Recording media that exhibit high dynamic range for digital holographic data storage

Lisa Dhar, Arturo Hale, Howard E. Katz, Marcia L. Schilling, Melinda G. Schnoes, and Fred C. Schilling

Bell Laboratories, Lucent Technologies, 700 Mountain Avenue, Murray Hill, New Jersey 07974

Received December 15, 1998

A general strategy for fabricating thick, optically flat photopolymer recording media with high dynamic range ($M/\#$) that exhibit low levels of recording-induced Bragg detuning for holographic data storage is presented. In particular, media with $M/\#$ values as high as 42 in 1-mm-thick formats are obtained. We believe that these results are the first demonstration of a holographic storage medium with a dynamic range of this magnitude. In addition, we report the holographic recording and recovery of high-capacity (480-kbit) digital data pages in these media, further illustrating their data-storage capabilities. © 1999 Optical Society of America

OCIS codes: 090.2900, 160.5470, 210.2860, 090.7330, 160.4890.

The lack of suitable recording materials has long been an obstacle to the development of holographic data storage. Holographic media must satisfy stringent criteria, including high dynamic range, dimensional stability, optical clarity and flatness, and millimeter thickness.[1] In this Letter we outline a general strategy for the design of photopolymer recording media that has made possible substantial advances toward meeting these goals. In particular, we demonstrate how this approach yields recording media that exhibit an $M/\#$ (Ref. 2) of 42, to our knowledge the highest dynamic range yet reported, in an ~1-mm-thick, optically flat format with low light scattering. We also report holographic storage and recovery of multiple high-capacity digital data pages in thick media fabricated by use of this method.

Optical holography promises storage densities and data-transfer rates that far exceed those of traditional magnetic and optical recording technologies.[3] Holographic storage can achieve high transfer rates because data are written and recovered in a pagewise fashion; high storage densities are possible because many high-capacity data pages can be recorded independently in the same volume of material. To enable one to achieve simultaneously high storage densities and rapid data-recovery rates, a material must have an adequate dynamic range to support large numbers of holograms with sufficiently high diffraction efficiencies. The storage capacity of a holographic material is typically characterized by parameter $M/\#$,[2] a measure of how many holograms of a given average diffraction efficiency can be stored. The $M/\#$ of iron-doped lithium niobate, the traditional choice for holography experiments, is typically 1–1.5 for 1-cm-thick crystals.[2] It is commonly believed that $M/\#$ values that are at least an order of magnitude higher will be required for compelling storage densities and transfer rates.[1] The thickness of the recording medium also independently determines how many holograms can be recorded in the same volume of material because of geometrical and angular bandwidth considerations.[4] Thicknesses of approximately 1 mm are thought to be required for high-density applications. Recording media must undergo only limited changes in their dimensions and bulk refractive index, as these changes lead to Bragg detuning or rotations in the Bragg angles of recorded holograms. Bragg detuning can degrade the fidelity of data recovery and ultimately limit the storage density of a material. Holographic media must also be optically flat so that high-capacity digital data pages can be imaged, recorded, and recovered with low probabilities of error.

Photopolymer materials[5] are attractive candidates for write-once–read-many-times data-storage applications because they can be designed to have large refractive-index contrast and high photosensitivity, record permanent holograms, and be easily processed. Most of the currently available holographic photopolymers,[5] however, have been optimized for display rather than storage applications. Typically, these materials can be used only as thin ($\leq$200-μm) layers and often exhibit significant dimensional and bulk refractive-index changes owing to the polymerization of the photosensitive species that occurs during recording.

To better meet the needs of holographic data storage we designed a new type of polymer system composed of two independently polymerizable and compatible chemical systems: low-refractive-index matrix precursors and high-refractive-index photopolymerizable monomers. The matrices of our media are formed by *in situ* polymerization, which yields a cross-linked network in the presence of the photopolymerizable monomers, which remain dissolved and unreacted. Holographic exposure polymerizes the photoactive monomer(s); the unreacted monomers diffuse through the matrix, producing a modulation in the refractive index that is determined by the difference between the refractive indices of the monomer and the matrix and by the relative volume fraction of the monomer. The most important aspects of this strategy that yield high-performance holographic storage media are (i) preforming the matrix *in situ*, which allows media to be shaped into the required thick and flat formats; (ii) creation of a cross-linked matrix as a support structure for stable holographic gratings; (iii) choice of compatible matrix and monomer systems that yield media with good optical clarity and low levels of light scattering; and (iv) design of independent matrix and monomer systems to allow us to avoid cross reactions that dilute the refractive-index contrast.

