Effect of saturable response to two-photon absorption on the readout signal level of three-dimensional bit optical data storage in a photochromic polymer

Min Gu[a)]
Center for Micro-Photonics, School of Biophysical Sciences and Electrical Engineering, Swinburne University of Technology, P.O. Box 218, Hawthorn, Victoria, Australia

Jose Omar Amistoso
National Institute of Physics, University of the Philippines, Diliman, Quezon City, 1101, Philippines

Akiko Toriumi
Department of Applied Physics, Osaka University, Suita, Osaka 565-0871, Japan

Masahiro Irie
Department of Chemistry and Biochemistry, Kyushu University, Hakozaki, Fukuoka, 812-8521, Japan

Satoshi Kawata
Department of Applied Physics, Osaka University, Suita, Osaka 565-0871, Japan

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We observe a saturable change in the refractive index induced by two-photon absorption in a photochromic polymer, while the erasable nature of the polymer is maintained. As a result, the support region of spatial frequencies of the recorded data bits caused by the refractive index change in the polymer is enlarged. This feature leads to the significant enhancement of the signal level in a reflection confocal microscope readout system for three-dimensional photochromic bit optical data storage. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1383999]

Three-dimensional (3D) bit optical data storage based on two-photon absorption has attracted more and more interest over the last ten years.[1–11] A change in the refractive index induced by two-photon absorption in crystal[3,8] and polymer[3,7,9] materials provides a mechanism for 3D erasable and rewritable memories. In particular, 3D data bits recorded by two-photon absorption in a photochromic polymer can be readout using a reflection confocal microscope,[7] which is impossible under single-photon excitation.[11–13] As result, a compact reflection optical system can be used in both the writing and reading processes for two-photon 3D photochromic optical data storage.[3]

However, the readout signal level from the photochromic polymer is limited because the support region of spatial frequencies of the refractive data bits caused by two-photon absorption partially overlaps the passband of the 3D coherent transfer function for a reflection confocal microscope readout system.[12,13] In this letter, we report on the observation of a saturable, but erasable, change in refractive index in a photochromic polymer under two-photon excitation. Consequently, the signal level from a reflection confocal microscope readout system is increased significantly.

The photochromic polymer used in the experiment was a poly(methylmethacrylate) film doped with photochromic molecules, cis-1,2-dicyano-1,2-bis(2,4,5-trimethyl-3-thienyl)ethene (B1536).[3,7] As B1536 is a yellow isomer which can be converted into a red isomer under ultraviolet illumination at a wavelength of 380 nm, it can be excited via two-photon absorption at a wavelength of 760 nm.[7]

Therefore, an ultrashort pulsed laser (Spectra-Physics: Tsunami), tuned at 760 nm and 80 fs, was focused by an oil-immersion objective of a numerical aperture of 1.4, onto the photochromic film of a thickness of 1 mm. The use of an oil-immersion objective can reduce the effect of spherical aberration caused by the mismatch of the refractive indices between the recording material ($n = 1.59$) and its immersion medium ($n = 1.518$).[14] The laser power in the focus of the writing objective varied from 4.2 to 6 mW and the writing time for each bit was 8 ms. Under these conditions, four data bits with a spacing of 5 μm were written into one line under the illumination of the same laser power. For reading out data bits, a Zeiss Laser Scanning Microscope (LSM) 410 reflection confocal scanning microscope with a He–Ne laser (3 mW) and an oil-immersion objective of a variable numerical aperture was used. The reading wavelength at 633 nm was outside the absorption band of the recording material,[7] so that the recorded data bits were not erased during the reading process.

The dependence of the measured intensity and the bit size, averaged over the four bits recorded with the same power, on the writing power is shown in Fig. 1. It is clear that the dependence is approximately quadratic when the writing power is less than 5 mW, which indicates that the change in refractive index induced by two-photon excitation is unsaturated. However, when the writing power is greater than 5 mW, the dependence becomes saturated. This situation is further demonstrated in the inserted plots in Fig. 2. It should be emphasized that all the bits recorded with writing power less than 6.5 mW can be erased under the illumination of a wavelength at 543 nm.[7] A further increase in writing power leads to permanent damage of the polymer.

[a)] Electronic mail: mgu@swin.edu.au

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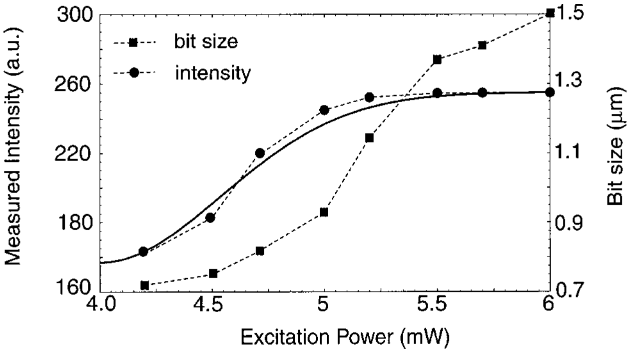

FIG. 1. Reflected intensity (circular spots) and bit size (square spots) as a function of the writing/excitation power for each bit are shown. The numerical aperture of the reading objective is 1.4 (oil). The solid curve gives a fitted relationship of the reflected intensity to the writing power.

