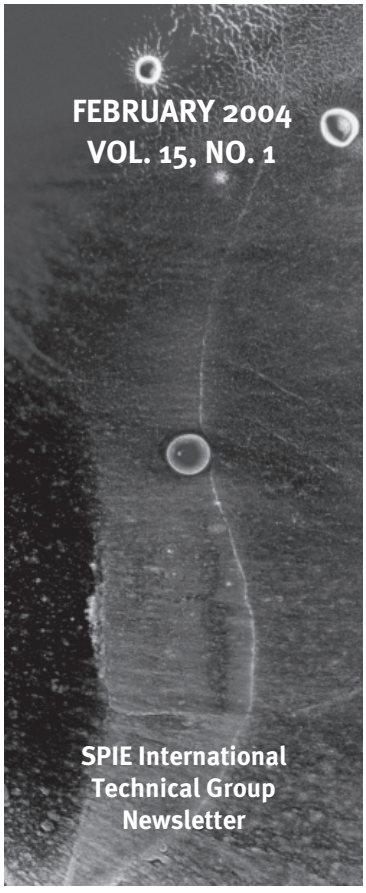

Special Issue on:

Part 2

Holographic Materials for Data Storage

(See URL at the top of the page for part 1)

Guest Editor
Dr. John Sheridan
University College, Dublin

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Optics in Information Systems

Re-writable high-density optical recording on azobenzene thin film

Azobenzene-functionalized polymers are interesting and potentially-useful materials for optical device applications. One of the key features of this material is that photo-anisotropic azobenzene moieties, upon irradiation with polarized light, exhibit photo-induced molecular reorientation through the *trans-cis-trans* photo-isomerization cycles. This re-orientation process has been widely investigated from the viewpoint of data storage: especially for volume holography. Another important feature is that the azobenzene-polymers allow light-induced mass transport resulting a formation of photo-induced surface relief (PSR). This unique phenomenon was reported in 1995[1,2] and many other researchers have investigated it since then.[3-7] So, for instance, if an azobenzene polymer thin film is irradiated by interference fringes (generated by two-beam coupling of coherent light with appropriate wavelength and irradiance) then photo-fabricated surface relief gratings (SRG) with sub-micron modulation height are easily obtained.

Figure 1 shows typical examples of SRG with 1μm pitch and 0.5μm modulation depth. Several experimental studies have revealed that this phenomenon does not originate in the laser ablation process, but in the molecular mass transfer from the bright region to the dark region. It is surprising but true that PSR formation is carried out in the solid-state thin film well below the glass transition temperature (T_g) of the polymer. Thus, this phenomenon is quite different from other conventional micro-fabrication techniques such as photolithography, laser ablation, and thermal deformation. The PSR structure can be precisely controlled by the intensity, spatial distribution, and polarization conditions of the irradiated light, as well as the irradiation time. Furthermore, though

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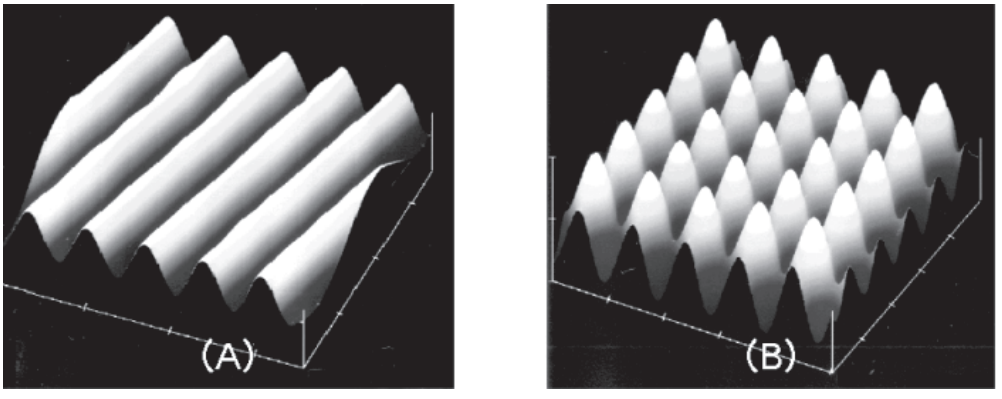

Figure 1. Topographic images of typical photo-fabricated surface-relief gratings observed using an atomic force microscope.

Editorial

Welcome to the second part of this special issue on holographic materials. Although many holographic applications exist for innovative materials, perhaps the most topical, as outlined in my previous editorial, is holographic data storage. Single discs promise hundreds of gigabytes of data storage. Many optical signal processing (OSP) issues and opportunities arise as holographic materials increase in quality, and as the optoelectronic and thermo-mechanical performance of the read-write hardware improves.

However, we must ask the following questions. Does a consumer market exist for a compact disc capable of holding hundreds of feature films? Does a high-end data storage market exist for such removable media? Can the management of data stored optically be improved using OSP techniques?

The first question begs a further one: what will consumers need this scale of storage space for? Even at my most imaginative I am unable to guess a fraction of the potential uses. But please take note of three facts. First, packs of 100 blank CDs (~70 GB) are sold in many high street stores today at less than 30 cents a CD. Second, I bought my first video tape ~15 years ago, the same year I bought a MacPlus with an amazing 10MB external hard disk. My 9-year-old son currently owns over 100 videos and plays games on an old computer with a 20GB HD. Finally, the digital world continues to develop: i.e. digital cameras/recorders, digital paper, high definition TV, broadband internet, DWDM, 3G telecommunication etc. A large consumer appetite for data exists.

As for our second question, very-large-scale data storage systems provide the backbone of every large business, academic, medical, and government organization. Commercial systems, made up of hundreds of tape drives capable of jointly storing tens of terabytes, are widely available. Whether for commercial, security, or legal compliance reasons, everything from e-mails to invoices must be retained for some period of time, and failure to do so can be financially disastrous. Large-scale science, such as the CERN, LIGO, and genome projects, generate terabytes of data in real time. Finally, the provision with each PC of a single encrypted disc containing every possible user system and version of system operator package becomes feasible using this technology.

To our final question: large data storage firms today are primarily involved in software management as opposed to hardware/device issues. Storage systems should provide long term security, low cost per bit, transparency, and high speed access. Moving to an optical storage system, which potentially involves massively parallel operation, opens up the possibility of incorporating ideas from optical signal processing: this includes all-optical correlation and encryption.

Holographic materials have reached the stage in their development where practical commercial systems for holographic data storage can be built. The implications for OSP (and optical computing) are very significant.

I thank again all the contributors for their excellent work. I encourage you to read each piece in this issue and the last (see the URL on the cover of this issue) and to contact those whose work is of interest to you. I also thank the chairs of the Optics in Information Systems Technical Group for hosting this second issue in their newsletter.

