Use of two-photon excitation for erasable–rewritable three-dimensional bit optical data storage in a photorefractive polymer

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We report what is believed to be the first use of a photorefractive polymer in erasable–rewritable three-dimensional bit optical data storage under two-photon excitation. We successfully demonstrate writing, erasing, and rewriting of multilayered information in a photorefractive polymer consisting of 2,5-dimethyl-4-(*p*-nitrophenylazo)anisole, 2,4,7-trinitro-9-fluorenone, 9-ethylcarbazole, and poly(*N*-vinylcarbazole). A three-dimensional bit density of 5 Gbits/cm^3 is achieved by two-photon absorption under pulsed beam illumination at an infrared wavelength of 800 nm in the recording process. Complete erasing of the recording information is achieved by use of ultraviolet illumination. © 1999 Optical Society of America

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Multilayered [or three-dimensional (3D)] optical memories have increasingly become a field of interest in the development of high-density optical data storage devices.[1–5] Systems that utilize multiple-layer recording can achieve a recording density from 100 to 10,000 times higher than that in conventional optical data storage devices.

The use of two-photon excitation in 3D bit optical data storage has grown, owing to its ability to increase the density of data in a given material by reducing the volume of recorded bits.[1–5] The probability of two-photon excitation is proportional to the squared intensity of the incident light; this effect produces excitation only within a small region of the focus spot of the recording objective. As a result there is less cross talk between neighboring data layers. Another advantage of two-photon excitation is the use of infrared illumination, which results in the reduction of scattering and permits the recording of layers at a deep depth in a thick material.

Over the years various materials have been used for 3D bit data storage under two-photon excitation. In studies of two-photon 3D bit recording in photopolymerizable[2] and photobleaching[4,5] materials it has been demonstrated that recording densities could reach terabits per cubic centimeter; unfortunately neither material is erasable. Photochromic materials[1,6] that undergo a change in isomer state and photochromic polymers[7] that produce a change in refractive index when they undergo two-photon excitation are both erasable materials. Another type of material that is of considerable interest is photorefractive material. The photorefractive effect, which is induced by the spatial distribution of electric charges when the material is illuminated,[8] results in modulation of the refractive index at the point of focus. One can reverse such a process by illuminating the medium again with a specific wavelength to produce a uniform redistribution of the electric charges, leading to erasing of the recorded information. A photorefractive material that has been used for erasable two-photon 3D bit data storage is photorefractive crystal[3] (e.g., $LiNbO_3$); however, these materials are expensive and difficult to manufacture.

In this Letter we propose using a photorefractive polymer[9] as a recording material for erasable–rewritable 3D bit optical data storage under two-photon excitation. The material that we used was the polymer poly(*N*-vinylcarbazole) (PVK) doped with the photosensitive material 2,4,7-trinitro-9-fluorenone (TNF), which provides absorption in the ultraviolet-to-visible region of the spectrum. The chromophore that was used was 2,5-dimethyl-4-(*p*-nitrophenylazo)anisole (DMNPAA), which also provides absorption and an electro-optic effect in the ultraviolet-to-visible region. In this experiment, unlike in the experiments reported in Ref. 9, the randomly oriented chromophores were not aligned by application of an electric field (poling) during the polymerization and recording processes. Such a poling electric field is not necessary because the local electric field in the focus that is produced by a high-numerical-aperture objective is 5 orders of magnitude larger than that of the incident beam over the objective aperture. This local electric field is strong enough to induce a detectable electro-optic effect. Finally, we added *N*-ethylcarbazole (ECZ) to reduce the glass-transition temperature of the material. The concentration of the materials DMNPAA:PVK:ECZ:TNF was 50:33:16:1 wt. %. We fabricated uniform films of 200-μm thickness by combining all the materials

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in a Teflon cast and then polymerizing the PVK at a temperature of 100 °C. We cut and polished the resulting polymer block to produce the sample that was used in the experiments. Figure 1 illustrates the absorption band of a 200-μm-thick sample, as detected by an Oriel ultraviolet-to-visible spectrophotometer that used a xenon arc-lamp source.