If the dependence of the reflected intensity I on the writing power P is mathematically fitted with the subtraction of the background intensity, we have

$$I = I_s[1 - \exp(-1.7P^2)], \tag{1}$$

where I_s is the saturation intensity. This relation can be expanded into

$$I = I_s(1.7P^2 - 1.4P^4 + 0.8P^6 + \cdots + a_j P^{2j} + \cdots), \tag{2}$$

where j is the order of the terms in the series. The first term $(j=1)$ in Eq. (2) shows the quadratic dependence confirming unsaturated two-photon absorption and the higher order terms $(j \geqslant 2)$ suggest that the two-photon absorption in the polymer is saturable when the writing power becomes high.

The physical origin of the saturable response to two-photon absorption in the photochromic polymer observed in Fig. 1 may arise from the fact that the conversion between the yellow and red isomers becomes balanced when their ratio approaches 60:40.[15,16] While this mechanism should be further explored using spectroscopy, the resultant saturable change in reflected intensity/refractive index can prove advantageous in terms of increasing the readout signal level of the 3D optical data storage. To demonstrate this feature, let us recall that the distribution of the writing power P is proportional to the 3D intensity point spread function for a writing objective, which can be expressed as $|h(v,u)|^2$.[13,14] Here v and u are transverse and axial optical coordinates, respectively.[14]

The support region of spatial frequencies of the recorded 3D data bits, $S(l,s)$, is given by the passband of the 3D Fourier transform of Eq. (1)[13,14] and enclosed by the following functions:

$$S(l,s) = \pm \begin{cases} 2jn_w k_w\left[1 - \cos\left(\sin^{-1}\left(\dfrac{NA_w}{n_w}\right)\right)\right], & 0 \leqslant l < 2jk_w NA_w, \\ \sqrt{(4jn_w k_w)^2 - (l - 2jk_w NA_w)^2} - 2jn_w k_w \cos\left(\sin^{-1}\left(\dfrac{NA_w}{n_w}\right)\right), & 2jk_w NA_w \leqslant l \leqslant 4jk_w NA_w \end{cases}, \tag{3}$$

where n_w, k_w, and NA_w are the refractive index of the immersion material, the wave number of the writing beam in a vacuum, and the numerical aperture of the writing objective, respectively. Here l and s are the transverse and axial spatial frequencies, respectively. Because the strength of the higher order term $(j \geqslant 3)$ is weaker than that of the first $(j=1)$ and second $(j=2)$ order terms, only the effect of the first two terms are considered in the following discussions.

The support regions for $j=1$ and $j=2$ in Eq. (3) are shown in Figs. 3(a) and 3(b). The regions enclosed by the dotted curves in Figs. 3(a) and 3(b) represent the passbands of the 3D coherent transfer function for reflection confocal

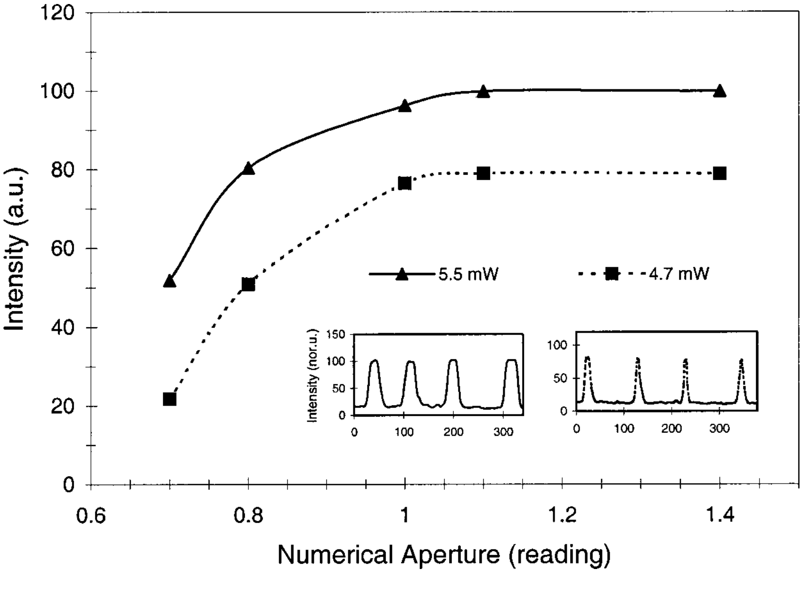

FIG. 2. Reflected intensity as a function of the numerical aperture of a reading objective under the writing powers of 5.5 mW and 4.7 mW, respectively is shown. The inserted plots represent the intensity profiles (full horizontal scale is 40 μm) of the bits exposed under the illumination powers of 5.5 (solid curve) and 4.7 mW (dotted curve).