John T. Sheridan
University College Dublin

Re-writable high-density

Continued from cover.

the PSRs are very stable, they can be erased by heating the film above T_g, or via uniform irradiation. Due to the simplicity of this process, it we expect it to be used for optical device fabrication, ultra-high-density optical data storage, and so on.

To make use of the PSR phenomenon, a novel optical-data-storage method has been proposed.[8] To improve storage density, reduction of pit size is a major focus of research in conventional digital data storage. With our, very different, method, it is possible to achieve ultra-high recording density—more than 1Tb/inch^2—by inscribing multi-code on the azobenzene polymer medium as a PSR pattern. The proposed read/write system is shown in Figure 2. A square indicates the area reserved for one pit, in which anisotropic dot series along different directions are inscribed as PSR patterns with different surface modulation depths (see part A). If we input an elliptical read-beam with a high axial ratio and rotate it around the pit center, then the intensity of the transmitted (or reflected) read-beam changes depending on the recorded PSR patterns because they act as diffraction gratings (or scatterers).

The read-beam intensity for such PSR patterns is shown in part B as a function of rotation angle. From this it is clear that one pit takes $(m+1)^n$ information codes from the combination of angular (n) and depth (m) possibilities in the PSR patterns. Therefore, assuming that the pit is 80μm square, and there are 10 and 4 angular and depth possibilities, respectively. The estimated recording density is approximately 1Tb/inch^2. A study of the feasibility of this recording method is ongoing, and related patents pending.

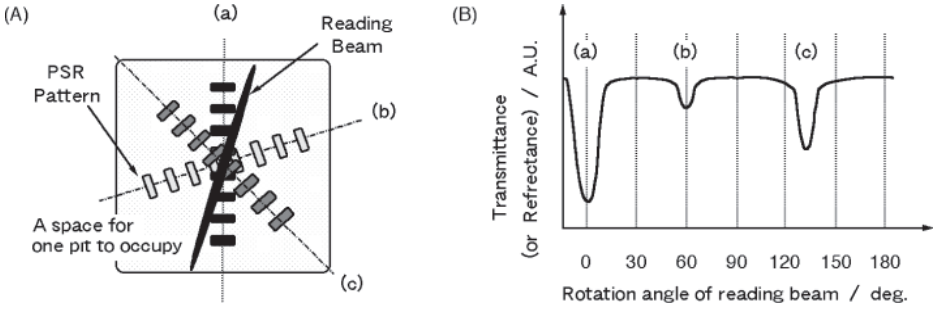

Figure 2. Schemata of a proposed multi-coding high-density recording method (A) and a read-out signal waveform (B). (Darker corresponds to deeper PSR and larger intensity modulation.)

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Improvements in holographic photopolymers at Weizmann Institute of Science

In recent years, we have investigated several photosensitive recording materials and tested them for use in making holographic optical elements (HOEs).[1-5] In some of these investigations, we considered photopolymer formations with both hydrophilic as well as hydrophobic binders, such as poly-vinyl alcohol (PVA), poly-vinyl butyral (PVB) and poly-methylmethacrylate (PMMA). These binders have additives that influence the exposure sensitivities, diffraction efficiencies (DEs), and both temporal and dimensional stabilities. Specifically, we introduced the use of iodonium salt sensitization into hydrophilic systems, using diphenyliodonium chloride (DPI-Cl), in conjunction with eosin Y and triethanolamine (TEA). We showed that a combination of both DPI-Cl and TEA can significantly increase exposure sensitivity. We also considered the influence of the PVA binder parameters on sensitivity during real-time holographic recording. Experimental results indicated that a higher molecular weight of the binder increased the polymerization rates, as expressed by increased exposure response.[2] This probably originates from dipole-dipole interactions between the binder and the monomer; so the diffusion coefficients in a less polar binder matrix increase, thereby leading to increased sensitivity.

Some representative results of our most recent investigations are shown in the figures. Figure 1 shows the experimental diffraction efficiency as a function of exposure for two photopolymers: one with a PVB, the other a PMMA, matrix. As is evident, the former exhibits significantly better exposure sensitivity than the latter. We believe that both the density of the binder and its dipole moments—which are larger in PMMA—are responsible for the reduced response.

To obtain the maximum diffraction efficiency for a given layer thickness, we investigated how to increase the dynamic range of refractive index variation (Δn). Such an increase may be achieved by choosing an optimal balance between binder and monomer indices, and their respective concentrations.[6] We increased the Δn in our photopolymers by introducing a new acrylic monomer species of phenylthioethyl acrylate (PTEA by Bimax). For PTEA, $n \approx 1.56$: significantly larger than that of phenoxy-ethyl acrylate (POEA) where $n \approx 1.51$. Figure 2 shows both the theoretical and experimental angular responses of holographic gratings recorded in a PTEA formulation. Here, the optimized formulation with PTEA exhibited $\Delta n \approx 0.012$ for holographic gratings of ~2000cy/mm recorded at 514nm in a ~45μm-thick layer: significantly higher than $\Delta n \approx 0.007$ with a POEA formulation.

Another innovation is the use of a new type of spectral sensitizer (photoinitiator) in our PVB-binder-based photopolymer materials. We successfully replaced the xanthine-type sensitizers (eosin, erytrosin, etc.) with a species labelled NU-470 (by Spectra Group Ltd.) of 5,7-diiodo-3-butoxy-6-fluorone, in conjunction with a iodonium salt accelerator (by Sartomer). While no dramatic change in exposure sensitivity was evident when compared to xanthine dyes, the new materials have better post-exposure bleaching properties and reproducibility: the former is of practical importance for HOEs with high transmittance requirements.

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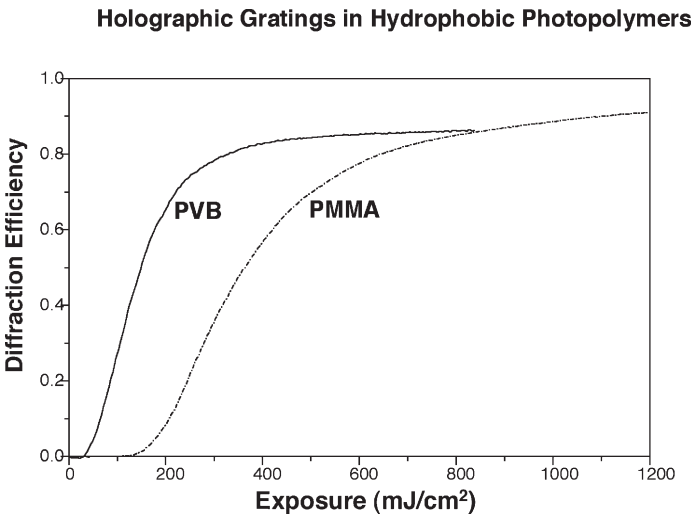

Figure 1. Diffraction efficiency (DE), as a function of exposure of holographic gratings, recorded in photopolymers of hydrophobic-based binder formulations.