0146-9592/99/070487-03$15.00/0 © 1999 Optical Society of America

Point (iv) ensures that the low refractive index of the background matrix is not detrimentally raised by the premature polymerization of the high-refractive-index monomer. Media with high $M/\#$ can be fabricated by use of small amounts of the high-index monomer, thereby minimizing the photopolymerization-induced dimensional and bulk refractive-index changes that occur during holographic recording.

To prepare samples for holographic recording we dispensed resins, consisting of the matrix precursors, photopolymerizable monomers, and a visible light-sensitive photoinitiator, between two optically flat glass slides. The *in situ* room-temperature formation of the matrix allowed routine fabrication of high-optical-quality media with polymer thicknesses of 200 μm to 1.5 mm. A transmission interferogram of a typical 1-mm-thick (polymer thickness) medium is shown in Fig. 1(a), and a surface plot of the variation in the optical flatness is shown in Fig. 1(b); the data show flatness within 50 nm ($\lambda/10$) over the diameter of the sample.

The strength of separable chemistries is highlighted by the ability to controllably optimize the dynamic range of the medium. In general, the dynamic range of a photopolymer material increases with increasing concentration of the writing monomer. This approach, however, carries the penalty of raising the Bragg detuning levels of the medium. For data-storage applications the detuning levels should be kept below the width that is typical of Bragg peaks obtained in thick materials. Our materials-design strategy enables us to improve the dynamic range of the medium while keeping the Bragg detuning levels constant by simply varying the difference between the refractive indices of the matrix and the writing monomers.

In Fig. 2(a) we show the $M/\#$ values of a series of 200-μm-thick media that were fabricated by use of the same matrix but with writing monomers of various refractive indices and sizes, where the concentrations of the monomers were adjusted to yield equal levels of Bragg detuning. Thirty-five plane-wave holograms were angle multiplexed within the samples by use of a frequency-doubled Nd:YAG laser (532 nm) as the recording source. We adjusted the recording exposure schedules[6] to consume all the photoactive species and yield holograms of nearly equal diffraction efficiency. After recording, we flooded exposed the samples to bleach any excess photoinitiator. We calculated the $M/\#$ of the media by summing the square roots of the diffraction efficiencies of the recorded holograms. The levels of Bragg rotation[7] for this comparison were chosen to be less than the angular separation between the first nulls of a Bragg peak of a 500-μm-thick medium. Each of the samples required rotations of $-0.13°$ and $0.19°$ for the two recording arms of the holographic system for recovery of the Bragg angle of a plane-wave grating written with an internal tilt angle of 17.3°; these rotations corresponded to changes of $\sim 0.35\%$ in thickness and $\sim 2.1 \times 10^{-3}$ in the bulk refractive index for the complete reaction of the photoactive monomers. Increases in $M/\#$ from 2 to 11 were realized while the same level of effective dimensional stability of the media was maintained.

Media with high $M/\#$ were obtained by fabrication of thick samples of our photopolymer materials. (We measured the $M/\#$ values in these experiments

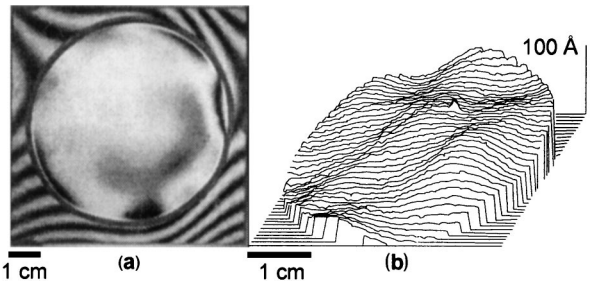

Fig. 1. (a) Transmission interferogram of a typical 1-mm-thick photopolymer medium. (b) Surface plot of the variation in the optical thickness of the inner 4 cm of the sample. The optical flatness of the sample varies less than $(\lambda/10)$/cm ($\sim$50 nm; 1 Å = 0.1 nm) over the entire area of the medium.

