Rewritable polarization-encoded multilayer data storage in 2,5-dimethyl-4-(*p*-nitrophenylazo)anisole doped polymer

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We report a rewritable polarization-encoded multilayer data storage method with a polymer film doped with the azo dye DMNPAA (2,5-dimethyl-4-(*p*-nitrophenylazo)anisole). It is found that under two-photon excitation by a linearly polarized femtosecond laser beam at wavelength 780 nm the optical axis of DMNPAA molecules can be oriented to the perpendicular direction of the beam via a trans–cis–trans isomerization process. As a result, multilayer polarization-encoded optical data storage is demonstrated by recording two letters of a bit spacing of 4 μm in the same region of a given layer. It is shown that erasing and rewriting a particular layer is possible.

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Because of its highly localized excitation nature, two-photon (2P) excitation by a laser beam focused through a high-numerical-aperture (NA) objective allows a higher storage density compared with single-photon excitation.[1] Furthermore, infrared illumination permits recording into the inner volume of a thick recording medium. In the past decade, 2P excitation has been widely used in three-dimensional (3D) bit optical data storage. 2P-induced multilayer optical recording has been successfully demonstrated in various erasable and nonerasable materials.[2–8] In the diffraction limited case, it can be estimated that the highest 3D data density under 2P excitation by a laser beam of wavelength 800 nm is approximately a few terabits per cubic centimeter.[9]

Another method for increasing the optical data storage density is to encode the data information in the polarization domain of a recording beam.[10–12] There have been a few reports on this so-called polarization-encoded bit data storage method. Three letters were multiplexed on the surface of a polarization-sensitive polymer under single-photon excitation.[10] Although 2P excitation has been recently adopted for inducing polarization anisotropy in a bacteriorhodopsin-doped film[11] and molecular orientation in a Disperse Red One contained film,[12] the recording was permanent and no recording of multiple layers has been demonstrated. In this Letter, for the first time to our best knowledge, we report a rewritable 2P-induced polarization-encoded multilayer data storage method in a polystyrene (PS) film doped with azo dye DMNPAA (2,5-dimethyl-4-(*p*-nitrophenylazo)anisole).

DMNPAA has been used as dye chromophores in photorefractive polymer.[4,13] In fact, just like the other azo dye molecules, its optical axis can be orientated to the perpendicular direction of a linearly polarized excitation beam via a trans–cis–trans photoisomerization process.[12] It has been demonstrated that the DMNPAA-doped photorefractive polymer can be excited under 2P absorption.[5] It is therefore possible to achieve a 2P-induced polarization sensitivity via the trans–cis–trans isomerization process if DMNPAA is doped in an appropriate polymer matrix. To this end, we doped DMNPAA in unplasticized PS rather than the conventional system of poly(*n*-vinylcarbazole) (PVK) plasticized with ethyl carbazole (ECZ), as neither charge transport nor plasticization are needed in this process. The sample was prepared in the following way. A mixture consisting of 30 wt. % of DMNPAA and 70 wt. % of PS was dissolved in chloroform. Then the solution was evaporated at 60°C for 20 min to get rid of solvents. The sample was placed on a glass slide between spacers and heated at 200°C for 3 min. The polymer sample was sandwiched and cooled to room temperature quickly. Since there is no linear absorption above 650 nm (see Fig. 1) a femtosecond pulsed laser beam at wavelength 780 nm can be used for 2P excitation, where the 2P absorption

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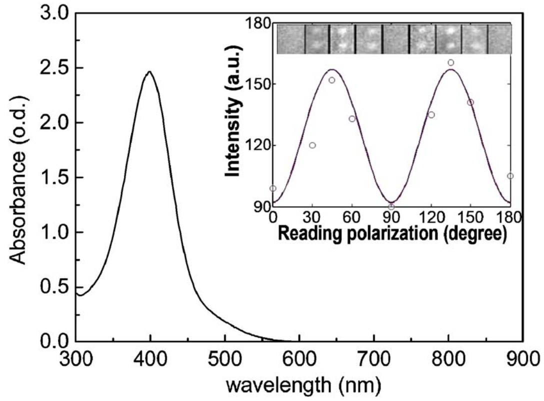

Fig. 1. Absorption band of the sample. Inset, bit intensity of the recorded bits as a function of the reading polarization direction. The sample thickness is 80 μm. Two vertical bits were recorded by a horizontally polarized beam. The recording power and the exposure time are 10 mW and 25 ms, respectively. The circles are the experimental bit intensity, and the solid curve is the theoretical prediction.

cross section we measured according to $I(d)=I_0/[1+(N\sigma_2/h\nu)I_0d]$ is 1×10^{-48} cm^4 s photon^{-1},[14] while the single-photon-induced recording cannot be observed up to the intensity of 0.5 GW/cm^2.