It can be seen from Fig. 1 that the absorption band of the new material is within 380–600 nm. Therefore a laser beam of an infrared wavelength at 800 nm can be used in the recording process to produce two-photon excitation at 400 nm. Since the absorption band cuts off at a wavelength of ~630 nm, a range of wavelengths from 630 to 750 nm can be chosen so that the recorded photorefractive data bits are read out without suffering significantly from single- or two-photon excitation.

The optical system for two-photon excitation in the recording process is illustrated in Fig. 2. A Spectra-Physics Tsunami Ti:sapphire laser was focused into a photorefractive polymer sample. In mode-locking operation the laser can produce an ultrashort pulsed beam with a pulse width of 80 fs and an average power of 800 mW. The wavelength of the laser was tuned to 800 nm, which corresponds to approximately twice the wavelength of the main absorption band of the sample. From the absorption curve in Fig. 1, one can see that there is no single-photon absorption at a wavelength of 800 nm; only two-photon absorption can occur at a wavelength of 400 nm. The logic state of the recorded information was controlled by a mechanical shutter linked to a computer. The recording material was mounted upon a Melles-Griot computer-controlled 3D translation stage. For recording, a Zeiss Fluar objective with a numerical aperture of 0.75 and a magnification factor of 20 was used.

For reading the change in the refractive index that was caused by the two-photon photorefractive effect we employed an Olympus FluoView microscope and used it in a differential interference contrast mode. A He–Ne laser of wavelength 632.8 nm was coupled into the microscope for reading of the recorded information, as the wavelength of 632.8 nm is on the edge of the absorption band and causes minimal damage to the recorded information (see Fig. 1). To erase the recorded information we illuminated the region of interest with the ultraviolet line of the mercury lamp in the microscope. In both the reading and the erasing processes an Olympus UPlanAPO objective with a numerical aperture of 0.7 and a magnification factor of 20 was used.

As a demonstration of the ability to record information as a change in refractive index by use of two-photon excitation, in Figs. 3(a) and 3(b) we show the change in refractive index that was produced at the focus spot. In recording, an average power of 5 mW at the focus spot was used. A pattern (the letter A) consisting of 24 × 24 bits was recorded in the sample. The bit separation was 3.2 μm, and the exposure time for each bit was 20 ms. In the reading process, scanning a laser beam with power of less than 5 mW across the sample produced the differential interference contrast image. Figure 3(b) shows the deterioration of the recorded information after it was scanned 1000 times. The contrast of the recorded bit in Fig. 3(b) is 50% of that in Fig. 3(a). This result illustrates that there is weak erasure of the information, owing to the absorption of the light that is used for reading.

Figure 4 shows the writing, erasing, and rewriting of information into the same region of the photorefractive polymer. Figure 4(b) shows the same region as in

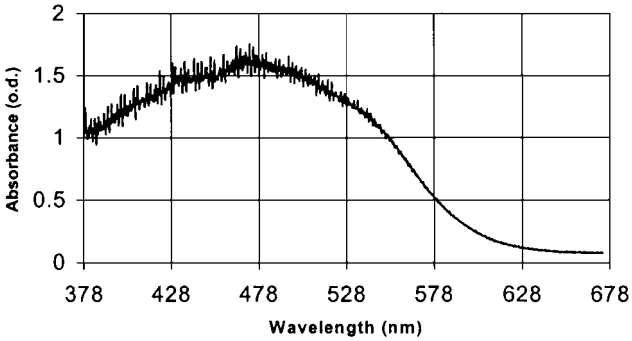

Fig. 1. Absorption curve of the photorefractive polymer.

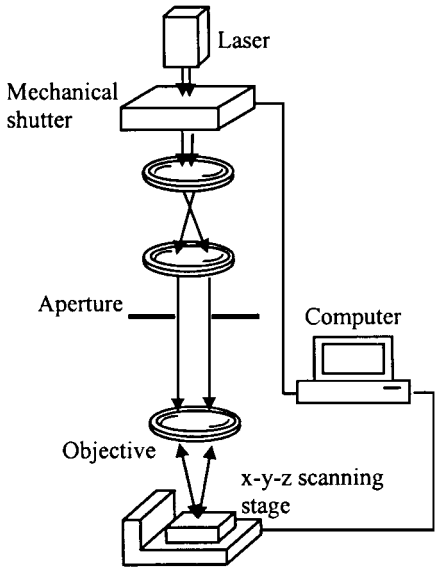

Fig. 2. Schematic diagram of the two-photon excitation microscope used to record 3D data bits in a photorefractive polymer.