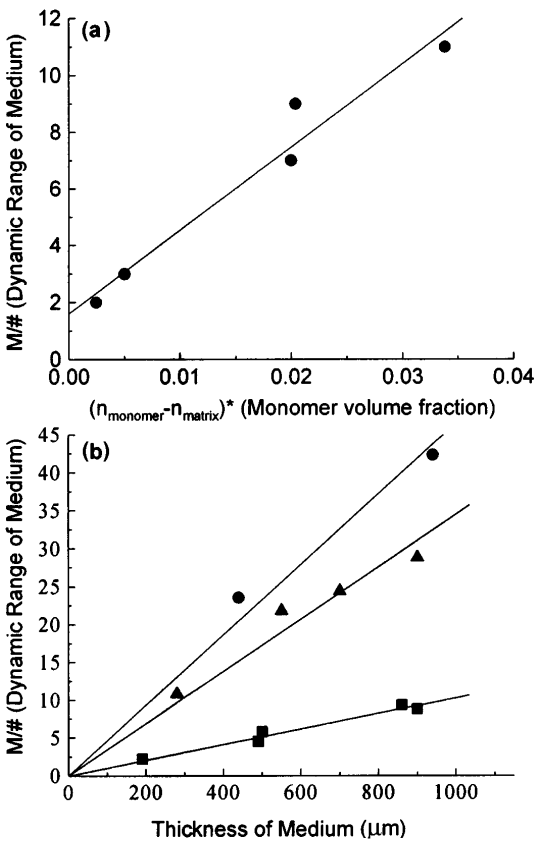

Fig. 2. (a) $M/\#$ versus the product of the difference between refractive indices of the photosensitive monomer and the matrix and the volume fraction of the monomer for five different resins. Adjusting the molar number of reactive groups of the photoactive monomer in each of the media yields equivalent amounts of recording-induced rotations in the Bragg angle upon recovery. The media are all 200 μm thick. (b) $M/\#$ versus thickness for photopolymer media fabricated with a typical writing monomer: squares, media exhibiting Bragg angle rotations of $-0.03°$ and $0.06°$ at an internal grating tilt angle of 17.3°; triangles, media exhibiting Bragg angle rotations of $-0.13°$ and $0.19°$; circles, media exhibiting Bragg angle rotations of $-0.16°$ and $0.25°$.

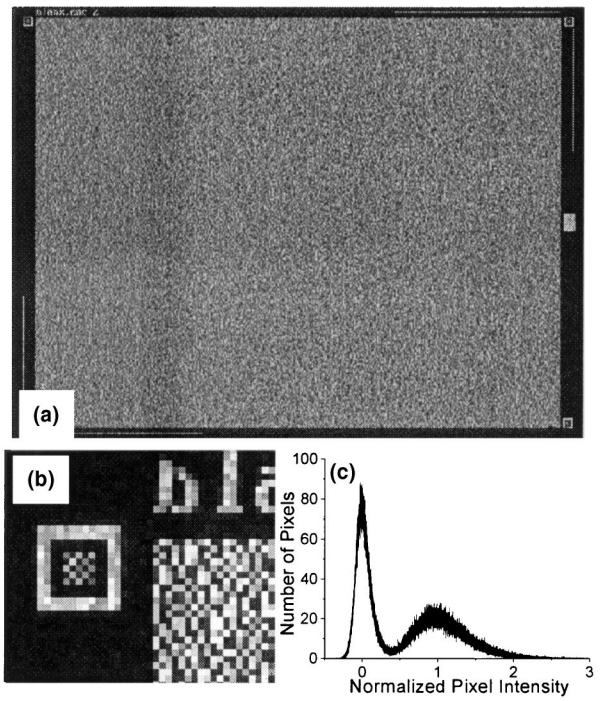

Fig. 3. (a) Tenth digital data page recovered from a set of 20 holograms multiplexed in a 500-μm-thick medium. The data page in an 800 $\times$ 600 array of random transmitting and opaque pixels. The probability of detecting errors in this data page is 3×10^{-4}, as determined by a global threshold method. (b) Expanded view of the top left-hand corner of the recovered data page, illustrating the fidelity of the recovery. (c) Histogram of the intensities of the pixels of the entire recovered data page.

by recording >250 holograms, using angle and peristrophic multiplexing methods.[8]) In Fig. 2(b) we show how $M/\#$ scales with thickness in media fabricated with a typical writing monomer. Data from three sets of samples are shown, with each set formulated with a different concentration of the monomer and therefore exhibiting a different level of Bragg detuning. The gains in $M/\#$ with increasing thickness are possible because of the low levels of light absorption and light scatter and the high level of effective dimensional stability of the media.