The experimental setup for 2P recording was a typical microscopy system. A linearly polarized Ti:sapphire ultrashort pulsed laser beam of pulse width 100 fs and repetition rate 82 MHz (Spectra-Physics Tsunami) at wavelength 780 nm was employed as a light source for 2P excitation. A quarter-wave plate and a Glan–Thompson prism were used to control the polarization state of the recording beam. An objective (NA=0.7, 20×) was used to focus the excitation beam onto the recording sample. A commercial transmission microscope (Olympus BX50) and a high-NA objective (NA=1.4) were used for readout of multilayer information. To avoid any erasure of recorded bits in the reading process, we used the Ti:sapphire laser at a continuous-wave mode with one fifth of the recording beam power. The sample was placed between two orthogonal polarizers, which were simultaneously rotated to choose a reading polarization state during readout.

The images in the inset of Fig. 1 show the two bits recorded by the parallelly polarized femtosecond laser beam at wavelength 780 nm as well as the corresponding readout intensity as a function of the reading polarization direction. It can be found that the highest- and lowest-intensity positions occur when the angles between the recording and the reading beam polarization directions are 45° and 90°, respectively. In other words, the readout intensity has an angular dependency of $\sin^2(2\theta)$, where θ is the angle between the polarization directions of the recording and reading beams, which is consistent with the theoretical prediction[15] (solid curve). This result confirms that the observed angular dependency is caused by the 2P-induced birefringence property via a trans–cis–trans process, where the optical axis of the excited molecules is intended to be orientated to the direction perpendicular to the polarization of the recording beam.[10,12]

In fact, not all molecules in the focus volume can be aligned to the perpendicular direction of the recording polarization because of their initial random orientation. Therefore one can encode two letters in the same layer by separating the two polarization directions of recording beams at an angle of 45°. Figure 2 demonstrates six 2P-induced polarization-encoded patterns recorded in three layers. In each layer, two letters are recorded when the polarization direction of the recording beam is orientated at 0° for one letter and 45° for the other. Each pattern consists of 21 ×21 bits with a bit spacing of 4 μm. Each bit was recorded with the given exposure condition (recording power of 16 mW and exposure time of 25 ms). The separation between the adjacent layers is 30 μm. Patterns P B, I J, and C D were multiplexed in the first, second, and third layers, respectively. Thus the estimated memory density of our system is approximately 50 Gbits per disk. However, because of the depolarization effect of high-NA objectives,[16] the bit intensity does not drop to zero (as shown in the inset of Fig. 1), therefore leading to a small amount of cross talk. In addition, irreversible changes in the storage medium may be another reason for cross talk.

Since the aligned molecules can relax back to their original states, the recorded bits can be erased completely when exposed to an environment temperature higher than T_g ($T_g\sim45°C$) or a He–Ne beam of polarization perpendicular to that of the recording beam. Figure 3 shows the erasing time as a function of the recording power, with a He–Ne laser of power 0.4 mW used for erasure. Under our recording conditions the recorded bits can be completely erased in 3 min. The inset illustrates the bit readout contrast as a function of the recording power. The circles present the results read out immediately after recording, while the triangles are the results read out after erasing. For a recording power level between 5 and 20 mW, the recorded bits are erasable and rewritable with high contrast. We estimated that the temperature rose to $\sim60°C$ at a recording power of 5 mW and exposure time of 25 ms. Beyond 20 mW,