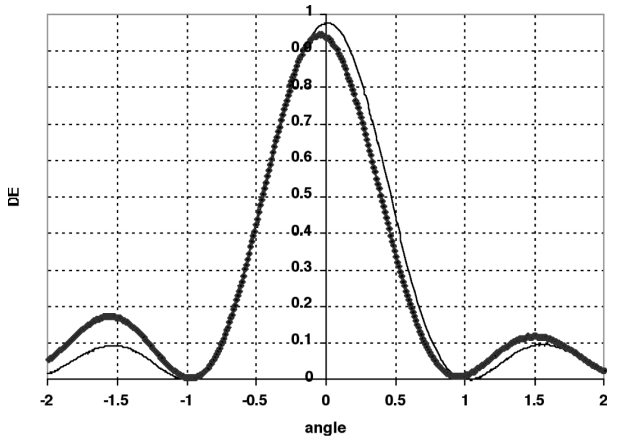

Figure 2. Theoretical and experimental DE as a function of readout angle (deviation from Bragg conditions) of holographic gratings recorded in the photopolymer layer of a PVB-based binder formulation, including the PTEA monomer. The theoretical angular response (solid line) fits the experimental data (diamonds) for a layer thickness of 45μm and a $\Delta n = 2 * n_1 \approx 0.012$, recorded at 514nm and read out at 633nm.

High-density, low-BER data storage with Aprilis technology

The Aprilis, Inc. HMD-120 disk and HMC-050 card holographic media technology were announced at InterOpto Japan in July 2002 after a number of years of invention and advanced development. The technology is based on holographic, photo-polymerizable recording materials: systems of organic molecules that rely on photo-initiated polymerization to record volume phase holograms. However, we use a proprietary chemistry method—Cationic Ring Opening Polymerization (CROP)—to replace more conventional free-radical monomers for recording holograms.[1-2] CROP provides several key advantages for holographic recording over other methods of photo-polymerization. These advantages are inherent to the recording chemistry and relate to a unique combination of key performance attributes such as high recording sensitivity, excellent image fidelity, and large, accessible dynamic range per unit thickness.[3-4]

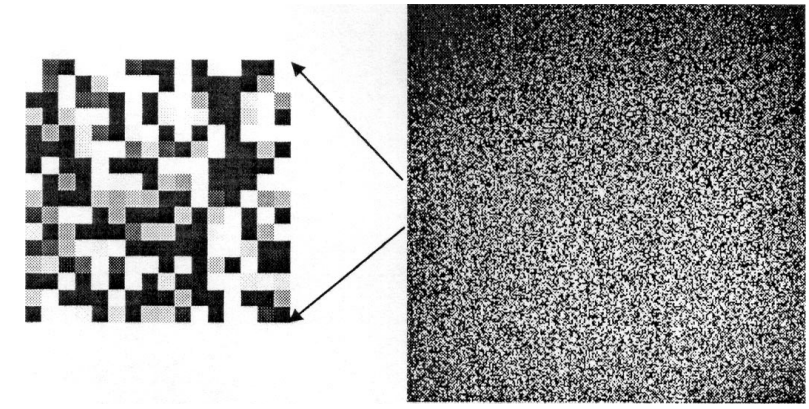

Figure 1. Sample hologram read out at 10Gbit/sec and an enlarged portion of it.

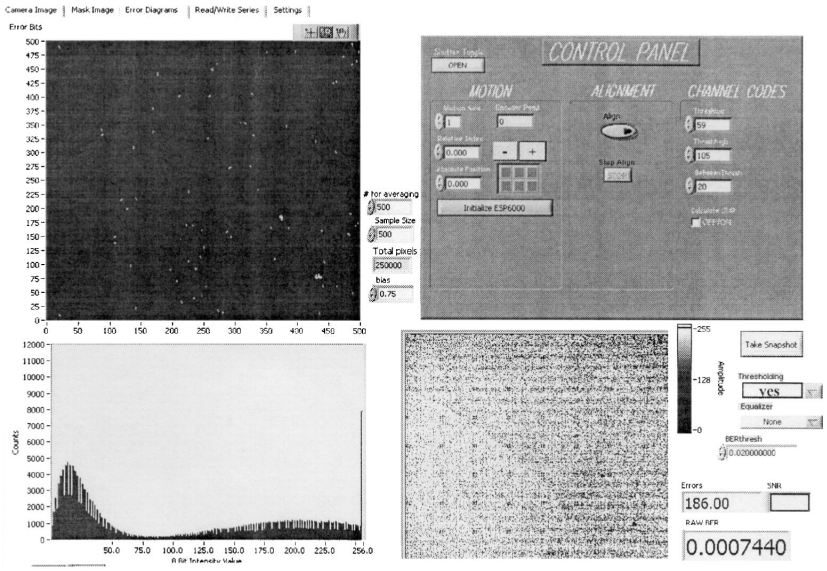

Figure 2. Bottom left is the histogram (1s and 0s) of the direct readout of the reconstructed image (bottom right). The raw BER for this page (#48/100 co-locationally-multiplexed digital 512² data pages) is 186. Top left is the error map, with a software control panel top right.

The improvement in image fidelity and accessible dynamic range is primarily a consequence of a substantial reduction in shrinkage, inherent to the ring-opening volume compensation of the CROP method: that corresponds to at least three times less shrinkage than for the equivalent number of free-radical photo-polymerization reactions during hologram recording.[4-5] Additionally, the method exhibits improved uniformity of spatial frequency response in the typical range used for data-page recording. This is due to the combination of fast polymerization and fast diffusion rates.[6]

The high recording sensitivity achieved is also related to the shrinkage advantage, in that more chemistry is available for recording information with fidelity. Typically, about 95% of the attainable dynamic range is achieved with a cumulative exposure energy of only about 200mJ/cm^2, which represents at least an order of magnitude improvement over other photopolymer systems used for digital data-page recording.[4] Free-radical photo-polymerization methods must consume significantly more chemistry in order to reduce shrinkage to the requisite level for a particular medium thickness, thus leaving less available for holographic recording. This difficulty mounts as thickness increases due to restrictions imposed by Bragg selectivity.[7] When recording must take place at higher extents of polymerization then these materials are more glass-like in their physical properties, which substantially retards diffusion rates. Additionally, only small amounts of reactive monomers are left available to participate in recording.

Aprilis' technology's advantages in performance won its selection by the DARPA-funded Photo Refractive Information Storage Materials (PRISM) consortium in 1999 as the, "medium of choice," out of 57 candidates evaluated for use in the Holographic Data Storage System (HDSS) demonstrations. A culmination of these efforts was a demonstration of the Aprilis technology in a holographic-disk digital data-storage system at Stanford University in November 2000. The system performed correlation multiplexing in a co-axial optical configuration with a rotating disk architecture. It was used to record megapixel digital data pages at a rate of 1Gbit/sec (channel) in an Aprilis disk moving at 300rpm.