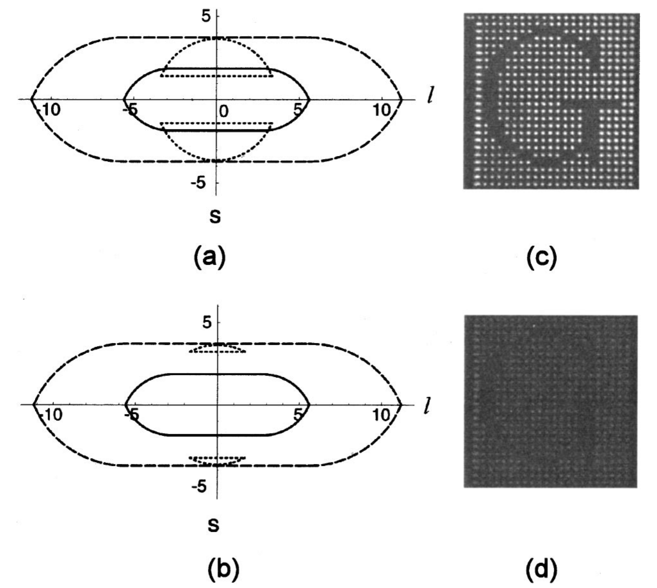

FIG. 3. The solid and dashed curves in (a) and (b) represent the support regions of spatial frequencies of data bits recorded by two-photon absorption under the unsaturated (solid) and saturated (dashed) recording conditions. The dotted curves in (a) and (b) represent the passbands of the 3D coherent transfer function for a confocal microscope readout system of an oil-immersion objective with numerical apertures of 1.4 (a) and 0.7 (b). The images of letter G in (c) and (d), consisting of 24×24 bits and a bit spacing of 5 μm, are recorded with an illumination power of 5 mW and readout under the conditions corresponding to (a) and (b), respectively.

microscopy with an oil-immersion objective of numerical apertures of 1.4 and 0.7, respectively. All passbands and support regions have been normalized by the writing wavelength λ_w. It is clear that the readout system in Fig. 3(a) transfers the spatial frequency components of both the first and the second order terms, while that in Fig. 3(b) transfers only a small part of the second order term. This feature leads to a higher readout signal level in the former case, which is confirmed by the experimental results in Figs. 3(c) and 3(d).

Figure 2 also shows the dependence of the measured readout signal level on the numerical aperture of the reading objective for illumination powers of 4.7 and 5.5 mW, respectively. The former corresponds to the unsaturated case while the latter to the saturated condition. As expected, for a given numerical aperture, the readout signal level of the data bits recorded under illumination power of 5.5 mW is increased by 40%.

The condition for confocal microscope readout of 3D data bits in the photochromic polymer can be derived in terms of the fact that the passband of the coherent transfer function should partially or completely overlap the support regions of spatial frequencies in Eq. (3),[13] which leads to

$$NA_r \geqslant n_r \sin\left\{ \cos^{-1}\left(\beta \frac{j n_w}{n_r} \left[1 - \cos\left(\sin^{-1} \frac{NA_w}{n_w} \right) \right] \right) \right\}, \tag{4}$$

where n_r and NA_r are the refractive index of the immersion material and the numerical aperture of the reading objective, respectively. $\beta = \lambda_r / \lambda_w$, where λ_r is the wavelength of the reading beam.

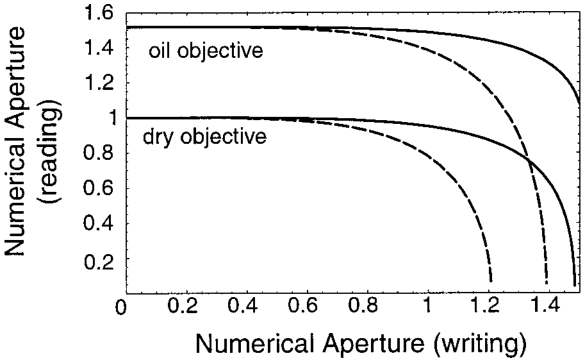

FIG. 4. The critical condition for the reading numerical aperture as a function of the writing numerical aperture under the unsaturated (solid curves) and saturated (dashed curves) conditions is shown. Only the second order terms have been considered.

It is seen that the condition in Eq. (4) depends on the wavelength and the numerical aperture used in the writing and reading processes. The critical condition which corresponds to the equity sign in Eq. (4) is shown in Fig. 4 for dry ($n_r = 1$) and oil ($n_r = 1.518$) reading objectives. Clearly, it is advantageous if the writing wavelength is longer than the reading wavelength, and more spatial frequency components from saturated data bits under two-photon absorption can be transferred in a reflection confocal microscope readout system.

In conclusion, it has been found that the erasable change in the refractive index in photochromic polymer under two-photon excitation is saturable as a result of the balance of the conversion between the two isomer states. Such a saturable change leads to more spatial frequency components in recorded 3D data bits. Consequently, the signal level in a reflection confocal microscopy readout system can be increased approximately by 40%.

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