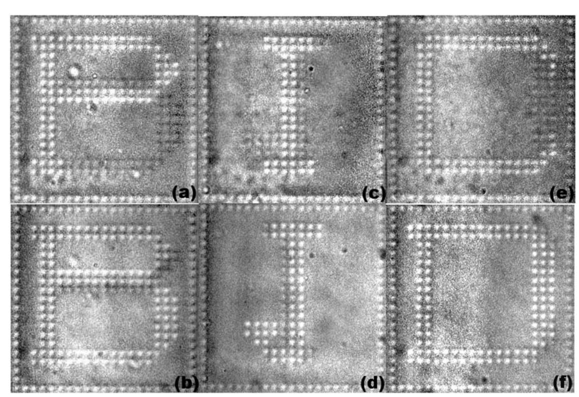

Fig. 2. Demonstration of 2P polarization-encoded patterns in the same region in three layers of the sample. (a) (b), Letters P and B recorded in the first layer; (c), (d) letters I and J recorded in the second layer; (e), (f) letters C and D recorded in the third layer.

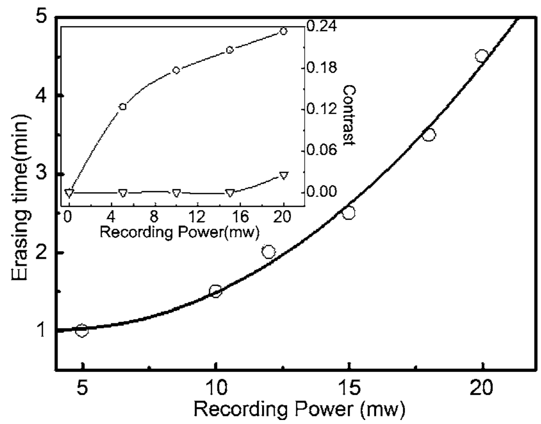

Fig. 3. For an erasing laser of power 0.4 mW, erasing time as a function of the recording power. Inset, readout contrast as a function of the recording power immediately after recording (circles) and after erasing by a vertically polarized beam (triangles).

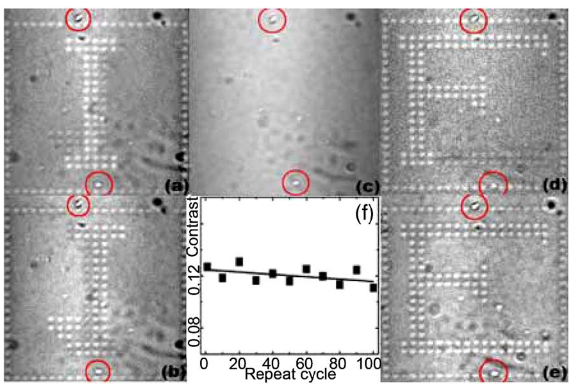

Fig. 4. (Color online) Erasing and rewriting patterns in a particular layer. (a), (b) Letters I and J encoded in the same region in the second layer; (c) the same area after letters I and J are completely erased by a vertically polarized laser beam; (d), (e) letters F and E are rewritten in the same region. The circled defects indicate that all the patterns are recorded in the same area. (f) Recording contrast (squares) as a function of the repeat cycle.

permanent recording was observed, caused mainly by thermal damage to the matrix. It has been noted (not shown here) that if DMNPAA is doped in a loose matrix of PVK:ECZ (concentration of 30:54:16), it is more difficult to achieve a stable erasable and rewritable region.

Figure 4 demonstrates the erasing and rewriting of information in a particular layer. Figures 4(a) and 4(b) show the two letters I and J recorded in the second layer with polarization encoding at 0° and 45°. For erasing, a vertically polarized He–Ne laser was focused by an objective (NA=1.4;60×) to scan the given layer for 3 min. Both patterns were completely erased as shown in Fig. 4(c). In Figs. 4(d) and 4(e) two new patterns, F and E, were rewritten in the same area at 0° and 45° to the recording polarization, respectively. The repeat cycle of our sample we estimated should be at least 1000 times, because there was no significant degradation after a test of erasing and rewriting up to 100 times at a recording power of 10 mW and an exposure time of 25 ms [see Fig. 4(f)].

In conclusion, we have developed the 2P-induced polarization encoding technique for multilayer bit-by-bit data storage in a DMNPAA chromophore-doped polymer. Two letters have been successfully encoded in the same region in each of the three layers and read out distinctly layer by layer by using the polarization interference method. Our results show a potential for multiple-state polarization-encoded optical data storage, which will significantly increase the data density under 2P excitation.

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