The rapid real-time recording was carried out using pulses of a Lightwave Q-switched 532nm Nd:YAG DPSS laser. The total numeric aperture of 0.75 of the HDSS custom double-Fourier-transform (FT) lens was divided between a central, high-resolution, low-distortion portion (NA=0.36, <±0.15nm distortions), which was used by the spatial light modulation (SLM), and the outer area, which was used by the speckle reference beam.[8-10] The Aprilis advantage in recording sensitivity under such rigorous recording conditions was at least 100 times other photopolymer media. Readout was also carried out while the Aprilis disk was moving at 300rpm. Retrieval of 12 simultaneous video streams was demonstrated corresponding to sustained data rates of 1Gbit/sec (channel), and feasibility for an optical transfer rate of 10Gbit/sec was demonstrated.[8-10] A sample megapixel digital hologram, read out at 10Gbit/sec, is shown in Fig-

ure 1.

These demonstrations emphasized the data-rate advantage of digital holographic data storage but at a moderate areal density (less than 10 bits/μm$^{2)}$. In 2003 we demonstrated significant new milestones for areal density in photopolymerizable recording media: we achieved a storage density >100 and >150 channel bits/μm^2 in only 300μm- and 400μm-thick recording layers, respectively.[11-12] This represents about a threefold improvement over previous results reported for all photopolymer candidates at any thickness. Fully co-locational 262kbit digital data pages were recorded in a storage location of area 0.27mm^2 by use of an Aprilis proprietary implementation of azimuthal and angle multiplexing. A classic 4f optical configuration was used for both the object and reference beams, and recording was carried out at the fractional FT plane. Average raw bit-error-rates (BER) of 4.4×10^{-4} and 2.3×10^{-3} were achieved for 100 and 150 channel bits/μm^2, respectively.[12]

Bottom left in Figure 2 is a representative histogram of the distribution of intensity values of 1s and 0s for reconstruction of #48 of 100 co-locationally-multiplexed 262kbit/page holograms recorded at 100 bits/μm^2. Bottom right is the reconstructed data page as imaged onto a CCD detector with 2/1 over-sampling. Top left is an error map of the reconstructed image page, directly determined by comparison of the original and output pages. In this map, white and black indicate correctly and incorrectly-assigned pixels, respectively. There are a few small clusters of errors found in all the data pages at the same pixel locations—at the *x*-direction positions near pixels #375 and #440. These were subsequently determined to arise from particles on the lens elements. The total number of errors for this page, after application of thresholding of intensity values in sub-groupings of the full page, is only 186: this corresponds to a low raw BER of just 7.4×10^{-4}. Such low values are typically only observed for photopolymer holograms recorded at an areal density of less than about 40bits/μm^2. The high fidelity recording characteristics of our imaging system, and the unique recording attributes of the CROP recording medium, allow us to achieve this at 100 bits/μm^2 and more.

In Figure 3 is shown a representative theta scan of the set of angle-multiplexed holograms for the φ=0° condition of the azimuthal multiplexed set at an areal density of 100 channel bits/μm^2 in a 400μm-thick layer. 100 sequentially-recorded holograms were multiplexed co-locationally, and hologram numbers 8, 17, 32, 40, 49, 57, 72, 80, and 91 are labeled for reference in the theta scan. The noise floor of 5×10^{-8}W is extremely low, and is uniform across the entire set of co-locationally multiplexed data pages. The recording schedule was reasonably optimized and thus the diffracted power varied by less than about a factor of two across the entire sequence of recorded data pages. These results show that Aprilis media technology also has the desirable property of extremely low scatter. Thus, noise gratings that are typically common to multiplexed data page holograms, and which typically degrade SNR at high areal density, are not present. The cumulative grating strength, $\Sigma\eta^{0.5}$, attained a high value of ~20 for a recorded areal density of 150 channel bits/μm^2 in a 400μm-thick layer.[12]

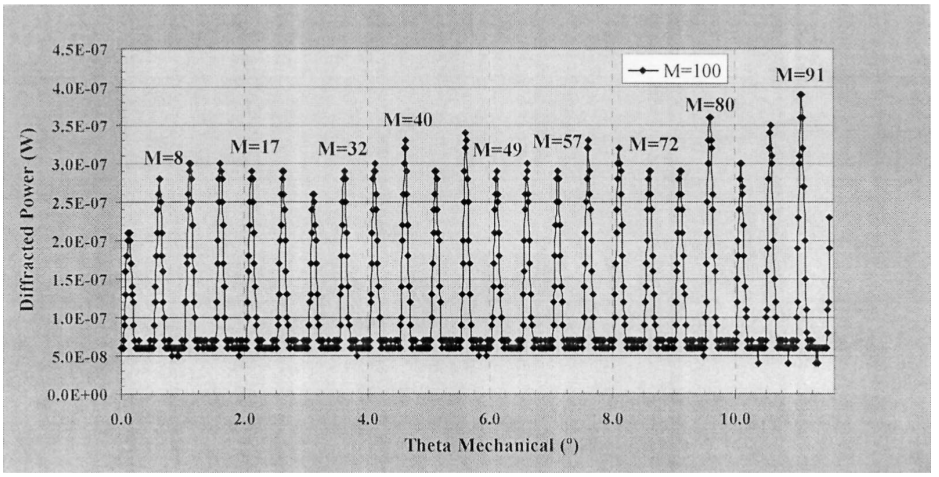

Figure 3. Scan of Bragg-selectivity profiles for co-locationally-recorded digital 512^2 data pages for angle-multiplexing values of θ_i ($\Delta\theta_i$=0.5° between holograms) at 0.01° increment per data point for holograms where the azimuthal angle is ϕ=0°.

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Commercial realization of high performance holographic data storage

The combination of extremely large storage capacities and fast transfer rates make holographic storage ideal for high-performance video applications.[1] Here, we present an overview of the InPhase Technologies holographic demonstration platforms. These compact, mobile systems are the first fully-functional, portable, holographic, recordable drives. The development of these devices paves the way for the commercialization of this technology.

The primary advantage of holographic storage comes from using the entire volume of the media, not just the surface to store information. Light from a single laser beam is split into two, the signal beam (which carries the data) and the reference. The hologram is formed where these two beams intersect in the volume of the recording medium. (In photopolymer materials, a photo-induced polymerization is responsible for the index modulation that creates the permanent hologram.) By varying the reference beam angle, wavelength, or media position, many different holograms can be recorded in the same volume of material. To read the data, a reference beam—with characteristics identical to that used to store the data—diffracts off the index modulation reconstructing the stored information. The reconstructed data page is imaged onto a detector that reads the data in parallel. This parallel recording and read out of data provides holography with its potential for very fast transfer rates.

