Fast Bits Recording in Photoisomeric Polymers by Phase-Modulated Femtosecond Laser

Yanlei Hu, Jianqiang Ma, Yuhang Chen, Jiawen Li, Wenhao Huang, and Jiaru Chu

***Abstract*— High storage capacity and recording-access rate are two critical factors that the current optical storage technology pursues. Efforts on shortening the laser wavelength, increasing the numerical aperture of objective, and increasing the optical drive speed have been made to reach this end. However, these methods have run their full course because of many existing difficulties on the physical and manufacturing aspect. People have to explore innovative ways to satisfy the requirement of both mass data and high rate. Multidimensional optical storage and holographic recording are two promising technologies to increase the capacity of memory and recording rate by orders of magnitude. We report on the fast bits recording in bisazobenzene functional polymer film by femtosecond laser holographic recording. The mechanism of the photo-induced anisotropy of the material is reviewed and discussed. The demonstration of multidimensional holographic recording is presented. Some influence factors in the holographic processing are discussed.**

***Index Terms*— Femtosecond laser, data storage, azobenzene, holographic recording, multi-dimensional storage.**

I. Introduction

FROM the overview of the development history of commercial optical disc from compact disc (CD) to current Blu-ray disc (BD), one can summarize that the engineers endeavor to increase the storage capacity and transfer rate by shortening the laser wavelength, increasing the numerical aperture (NA) of objective and increasing the optical disc rotating speed. However, these techniques are approaching their end because of some physical or technical limitations, e.g., the use of laser source with wavelength less than 405 nm is problematic due to the high-cost and probably strong absorption of the disk substrate medium, the manufacturing of high-NA objective lens without immersion is still a grand challenge [1], and moreover, the fast disc rotation brings a multitude of problems such as difficulties in tracking feedback control and signal processing. Therefore, for developing next generation storage technology, some novel approaches having capacity to store up more information with higher recording rate superior to Blu-ray disc, such as Multiplexed Optical Data Storage (MODS), Holographic Versatile Disc (HVD) and Protein-coated Disc (PCD), are greatly desired and intensively studied.

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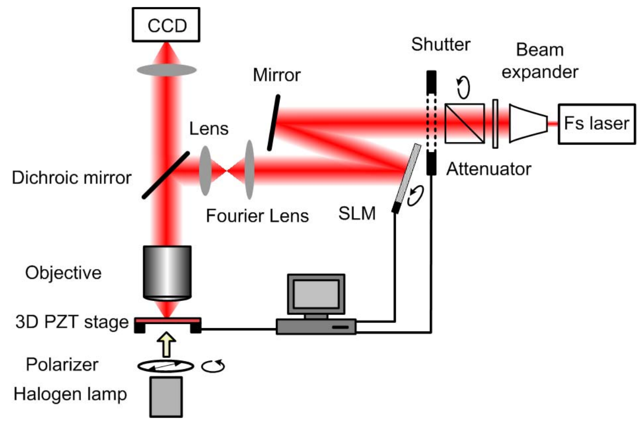

Fig. 1. Optical configuration for holographic recording and reading.

Aided by the nanometric resolution of two-photon absorption (TPA), multilayer data recording can be easily achieved. As many as hundreds of bits layers can be expected to be stored by two-photon induced physico-chemical reaction inside bulk photosensitive materials. Based on this, multi-dimensional data storage technology is exploited in recent years by integrating other recording constrains such as polarization and wavelength to further increase the storage capacity [2]. The bits density of up to Tb/cm^3 and good compatibility with former storage technologies make it a potential candidate for next generation optical storage. However, recording rate has not been fully considered in multi-dimensional storage, which becomes more crucial when the bits amount is greatly increased.

In this letter, we perform polarization-multiplexed and holographic recording in bisazobenzene functional polymer films by using femtosecond laser. Two-photon induced isomerization and orientation are studied. Versatile holographic recording are utilized to ultimately increase the recording rate. And moreover, some influence factors in the holographic recording are discussed.

II. Femtosecond Laser Holographic Recording

A femtosecond laser holographic recording system is built, as illustrated in Fig. 1. A femtosecond pulsed laser emitted

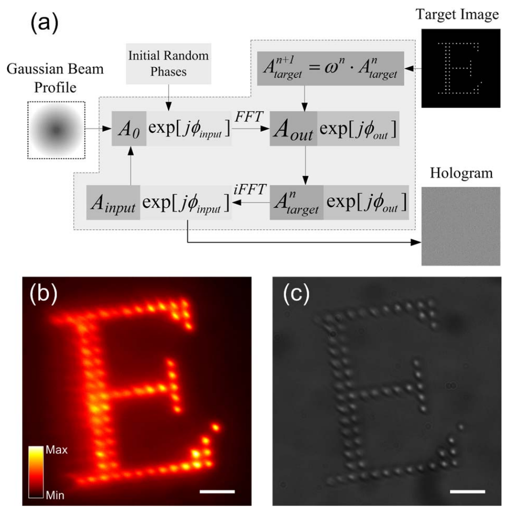

Fig. 2. (a) Flowchart of the weighted GS algorithm. (b) The intensity distribution map of the resulting multiple foci pattern. (c) Recorded bits by a single-shot of femtosecond laser. Scale bar: 10 μm.