We believe that the observation of an $M/\#$ of 42 is the first demonstration of a holographic storage medium with a dynamic range of this magnitude. Moreover, our strategy allows us flexibility in further optimization of our material. For example, media with high $M/\#$ values yet the same effective dimensional stability can be fabricated by use of monomers with increased refractive index or monomers with higher molecular weights that would occupy larger volume fractions per reactive group.

As a preliminary demonstration of the capabilities of these media, one of the moderate-response versions was used to store and recover multiple high-capacity (480-kbit) digital data pages. Figure 3 shows the tenth recovered data page from a set of 20 holograms multiplexed in a 500-μm-thick medium. The probability of detecting errors[9] in this data page was well below the level required for error-free recovery.[10] Higher storage densities (~48 channel bits/μm^2, or ~31.5 channel Gbits/in.^{2}) were demonstrated by use of established multiplexing techniques[11] to record and recover 3000 digital data pages in a 750-μm-thick medium.[12] The data suggest that even-greater storage densities are possible by use of available higher-response versions of our materials.

The materials described here represent substantial advances in the development of recording media for holographic data storage. Their success, however, rests upon a few important issues. Ongoing work is focused on the long-term archival life of stored data and on the effect of changes in temperature of the polymer media[7] on the fidelity of data readout. Nonetheless, the unprecedented $M/\#$ values reported here, the demonstrated data-storage capabilities of the systems, and the ability to improve the material further greatly enhance the promise of photopolymer media for holographic data storage.

We thank C. Boyd for sample fabrication and H. Bair, L. Blyler, M. J. Cardillo, E. A. Chandross, S. Campbell, K. Curtis, M. Galvin-Donoghue, A. L. Harris, A. Hill, N. Levinos, S. Patel, S. Popielarski, X. Quan, E. Reichmanis, M. Tackitt, W. Wilson, and P. Wiltzius for their many contributions. The National Media Laboratory is also thanked for support of this research. Correspondence and requests for materials should be addressed to L. Dhar (e-mail: ldhar@lucent.com).

References

1. G. T. Sincerbox, Opt. Mater. **4,** 370 (1995).
2. F. H. Mok, G. W. Burr, and D. Psaltis, Opt. Lett. **21,** 896 (1996); A. Pu, K. Curtis, and D. Psaltis, Opt. Eng. **35,** 2824 (1996).
3. D. Psaltis and F. Mok, Sci. Am. **273,** 52 (1995); J. F. Heanue, M. C. Bashaw, and L. Hesselink, Science **265,** 749 (1994); H. Coufal, Nature (London) **393,** 628 (1998).
4. H.-Y. S. Li and D. Psaltis, Appl. Opt. **33,** 3764 (1994).
5. H. J. Bjelkagen, ed., *Selected Papers on Holographic Recording*, Vol. 130 of SPIE Milestone Series (Society of Photo-Optical Instrumentation Engineers, Bellingham, Wash., 1996).
6. M. Schilling, V. Colvin, L. Dhar, A. Harris, F. Schilling, H. Katz, T. Wysocki, A. Hale, L. Blyler, and C. Boyd, Chem. Mater. **11,** 247 (1999).
7. L. Dhar, M. Schnoes, T. Wysocki, H. Bair, M. Schilling, and C. Boyd, Appl. Phys. Lett. **73,** 1337 (1998).
8. K. Curtis, A. Pu, and D. Psaltis, Opt. Lett. **19,** 993 (1994).
9. R. M. Shelby, J. A. Hoffnagle, G. W. Burr, C. M. Jefferson, M.-P. Bernal, H. Coufal, R. K. Grygier, H. Gunther, R. M. Macfarlane, and G. T. Sincerbox, Opt. Lett. **22,** 1509 (1997).
10. L. Dhar, K. Curtis, M. Tackitt, M. Schilling, S. Campbell, W. Wilson, A. Hill, C. Boyd, N. Levinos, and A. Harris, Opt. Lett. **23,** 1710 (1998).
11. D. Psaltis, M. Levene, A. Pu, G. Barbastathis, and K. Curtis, Opt. Lett. **20,** 782 (1995).
12. K. Curtis, W. Wilson, M. Tackitt, A. Hill, and T. Richardson, paper presented at the International Conference on Photonics, Taipei, Taiwan, December, 14–18, 1998.