The major challenge to implementing holographic storage has been the development of a suitable storage medium.[2] *Tapestry*™ satisfies the many stringent criteria for a viable storage material including high dynamic range, high photosensitivity, dimensional stability, optical clarity, manufacturability, nondestructive readout, thickness, and environmental and thermal stability. In addition to developing this new class of materials, InPhase Technologies also developed the *ZeroWave*™ manufacturing processes, which enable the cost-effective fabrication of high-quality optical media.

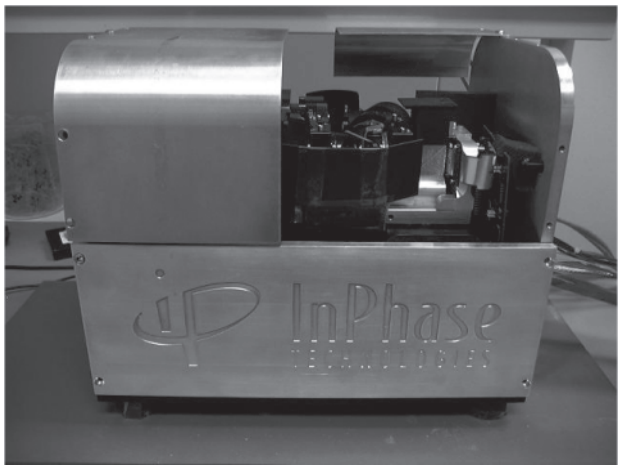

Figure 1. Angle-multiplexed demonstration platform.

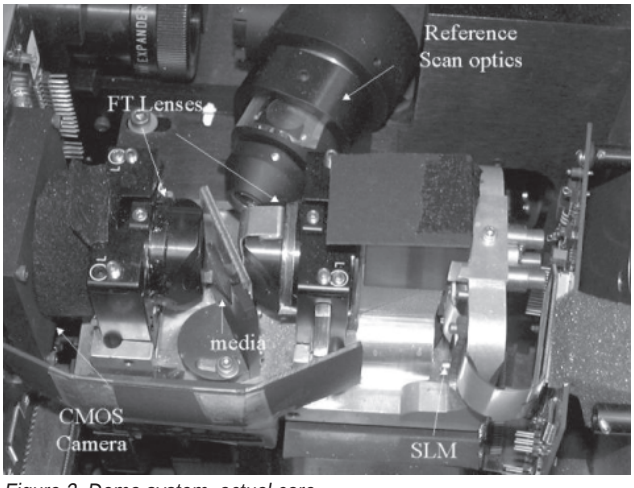

Figure 2. Demo system, actual core.

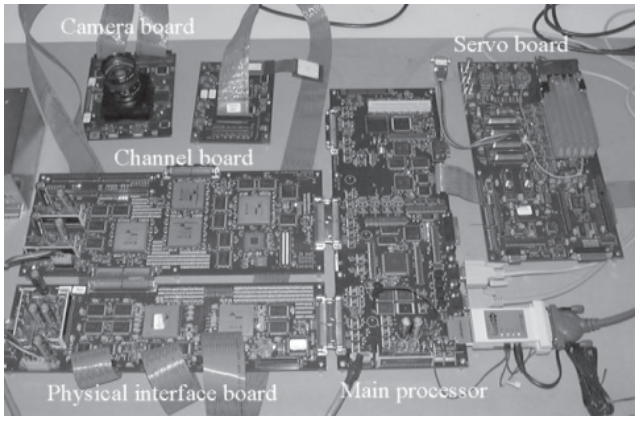

Figure 3. Electronics in FPGA form.

Demonstration platforms

The development of *Tapestry*™ storage media has enabled the design and construction of a series of first-generation holographic storage demonstration platforms (see Figure 1).[3,4] This small portable unit records and plays digital video content, and has been developed to demonstrate the feasibility and potential for commercialization of the technology. The system uses angle multiplexing to record multiple superimposed holograms in the volume of a 0.8-1.0mm thick photopolymer medium. The reference-beam angle is controlled using a Cambridge Systems precision galvo mirror, imaged by a set of scan optics onto the recording medium. Holograms are stored at 0.065˚ spacing. The system operates in a normal room environment with no active vibration isolation. Figure 2 shows the optical-mechanical assembly for that unit. This subassembly contains the modulator, the Fourier-transform optics, and the media mount. (The optical path from modulator to camera is less than 10cm.)

The input modulator is a Displaytech 1280×768 ferro-electric liquid crystal panel driven by InPhase-designed control electronics. The custom camera electronics use a Micron MV-13 CMOS APS camera, (1280×1024) that records data pages with high fidelity. Single holograms easily exhibit SNRs in the 9-10dB range, (less than 0.5% pixel saturation). The excellent optical quality of the medium makes this fidelity possible.

The electronics layout is shown in Figure 3. The main controller uses a Motorola Coldfire processor and the channel, servo, and control hardware have been implemented in FPGAs. The system has a 2MB/s data rate, (the electronics as designed should run at 20 MB/s—full ASICS).

Holographic recording performance

The demonstration system as currently designed is limited in capacity by the scan optics used. The unobstructed scan range is ~11˚, limiting hologram stacks to 90-100 recordings. For dem-

onstration purposes, this range is more than adequate. With current data overhead, more than 5Mbytes of user data can be stored in a single location. The recording performance of the system is shown in Figure 4, which shows a scan of a video file (~90 holograms.) The diffracted intensity of each data-page hologram as a function of galvo half-angle is shown. Here, the first exposures are of the order of 50ms and the final exposures are in the 75-100ms range. Note that the media used for demonstration purposes is much less sensitive (10 times or more) than our commercial media. This allows operation of the device open to ambient light so that recording and readout can be observed.

The average SNR of the data set is 6.5dB, typical for this platform. In our system, all holograms with SNR above ~2dB can be recovered error free. Figure 5 shows the raw recovered images of the first and last holograms in the data set. The hologram histograms shown are derived using the entire data page with no pre-processing. The overlap of the data distributions is exaggerated due to the intensity variations across the image mentioned earlier. In practice, we apply equalization algorithms to the recovered data page before decoding to eliminate this variance.

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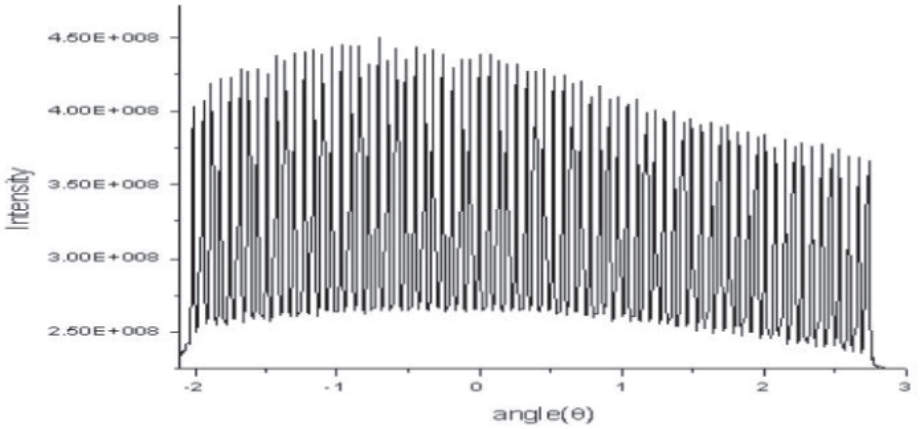

Figure 4. Angle scan of a set of ~90 holograms, (>5MB of MPEG 4 video).

