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Use of polarization sensitivity for three-dimensional optical data storage in polymer dispersed liquid crystals under two-photon illumination

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We report on the use of the polarization-dependent fluorescence property in photorefractive polymer dispersed liquid crystals for three-dimensional (3D) optical data storage under two-photon absorption. The liquid crystals, which have optical anisotropy, exhibit an alignment-based fluorescence feature that depends on the polarization state of an excitation field. Such a polarization sensitivity can be confined to the small focal region under two-photon excitation, allowing the stable recording of a 3D bit array. A 3D data density of 204.8 Gbits/cm^3 is achieved using ultrashort pulsed illumination at a wavelength of 900 nm. It is also demonstrated that the recorded data can be erased in bulk or as single bits and then rerecorded.
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The need for high-density optical data storage has been a topic of interest for a number of years. Two-photon (2-p) excitation has been demonstrated as a promising method for three-dimensional (3D) storage of bit optical data as it allows spatial confinement of the focal spot in three dimensions.[1–9] Currently, polymer-based photochromic,[1,2,5,6] photopolymerizable,[3] photobleaching,[7] and photorefractive[8,9] materials show the most interest because they are inexpensive to manufacture and their physical and chemical properties can be easily modified. In a photorefractive polymer, a modulation of the refractive index is generated by a space-charge separation.[10,11] This has led to a localized 2-p photorefractive mechanism for erasable and rewritable 3D data storage,[8,9] although a phase sensitive readout system is needed to collect the signal from the small refractive-index modulation. The refractive-index modulation can be increased in photorefractive polymer dispersed liquid crystals (PDLCs) as a result of the alignment of the liquid crystal directors induced by the internal space-charge field.[12–15] Because liquid crystal domains in PDLCs are in a submicrometer scale, the generation of a localized 2-p photorefractive effect[9] in the focal region in PDLCs remains a challenge.

However, it is known that liquid crystal domains in PDLCs have a microscopic inhomogeneity in molecular alignment and mobility and therefore exhibit fluorescence.[16,17] This type of fluorescence in PDLCs is also dependent on the realignment of the liquid crystal directors under polarized illumination and thus provides a mechanism for 3D optical data storage. When a linearly polarized writing beam is focused into a PDLC sample, a certain number of liquid crystal directors in the focal region are aligned with the illumination polarization direction and the resulting fluorescence varies with the polarization state of a reading beam. However, in the region without illumination (where liquid crystal directors are randomly oriented) such a polarization dependence is not be pronounced. In photorefractive PDLCs, the aligned directors can be trapped due to the space-charge field, which increases the stability of recorded bits. The aim of this letter is to demonstrate the polarization-dependent fluorescence property in PDLCs under 2-p excitation for 3D optical data storage.

The PDLCs used in our experiment consist of poly(methyl methacrylate) (PMMA) doped with liquid crystals, a photoabsorber, and a plasticiser.[12] The liquid crystal, 4-pentyl 4-cyano biphenyl (E49), was purchased from Merck. The absorber, 2,4,7-trinitro-9-fluorenone (TNF), has absorption from the UV to visible region of the spectrum. The plasticiser, *N*-ethylcarbazole (ECZ), reduces the glass transition temperature of the polymer. The concentration of the components (PMMA:E49:ECZ:TNF) was 45:33:21:1 wt %. The solvent induced phase separation method was used to form the PDLCs onto a glass slide with chloroform as the solvent. The thickness of the sample was approximately 320 μm. The absorption of the sample (see Fig. 1) was found to be negligible at a wavelength of 900 nm. Therefore, an infrared wavelength at 900 nm can be utilized to produce 2-p excitation.

The optical set up for recording and reading experiments was the same as that used before.[18] The illumination source was an ultrashort pulsed laser (Spectra-Physics Tsunami) operating at wavelength 900 nm and focused into the sample by an Olympus ULWD MSPlan100-IR objective with numerical aperture 0.80. The polarization state of the writing and reading beams was controlled with a $\frac{1}{4}$ wave plate and a Glan–

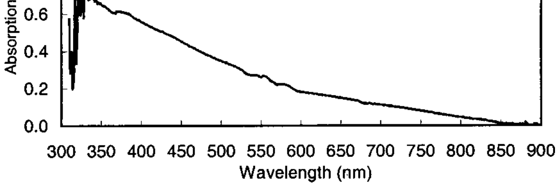

FIG. 1. Absorption spectrum of the PDLC sample used in the experiment.