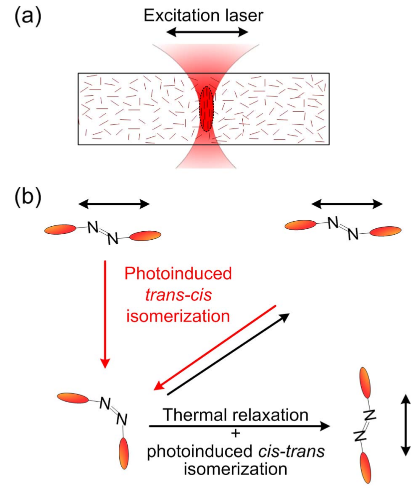

Fig. 3. (a) Photoinduced reorientation of the bisazobenzene molecules. (b) The detailed process of photoinduced isomeric orientation.

from a mode-locked Ti:sapphire laser oscillator (Coherent, Chameleon Vision-S) with central wavelength of 800 nm and pulse duration of 75 fs is used. The output power is >3W and one part of it is split and introduced into a regenerative amplifier (Coherent, Legend) to generate low repetition femtosecond pulses at 1 kHz. The combination of a half-wave plate and a Glan laser prism is used to control the polarization state and the power of recording beam. A reflective pixelated liquid crystal on silicon (LCoS, resolution: 1920 $\times$ 1080 pixels) produced by Holoeye is employed as spatial light modulator (SLM). The modulation effect of the SLM is incoming polarization dependent; therefore it is designed to be switchable between two states (horizontally or vertically mounted) in order to match the polarization of the incident laser. A Fourier lens (f = 600 mm) is placed behind the SLM and light spots for desired bits sequence are generated at its back focal plane. A convex lens with focal length of 200 mm and a 50 $\times$ objective lens (*NA*= 0.8, f =3.6 mm) compose a $4f$ optical system to reduce the light spots array into the storage medium for parallel recording. A mechanical shutter and a piezo-driven nano-positioning stage are synchronously controlled to determine the exposure time and bits sequence location. A CCD camera, a dichroic mirror and a halogen lamp with a polarizer above it are used to read out the recorded bits.

A weighted Gerchberg-Saxton (GS) algorithm is developed to calculate the computer-generated hologram (CGH) for holographic recording [3], as shown in Fig. 2 (a). A random phase distribution and a Gaussian amplitude distribution are selected for initialization. After performing a fast Fourier transform (FFT), the light field in the Fourier plane is obtained. Then the amplitude is replaced by the desired pattern A_{target}, and meanwhile the phase ϕ_{out} remains the same. An inverse fast Fourier transform (iFFT) is implemented to shift back to the input plane. The amplitude is updated by the Gaussian distribution and one iteration loop is finished. Most essentially, the intensity of each spot in desired target multi-foci pattern is monitored during the iteration, and corresponding weighting factor ω_n is employed to update the original target pattern for the next iteration loop. Convergence occurs after several iterations and the phase distribution (i.e. CGH) for the input plane is achieved. Fig. 2 (b) shows the intensity distribution map of the resulting multiple foci pattern. An oil-soluble CdSe nanocrystals solution (Jiayuan Quantum dots Co., LTD, China) is used as imaging film under the irradiation of 800 nm femtosecond laser and a CCD is used to detect the fluorescence intensity. It can also be seen that the intensity of each light spot is not so uniform. The spots in the right (nearer the zero-order light) are a little brighter than the left ones. The slight uniformity of the spots intensities results from the inherent constraints of the SLM and can be readily compensated. Fig. 2 (c) shows the recorded bits in our storage medium by a single-shot using the same CGH. Corresponding to the measured intensity distribution, the bits signal in the right part is relatively strong because of the higher writing power.

III. Multi-Dimensional Recording

Polymers containing bisazobenzene chromophores (M2BAN), which have two alternated azo bonds between three benzene rings, is used as the storage medium here [4]. Compared with azobenzene chromophores, the bisazobenzene is considered to have better nonlinear optical properties, long-time stability, and enhanced birefringence [5]. Two-photon induced isomeric orientation in bisazobenzene polymer is similar to the single-photon isomerization (see Fig. 3 (a)). Under the irradiation of purely polarized femtosecond laser, two kinds of geometric isomers will interconvert into each other rapidly, as depicted in Fig. 3 (b). Before irradiation, only the thermodynamically stable *trans* isomers exist and their angular distributions are in a random arrangement. Generally, the probability of TPA occurrence is proportional to $\cos^4\theta$ [6], where θ is the angle between the TPA transition dipole moments (TDMs) and the polarization direction of the excitation laser. Therefore, only the *trans* isomers with TPA-TDMs parallel to the laser polarization absorb photons and finally result in photoinduced *trans-cis* isomerization.

