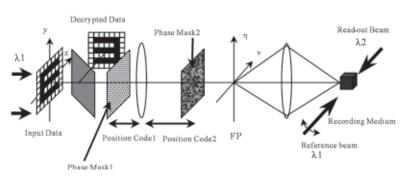


Figure 1. A schematic of a secure holographic memory with multi-dimensional keys. FP denotes the Fourier plane.

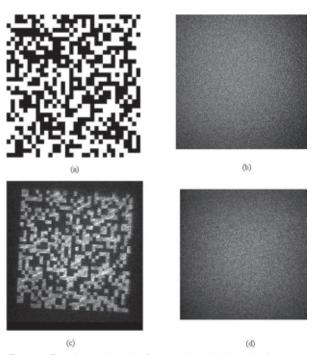


Figure 2. Experimental results of encryption and decryption in our secure holographic memory: (a) original input image; (b) encrypted image: (c) and (d) reconstructed images by use of correct and incorrect phase keys, respectively.

data within meaningful time without knowing the right one.

Figure 1 shows our concept of a secure holographic memory system that employs wavelength, three-dimensional key position, and polarization to modulate the optical wave: this is in addition to random phase keys. The number of potential combinations of keys is large enough to provide a very high level of security. In Figure 1, an original binary image is modulated

by two random phase masks located in the Fresnel domain. The modulated wave is Fourier transformed and then recorded in an intensity-sensitive medium such as a photorefractive material or photopolymer. Sequences of binary images are stored by angular or shift multiplexing. The phase-conjugate reconstruction can recover the original binary image by compensating for the phase modulation from the random-phase masks. The wavelength of the recording beams can be used to protect the stored data when the recording medium is sensitive to a wide spectral range of light. Due to the Bragg condition, mismatch between the recording and reconstruction wavelengths prevents the holograms from being read out. The combination of these encoding methods makes it impossible to decrypt without the key information. Two random phase keys can be located in the Fresnel domain. In addition to the phase information, the positions of two phase-masks are used as new keys for successful reconstruction of original data. The position of the masks can have as many as three

Continues on page 2.

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