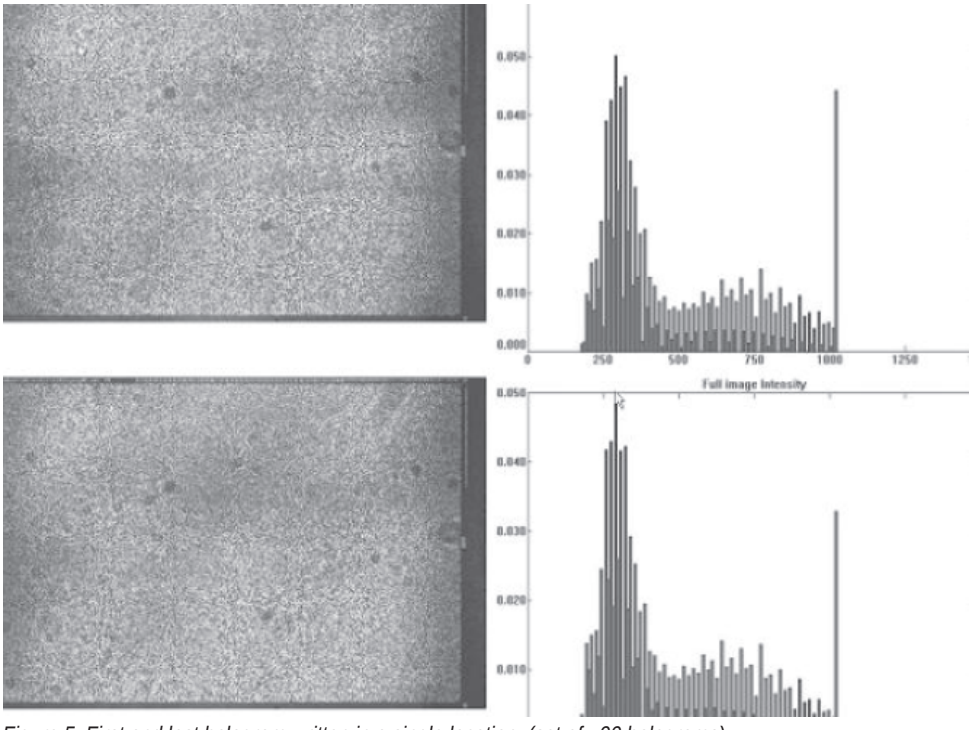

Figure 5. First and last hologram written in a single location, (set of ~90 holograms).

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Photopolymerizable hybrid sol-gel materials for holographic recording.

In the last eight years, the INTA-CSIC group has been working on the development of photopolymerizable organic-inorganic hybrid materials, prepared by the sol-gel method, for holographic applications. The sol-gel approach was first proposed for the preparation and manufacturing of such materials in 1996.[1] It was suggested as a way of obtaining a rigid final product, but avoiding the Bragg detuning that shrinkage of pure organic materials usually causes. Holographic sol-gel materials can be made through the incorporation of a monomer and a photoinitiator at the solution stage of the preparation process. Thus, the organic molecules are homogeneously embedded within the porosity of the resulting inorganic matrix (e.g. silica), which shows excellent optical quality and can be processed in different forms and sizes (thin or thick films and bulk samples). The refractive index of the hybrid material proves to be tunable through the partial modification of the binder and/or of the photosensitive organic component.

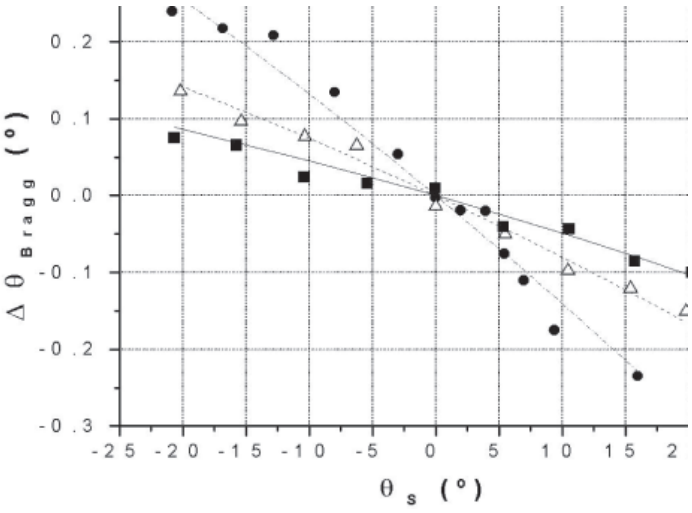

Figure 1. Bragg detuning experimental data (symbols) and best fit (lines) as a function of the slanted angle (θ_s) for 0%, 10%, and 20% TMOS samples (circles and dot-dashed line, open-up-triangles and dashed line, and squares and solid line, respectively).

In the first demonstration of the validity of uing a sol-gel material to record a Bragg grating with UV irradiation, the diffraction efficiencies were close to 100%, although with moderate index modulation (Table 1). The recorded holograms were highly stable; i.e. the diffraction efficiency decreased by just 5% over more than six months.[1,2] Since then, modifications to the chemical composition have shifted the recording wavelength (λ) into the visible and improved the overall recording properties (see Table 1).[1-4] Although photosensitivity improvement is still under way,[3,4] the sol-gel approach allows the porous silica matrix, which acts as a binder, to provide an easy way of controlling both pore diameter and size distribution. In addition, the porosity can be functionalized. This facilitates: diffusion of the monomer through the matrix during recording and the tailoring of the shrinkage during the UV post-recording process.

Recently, the performance of a sol-gel material set based on a mixture of organic monomers (acrylamide and *N*,*N*'-methylene-bis-acrylamide) and a photoinitiator complex (Rose Bengal and triethanolamine), dispersed within the porosity of different silica matrices, was reported.[5] The supporting matrixes were obtained using different mixtures of sol-gel precursors (which implies different amounts of organic groups in the porosity of the final binder). Such a binder modification provides a higher degree of crosslinking: a post-recording shrinkage evolution from 1.3% to a 0.4% of the initial material thickness was obtained with a simple matrix modification (see Figure 1). A theoretical study of how to compensate for Bragg-condition angular deviation during the UV post-recording process over a slanted-angle range of interest has revealed that it could be done by tailoring the binder shrinkage (s) and/or the maximum refractive index modulation capacity of a photosensitive mixture (Δn).