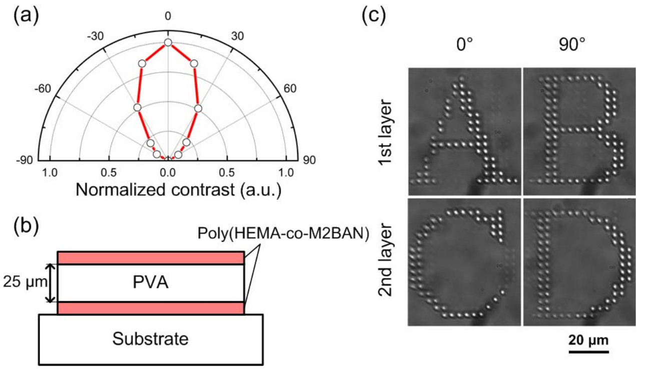

Fig. 4. (a) Readout bits contrast versus the angle between recording and reading polarization. (b) The structure of the storage medium. (c) Readout results of the two-layer polarization-multiplexed data.

There are two ways that the molecules come back to *trans* state. One is the spontaneous thermal reaction because the *cis* isomers are not thermal stable in room temperature. The other is the photoinduced reverse *cis-trans* isomerization. The *trans* isomers whose TDMs are perpendicular to the laser polarization will no longer absorb photons, while the other isomers will continue to undergo the two-photon isomerization until all the *trans* isomers orientation is perpendicular to the direction of the laser polarization.

The isomeric orientation of bisazobenzene polymer results in macroscopic optical anisotropy. As shown in Fig. 4 (a), the readout bits signal varies with the angle between the recording beam polarization and the reading light polarization. It is indicated that when the direction of reading polarization is parallel to recording polarization, the readout intensity and contrast reach the highest. The bits contrast gradually decrease when changing the angle from 0° to 90° . When the polarization of reading light is perpendicular to that of recording beam, no bits intensity can be observed. This promising feature makes multiplexed storage feasible, as researchers have demonstrated in laboratory [7]. It is also worth mentioning here that the expose dose is relatively low and no photoinduced surface deformation is involved. If the recording power is high enough, the surface of the material will deform and the appearance of readout bits will change. Both bright and dark bits can be observed as reported before [8]. It also can be seen from Fig. 4 (a) that the angular dependence of the bits contrast is narrower than expected, i.e., the contrast decrease to nearly zero when the angle is larger than 60°. This property can significantly reduce the cross talk between the multiplex data domain even when more than two kinds of bits sequences are recorded in the same region.

In order to demonstrate the fast multi-dimensional data storage integrating both spatial dimension and polarization, we prepare a dual-layer poly(HEMA-co-M2BAN) with better film forming property than poly(MMA-co-M2BAN) that is previously synthesized [4], and a polyvinyl alcohol (PVA) layer is inserted between two functional layers as the spacer layer. The geometric structure of the film is shown in Fig.4 (b). The thicknesses of the functional layers and transparent PVA layers are 5 μm and 25 μm, respectively. The two-layer polarization-multiplexed holographic recording results are successfully demonstrated in Fig. 4 (c). The phase of recording beam is modulated by the CGH and foci array patterns like letter “A” are generated. The patterns “A” and “B” are recorded in the same region of the first layer with two orthogonally polarized femtosecond laser, “C” and “D” are recorded in the second layer. The corresponding polarization of the reading beams is the same as the recording beams.

In principle, arbitrary spots patterns or bits sequences can be rapidly recorded with this method. However, the amount and arrangement of the foci spots are still limited by the following two aspects: the output power of laser and the physical apertures of the system. The total power of the recording laser is constant and when too many spots are generated, the power of each spot will not reach the recording threshold of the material. And to make matters worse, the diffraction efficiency of the SLM is difficult to be improved as part of the incident power is wasted by the unwanted zero-order diffraction beam, higher order beam and DC noise [9]. On the other hand, the area occupied by the foci spots is restricted by the optical apertures of the optics in the system, especially the aperture of the objective. That is to say, when the area of the predefined pattern is too large, only the central part of them can pass through the optics and finally reach the sample.

IV. Summary

Multi-dimensional optical storage is demonstrated in photoisomeric film by adopting femtosecond laser holographic recording technology. Both storage capacity and recording rate are significantly improved without changing the objective NA and the laser wavelength. The two-photon induced isomeric orientation is studied. And some factors affecting the amount and arrangements of multi-foci patterns for holographic recording are discussed.

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