



# Two-photon three-dimensional optical storage of a new pyrimidine photobleaching material



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## ABSTRACT

Two-photon three-dimensional high-density optical storage is an important method of optical storage. Based on two-photon absorption three-dimensional optical information storage principle of light-induced bleaching, the storage information is written and readout by using titanium sapphire femtosecond pulsed laser, the experiment of the storage information written and readout is carried out in a novel photobleaching material pyrimidines, six layers storage information is achieved, information point spacing is 4  $\mu\text{m}$  and the information layer spacing is 12  $\mu\text{m}$ . The signal strength information of six layers information is recognized using MATLAB software. And the experiment of the stability of the pyrimidine photobleaching material is carried out, experimental results indicate that the material has good stability. The experiments show that the pyrimidine photobleaching materials can be used for two-photon three-dimensional optical storage, which has laid a solid foundation for multi-layer high density and high density optical information storage materials research.

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## 1. Introduction

Currently the information point size of the traditional two-dimensional optical disk has reached the limit because of the optical diffraction limit, and thus its storage density will reach the limit. To break through this limit, it is working to develop a short-wavelength small lasers and the objective lens of greater numerical aperture, such as blue-ray disk is appeared on the market. In addition there are some ways to increase storage density methods, such as spectral hole burning [1], holographic storage [2] as well as near-field optical storage [3], etc. but during practical obstacle is encountered relatively large.

Since in 1989 Rentzepis realized two-photon absorption technology for optical storage, two-photon technique that demonstrated technological superiority attracted the world wide attention [4–9]. This new type of two-photon three-dimensional high-density optical information storing method, relative to the two-dimensional optical information storage, increase axial

one-dimensional, which can achieve multi-layered information storage and increase the storage capacity significantly.

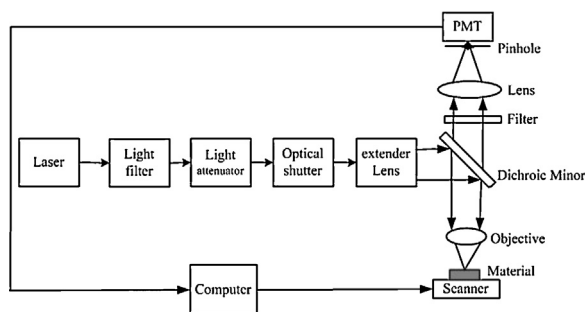
Kawata in Osaka University Japan used two-photon microfabrication technology to produce 10  $\mu\text{m}$  long, 7  $\mu\text{m}$  high dimensional bull graphics, which made two-photon femtosecond laser processing technology quickly become a hot topic [10]. According femtosecond laser two-photon absorption photopolymerization nanofabrication technology features and requirements, based on a set of self-built processing system, Sun Hongbo research group in Jilin University fabricated a high diffraction efficiency Fresnel zone plates which has a good performance on in the focus and imaging [11], and successfully fabricated fiber Bragg grating [12]. Because of the nonlinear two-photon writing process, two-photon absorption occurs only in the range near the focus of  $\lambda^3$ , which will not interfere with adjacent layers in the high storage density. In order to obtain a better two-photon three-dimensional optical storage materials, this paper carried out the experimental study that the storage information is written and read out in a novel pyrimidine photobleaching materials.

## 2. Photobleaching mechanism

Fluorescent dye polymer radiates fluorescence under the laser excitation, and fluorescence intensity increases with excitation

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**Fig. 1.** Optical setup for the writing and readout of two-photon three-dimensional bit information.

light intensity increased. After the excitation light intensity exceeds a certain value, the optical absorption will tend to saturate. If excitation light intensity is continued to increase, excited molecules will irreversibly be destroyed and photobleaching will occur. In addition, a sufficient exposure time will make the fluorescent dye lost fluorescence, photobleaching occurs.