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Composition	sf (lp/mm)	d_{eff}-d (μm)	η(%)	Δn	E (mJ/cm²)	S (cm²/mJ)	λ (nm)	ϑ (°)
TMOS/IRG-651/MMA	54	2000-2500	93	$1.1\cdot10^{-4}$	3000	$3.2\cdot10^{-4}$	351	-
TMOS/RB/POEA	676	177-1000	27	$6.0\cdot10^{-4}$	840	$6.5\cdot10^{-4}$	514	0.15
GPTMS/RB/AA+BAA	676	250-350	93	$1.0\cdot10^{-3}$	300	$3.4\cdot10^{-3}$	514	0.097

Table 1. The recorded grating spatial frequency (sf), the effective and real thickness (d_{eff}, d) of the sample, the diffraction efficiency (η), the refractive index modulation (Δn), the exposure energy (E), the photosensitivity (S), the recording wavelength (λ) and the half width at half maximum of the angular selectivity curve (ϑ) are given for the different sol-gel holographic materials prepared by our group.

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Self-developing polymer compositions for holography

Photopolymer compositions for holography—PPC-488, PPC-520 and PPC-650—have been developed at the Kiev Institute of Physics, Ukraine.[1] A PPC is a multi-component liquid mixture, the main constituents of which are polymerizable compounds (oligoetheracrylates), an initiating system, and a neutral component (NC) that stabilizes the hologram. The PPCs are self-developing holographic recording materials sensitive in the 350-670nm region. The amplitude of refractive index modulation n_1 is 0.015-0.03, exposure energy ranges from 60-600mJ/cm^2 for different compositions, and the thickness of recording layers can be varied from 10-1000mm.

Volume and surface-relief transmission gratings (see Figure 1), and volume reflection gratings (see Figure 2), can be recorded in PPC media. A technique for shrinkage prevention has been proposed for recording reflection gratings. Real-time image development extends PPC application from production of various holographic optical elements to holographic interferometry and devices of correlation optics. For instance, volume transmission gratings have been used in the dispersive resonator of a single-frequency tunable dye laser, with a polymer-active element doped with generative dye.[2] The materials have also been used for recording spatial matched filters in Vander-Lugt-type correlators.[3] Thick Fourier filters, with diffraction efficiency up to 50%, were made with a correlation plane signal-to-noise ratio of 40dB.

We have shown that the phase separation of the initial mixture plays an important role in the recording process.[3] Since the polymer network has limited thermodynamic compatibility with the NC, polymer formation gives rise to its displacement from the network and to the phase separation of the initial mixture. In an interference pattern, a spatially-organized two-phase structure with a periodic distribution of phases is formed. Such a system is thermodynamically at quasi-equilibrium, and the diffusion of components between the phases is forbidden: thus a permanently stable hologram is established. The value of n_1^{max} depends on the thermodynamic compatibility of the polymer and NC, and on the difference in their refractive indices: $\Delta n_{P,NC}=|n_p-n_{NC}|$. It can be quantitatively estimated by the difference of their solubility parameters: $\Delta\delta=|\delta_P-\delta_{NC}|$. For effective holographic recording ($n^{MAX}\geq 0.01$) the values of $\Delta\delta_{P,NC}$ and $\Delta N_{P,NC}$ should satisfy the following conditions: $|\Delta N_{P,NC}|\geq 0.1$; $\Delta\delta_{P,NC}\leq 5 MPa^{1/2}$, if hydrogen bonds are not formed; or $\Delta\delta_{P,NC}\leq 10 MPa^{1/2}$, if hydrogen bonds can be formed. Results obtained can be used for the development of materials with improved holographic characteristics. Recently, we have also shown that the phase separation of PPCs can be used for the creation of polymer-nanoparticle periodic structures.

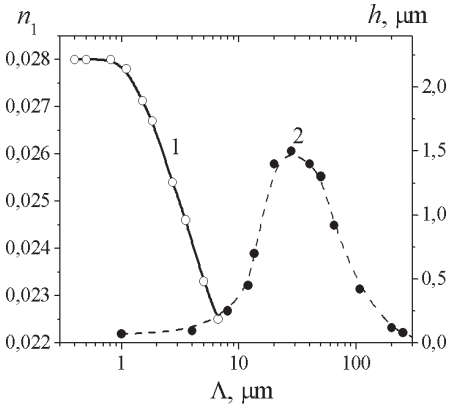

Figure 1. Variation of n_1 (1) and h (2) according to a grating period (Λ) for transmission volume (1) and relief (2) gratings in PPC-488. The wavelength of the recording beams λ_r is 488nm. The wavelength of the testing beam λ_t is 632.8nm. The relief depth is h.

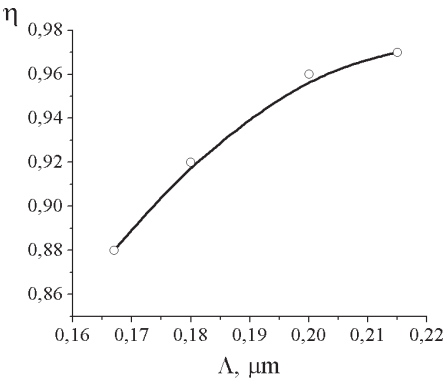

Figure 2. Dependence of n_1 on period (Λ) for reflection gratings in PPC-488. ($\lambda_r=\lambda_t=488$nm).

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High-thickness PVA/acrylamide photopolymers for data storage applications

Holographic techniques are particularly attractive for data storage systems and the fabrication of optical elements.[1] Particularly for the former application, the thick recording materials used in holography offer an advantage over currently the surface-recording techniques used for CDs and DVDs. Thanks to this volume recording, the theoretical information storage capacity of holographic memories greatly exceeds that of current devices such as the CD-ROM (by 1000 times) and random access time is significantly lower (just 10%). Photopolymers make good holographic recording materials[2] because they have: acceptable energetic sensitivity, a variable spectral sensitivity depending on the sensitizer dye used, good resolution, high diffraction efficiency, and good signal/noise ratio. Their low cost, easy preparation, and lack of complicated developing processes make them even more attractive for use in large-scale WORM- (write once read many times) type memories.