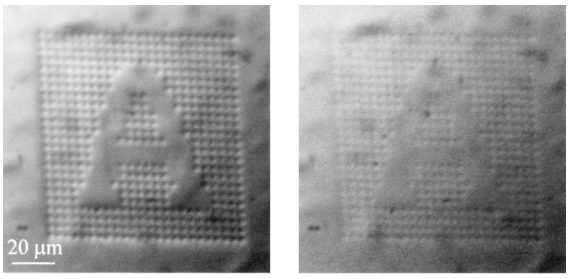

Fig. 3. Images of 24 × 24 bit patterns recorded by two-photon excitation in a photorefractive polymer: (a) letter A after the first reading, (b) letter A after it is read 1000 times.

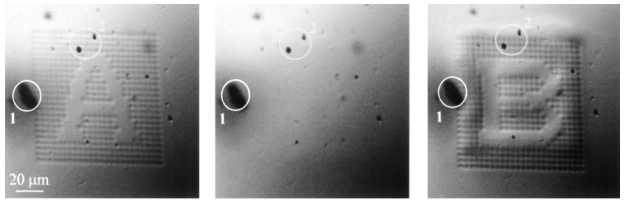

Fig. 4. Demonstration of writing, erasing, and rewriting in the same area: (a) letter A is recorded, (b) letter A is erased after being exposed to ultraviolet illumination for 1–2 s, and (c) letter B is recorded in the same area. The marked artifacts 1 and 2 indicate that the images are in the same area.

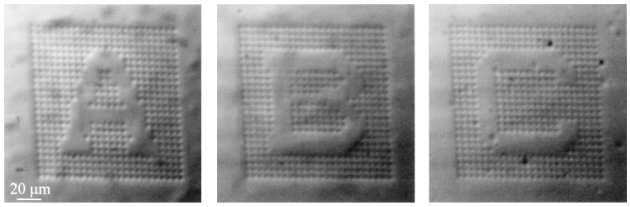

Fig. 5. Recorded 24 × 24 bit patterns at different depths in the photorefractive polymer under two-photon excitation. The spacing between adjacent layers is 20 μm, and the bit separation is 3.2 μm. (a) first layer, including the letter A; (b) second layer, including the letter B; (c) third layer, including the letter C.

Fig. 4(a), including a recorded pattern (the letter A) after it is exposed to ultraviolet illumination for 1–2 s. The result shows complete erasure of the previously recorded information. In Fig. 4(c), a new pattern (the letter B) is written into the same area shown in Figs. 4(a) and 4(b). In Fig. 4 the two artifacts, labeled 1 and 2, show that the same area was used to rewrite information.

In Fig. 5 we demonstrate the ability to record and read changes in the refractive index of multiple layers of information. Three layers of information were recorded, with a layer separation of 20 μm. Each layer consists of a pattern of 24 × 24 bits. The first layer, recorded near the surface, contains a pattern of the letter A. The second and third layers consist of the letters B and C, respectively. Thus the achieved 3D data density is approximately 5 Gbits/cm^3, which is comparable with that achieved in the photochromic polymer,[7] the photorefractive crystal,[3] and the photobleaching materials.[5,10]

The small amount of cross talk between the data layers is due to spherical aberration resulting from the refractive-index mismatch between the recording material ($n = 1.75$) and the immersion medium ($n = 1$).[5] The refractive-index mismatch results in a distortion of the diffraction spot of a recording objective at a deep position of a volume recording medium. As a result, a series of axial side peaks occurs along the depth of the recording medium,[5] resulting in cross talk between neighboring data layers. This problem can be overcome, and hence the density can be increased, by use of a variable-tube-length method.[5] Modifying the absorption band of the recording material will lead to less deterioration of the recorded signal during the reading process.

In conclusion, we have demonstrated, for what is believed to be the first time, that erasable–rewritable multiple-layer recording in a photorefractive polymer by use of two-photon excitation is possible. Such erasable–rewritable 3D bit data storage in the proposed photorefractive polymer can be achieved by use of a 1-W cw beam,[10] because the photorefractive polymer can be excited by an average power of 5 mW under pulsed beam illumination.

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