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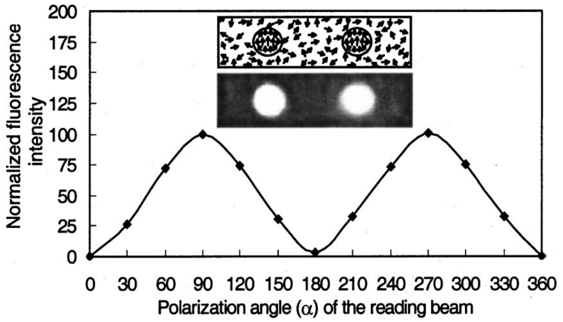

FIG. 2. Normalized two-photon fluorescence intensity as a function the polarization angle α of the reading beam. Inset (a) gives a schematic mechanism for the alignment of liquid crystal directors in the exposed and unexposed regions. Inset (b) shows two fluorescing data bits ($\alpha = 90°$).

Thompson polarizor. The Glan–Thompson polarizor has an extinction ratio of 100 000:1. The fluorescence from the sample (at a wavelength of 574 nm) is collected via a dichroic beamsplitter and a short pass filter into a photomultiplier tube. The average laser power in the focus of the objective was 20 mW for writing with an exposure time of 100 ms and 6 mW for reading.

E49 is an optically uniaxial medium with positive dielectric anisotropy. In the absence of an electric field, the orientation of the liquid crystal directors within the droplets is random. In the focal region, the directors tend to align parallel with the applied electric field. This situation is schematically shown in inset (a) of Fig. 2. As a result of the alignment of the directors, the fluorescence intensity from the illuminated region is highly dependent on the polarization state of a reading beam. The dependence of the fluorescence from an exposed region on the polarization angle α (defined to be the angle between the writing and reading polarization states) is depicted in Fig. 2. It is clear that the maximum fluorescence intensity occurs when the polarization state of the reading beam is perpendicular to that of the writing beam (i.e., when $\alpha = 90°$). The appearance of the fluorescence intensity maximum at $\alpha = 90°$ may be caused by the less absorption when the directors are orthogonal to the polarization state of the reading beam. Such a relationship was not observed in the unexposed region, which leads to sharp contrast of readout bits as shown in inset (b) of Fig. 2.

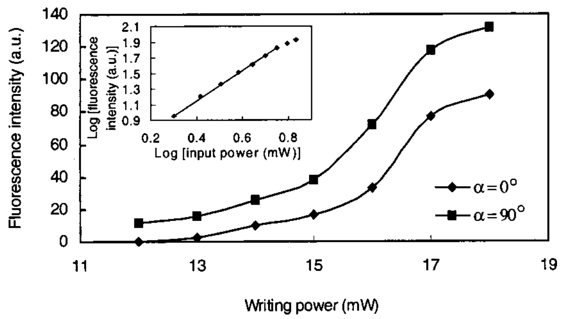

FIG. 3. Two-photon fluorescence intensity as a function of the writing power for a reading beam (6 mW) at $\alpha = 0°$ and $\alpha = 90°$, respectively. The inset shows the dependence of the two-photon fluorescence intensity on the reading power in a log scale (writing power = 20 mW).

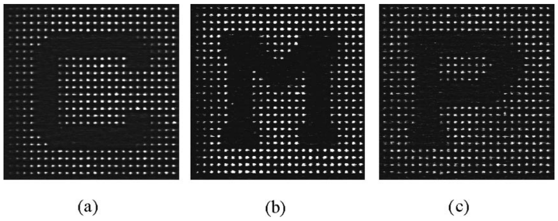

FIG. 4. Three layers of data bits recorded at different depth of the PDLCs sample with a point spacing of 1.6 μm and a layer spacing of 2 μm: (a) The first layer of letter C recorded at a depth of 2 μm; (b) The second layer is letter M; (c) The third layer is letter P.