For holographic memories to be competitive, the recording layer must be 500µm or thicker.[1] More holograms may be recorded with thicker layers, because the angular Bragg selectivity increases due to the fact that the width of the angular response curve is very small.[3] It is not easy to make such thick layers with currently-available recording. Photopolymers based on acrylamide are of interest due to their mean energetic sensitivity as compared to other available materials; the possibility of easily adapting their spectral sensitivity to the type of recording laser through changing the sensitizer dye, high diffraction efficiency, and both an acceptable resolution and signal/noise ratio.[4]

In recording materials based on polyvinyl alcohol (PVA)/acrylamide (AA), a solution of PVA in water forms the matrix and this is used to prepare the mixture of monomer (AA) and photopolymerization initiator system: triethanolamine (TEA) and yellowish eosin (YE). The solution is mixed using a conventional magnetic stirrer, under red light and in standard laboratory conditions (temperature, pressure, relative humidity). The solutions are deposited, in polystyrene or polymethylmethacrylate molds, and left in the dark to allow the water to evaporate, while recording the laboratory conditions (temperature and relative humidity) during the process. When part of the water has evaporated, the 'dry' material is removed from the mold, cut into squares, and attached—without the use of adhesive—to the surface of glass plates measuring 6.5×6.5cm^2. The plates are then ready for exposure, which takes place immediately. The thickness of the material is measured using a conventional micrometer. A detailed study of this method of preparation and the factors governing the process enable a thickness of around 1000µm to be reached.[3] Additionally, optimization of the composition allows good results to be obtained for the main holographic parameters, measured to characterize the material, such as diffraction efficiency (DE), energetic sensitivity (S), or the minimum energy required to reach maximum diffraction efficiency (DE_{max}).

To evaluate the material capacity for the recording of many diffraction gratings, we recorded seven in the same zone of a 700µm-thick recording material by using the light from an Argon laser emitting at 514nm. We used angular multiplexing, changing the angle of the reference beam in steps of 0.5°. In this way, a 4.5° variation in the angle suffices to include the seven gratings. Figure 1 shows the diffraction efficiency (DE) as a function of the reconstruction angle for the multiplexed gratings stored in the PVA/acrylamide photopolymer. We obtained high values for each individual maximum DE, sufficient separation between the gratings to enable their independent reconstruction, and good dynamic range (M#=25.91).[1]

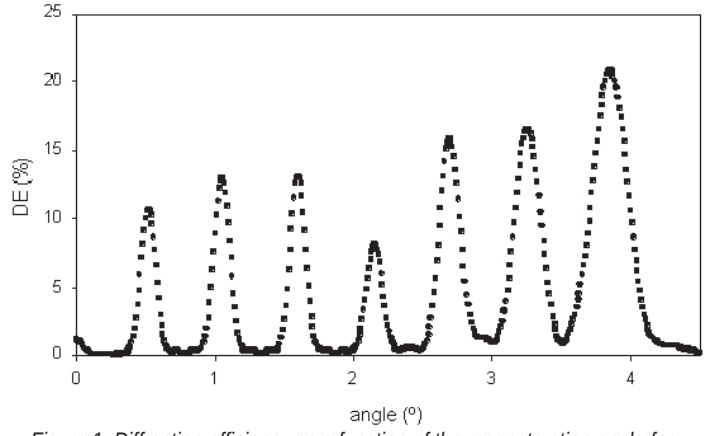

Figure 1. Diffraction efficiency as a function of the reconstruction angle for multiplexed gratings recorded in a PVA/acrylamide photopolymer.

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Organically-modified silica glasses for holographic recording

The fundamental motivation for using sol-gel materials is to replace high-temperature glass and ceramic fabrication techniques by a process that can takes place at lower, even close to room, temperatures. This way it is possible to incorporate organic molecules with low thermal stability into inorganic matrices, resulting in hybrid organic-inorganic materials, also called ormosils (organically modified silicates) or ormocers (organically modified ceramics). Combining the properties of organic and inorganic components in composites opens new opportunities in the development of innovative materials, including those for holographic data storage.

In 1996, the first organically-modified sol-gel material suitable for recoding of volume holograms was demonstrated.[1] This material resulted from an effort to overcome the problems of limited maximum thickness of commercial holographic photopolymers, as well as with the material shrinkage upon polymerization typical of acrylic-based materials. The basic idea here is to disperse organic photopolymerizable species in an inorganic host matrix rather than in an organic binder typically used for this purpose: the inorganic host matrix should significantly improve physical properties of the holographic recording material, such as its rigidity, environmental stability, dimensional changes upon holographic exposure, maximum achievable thickness, and the ability to accept an optical-grade polish. The support matrix of this organic-inorganic material, in contrast to the Vycor glass holographic materials,[2] was formed by in-situ polymerization (sol-gel reaction) of liquid silica precursors in the presence of dissolved photo-initiating and photo-polymerizable species: Irgacure 651 sensitizer and methyl methacrylate (MMA) monomer. The material was prepared in the form of monoliths, a few millimeters thick, and volume gratings were holographicaly recorded with diffraction efficiencies greater than 90%.

Sol-gel also allows the mixing of inorganic and organic components at the nanometer scale, in virtually any ratio. The small domain sizes of organic species minimizes optical scattering even when the refractive indices of the dopants differ from that of the host matrix. Among other advantages, we are particularly concerned with the following: decoupling the host matrix formation chemistry (sol-gel) from the hologram formation chemistry (photopolymerization); efficient liquid monomer transport facilitated by a porous host; reduced matrix contraction in the areas of low illumination; and the possibility of combining a porous, low-refractive-index host with high-refractive-index monomers to maximize the index modulation.

Using this strategy, a new sol-gel glass[3] was developed with refractive index modulation of up to 0.0043, a diffraction efficiency of 98%, and exposure of 230mJcm^{-2} at 514.5nm. To our knowledge, these are the highest refractive index modulation and sensitivity reported to date in a photopolymerizable material of similar thickness. Our material consists of a glassy host containing an ethylenic unsaturated monomer ethylene glycol phenyl ether acrylate, and a free-radical-generating titanocene photoinititiator bis(μ^5-2,4-cyclopentadien-1-yl)-bis-[2,6-difluoro-3-1H-pyrrol-1-yl)phenyl]titanium. It was fabricated both as thick films and monoliths. These encouraging results imply that sol-gel holographic materials are an important candidate for data storage applications.[4]

Recently, a photorefractive grating with refractive index modulation of 0.002 and a two-beam coupling gain of 444cm^{-1} was demonstrated in an organically-modified permanently-poled sol-gel glass.[5] The azo-dye 2,5-dimethyl-4-(2-hydroxyethoxy)-4'-nitroazobenzene (DMNPAA) was used as a nonlinear-optical chromophore. The chromophore molecules were covalently bonded to the silica glass backbone in order to achieve the high dye concentration required for efficient nonlinear-optical properties, while avoiding the dye crystallization that is typically observed in guest-host photorefractive polymers. We used 2,4,7-trinitro-9-fluorenone (TNF) as a photosensitizer and N-ethylcarbazole (ECZ) as the charge-transporting agent: both being present as guests in the glass, i.e. without being covalently attached to the matrix. Excellent resistance against chromophore crystallization is achieved by covalently bonding the chromophore. High stability of electric-field-induced chromophore alignment is due to a gradual heat-induced densification of the initially low-T_g gel during the electric field poling, yielding a high-T_g hard glass film. This densification process is essential for slowing down diffusive randomization of the chromophore alignment, and for improving the mechanical, electrical, and thermal properties of permanently-poled photorefractive glasses.

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