The image contrast of the recorded bits depends on the writing power. As indicted by Fig. 3, if the writing power is too high, a saturated nature occurs, which results in a significant reduction in image contrast. The dependence of the fluorescence intensity for $\alpha = 90°$ on the reading power, depicted in the inset of Fig. 3, shows a slope of 1.98 and thus confirms the quadratic dependence of the fluorescence emission under 2-p excitation.

The feasibility of 3D recording and reading in terms of the polarization-dependent fluorescence property is demonstrated in Fig. 4. The first layer recorded near the surface of the sample shows letter "C" composed of a grid of 24×24 data bits [Fig. 4(a)]. Subsequent two letters "M" and "P" [Figs. 4(b) and 4(c)] were written every 2 μm along the depth of the polymer. With a point spacing of 1.56 μm and a layer spacing of 2 μm a data density of 204.8 Gbits/cm^3 is achieved, which is equivalent to a capacity of 2.4 Tbits or 298.8 Gbytes for a CD size disk.

Since the space charge field formed in the photorefractive PDLCs when the region is exposed to the writing beam traps the position of the aligned liquid crystals, the recorded data bits are quite stable for a few days or after being read constantly. This feature was confirmed in a sample without doping TNF, in which case the recorded bits self erase in a few minutes.

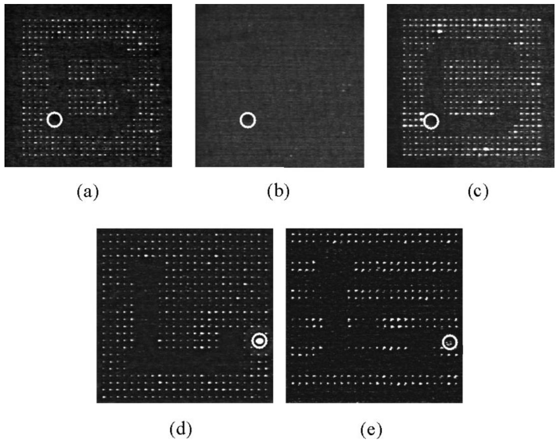

FIG. 5. Demonstration of the erasable nature of the PDLCs sample. The three images in (a), (b), and (c) show the bulk erasing nature, while the two images in (d) and (e) exhibit the bit erasing nature. (a) Letter B is recorded at a depth of 8 μm; (b) Letter B is erased after UV exposure; (c) Letter C is written in the same area as the previous letter B (the white ring highlights a defects in the polymer, confirming the same location and depth in three images). (d) Letter L recorded at a depth of 4 μm. (e) Letter L read after a few lines of data bits are illuminated with a beam of a polarization state perpendicular to that of the writing beam.

The data stored in the PDLCs sample can be erased in three ways. First, heating the sample above the glass transition temperature will allow the directors of the liquid crystals to relax back to their random orientation, thereby removing the stored information. Second, the data can be erased in bulk by unpolarized UV radiation. Figure 5 shows the results of bulk erasing. After the uniform unpolarized UV exposure for 5 s, the directors of the liquid crystals within the droplets in the recorded region [Fig. 5(a)] were reoriented back to their random state. This reduces the fluorescence strength to the background level and effectively causes the data bits to be deleted [Fig. 5(b)]. A grid of data bits can be rewritten in the same region [Fig. 5(c)].

The third erasing method in the PDLCs is in a bitwise fashion based on the fact that the fluorescence strength of the data bits is a function of their alignment (Fig. 2). By realigning the directors of the liquid crystals the fluorescence at individual bits may be extinguished and bit erasing can be accomplished. Figures 5(d) and 5(e) demonstrate this ability. In this case, every two lines of the recorded data bits [Fig. 5(d)] were illuminated by the laser beam with its polarization state perpendicular to that of the writing beam. As a result, they cannot be readout by a reading beam at $\alpha = 90°$.

In conclusion, we report on the observation of the polarization-dependent fluorescence property in the PDLCs under 2-p excitation. Because this polarization sensitivity is based on the alignment of liquid crystal directors in the focal region, it has been adopted for 3D high-density optical data storage and provided a mechanism for single bit erasure of the recorded data.

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