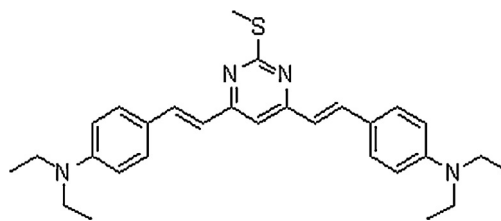
Photobleaching is an inherent property of fluorescent dye groups in the fluorescent microscope. In two-photon three-dimensional bleaching optical storage, photobleaching is used for optical information storage in the fluorescent dye doped polymer. In the bleaching process of two-photon excitation, when the excitation light intensity is low the radiation intensity of the fluorescence is proportional to the square of the incident light intensity. In the excitation of high-intensity light, dye of excitation point is bleached to get a bleaching point, bleaching point is no longer radiated fluorescence. In the optical storage bleaching point represents data "1" and the unbleached area represents data "0", which achieves information storage.

Two-photon bleaching excitation light is generally a pulsed laser, the pulse laser has a high power density that can achieve the fluorescent dye bleaching within a very short time, which can beneficially increase the speed of information storage and reduce the laser write power. When reading out the stored information, the weaker intensity two-photon laser or continuous laser is used for two-photon fluorescence excitation or single-photon fluorescence scanning, the recorded information does not emit fluorescence under the irradiation of the excitation light, and not recorded molecule fluoresces emit fluorescence, so detecting fluorescent light in the medium can read out the stored information.

### 3. Experimental systems and materials

Single-beam two-photon three-dimensional optical storage system is shown in Fig. 1. Ti:sapphire laser is the two-photon writing and reading light, whose center wavelength is 800 nm, pulse width is 80 fs, repetition frequency is 80 MHz.

When carrying out storage experiments, 800 nm pulsed laser through the filter, attenuation, and collimated beam expansion, through the objective lens (NA=0.65) is focused on the storage medium, PZT scanning stage make the storage medium for scanning movement, to achieve two-photon three-dimensional optical information storage under the control of the computer. When the reading experiment is carried out, through the filter, attenuation, and collimated beam expansion, through the dichroic mirror and the objective lens 800 nm pulsed laser is focused in the storage medium, optical switch is opened to scan, the fluorescence from the storage point is returned to the scanning optical system, and pass the dichroic mirror, the aperture stop to the photo multiplier tubes, photomultiplier tube signal is sampled by computer acquisition card, gray scale values provided by the computer can generate the storage light intensity image point by point in the monitor.



**Fig. 2.** Molecular structure.

This read-out method is a reflection confocal scanning read mode, which has a simple optical system and the high axial resolution of the storage layer, and can reduce the crosstalk of the storage layer generated by the storage medium and the substrate.

This experiment used a photobleaching material of 4,6-dimethyl-pyrimidine-based material, the molecular structure is shown in Fig. 2. The molecular structure has a D-A-D structure (D denotes an electron donor, A denotes an electron acceptor), 4,6-dimethyl-pyrimidine group is as a central core element, is derived to obtain curved molecules. After testing, two-photon absorption cross section of this material reaches 2278.6 GM.

In the high-intensity light irradiation, the two dipoles which connect N atom will absorb photons, the energy is passed to the middle pyrimidine ring, electron of a pyrimidine ring group is excited to an excited state, followed by electron radiate fluorescence and jump to the ground state. If the intensity of excitation light is too large, breakage of olefinic bond is likely to occur, the molecular structure is destroyed, energy absorbed by dipole can no longer be transferred to the pyrimidine ring, a molecule can no longer radiate fluorescence. Thus, we can write information that damaged points indicate "1", the no damaged point indicate "0".

The organic molecules were doped in the polymer using in situ polymerization method. The in situ polymerization method is better than directly doped polymer method, because the direct doping is difficult to find a suitable solvent to fully dissolve polymers, this will result in a doping sample inhomogeneity. In addition, costs of film production using in situ polymerization method were lower than direct mixing. Production methods are as follows: weigh organic molecules  $5 \times 10^{-5}$  mol, place in a small beaker, add 2 mL DMF to dissolve, filter small amount of insoluble precipitate, add the monomer PMMA (methyl methacrylate) 5 mL, and then add free radical initiator (azobisisobutyronitrile) 0.05 g, and free to initiate polymerization in heating or light conditions. We Heated for half an hour at 50 °C using heated water bath, then there will be more viscous state, remove and replace it with paper towels truss, form a solid after about a day.

Absorption spectra and fluorescence spectra of polymerization solid materials before and after exposure is shown in Fig. 3. Before exposure, 4,6-dimethyl-pyrimidine polymeric film has two broad absorption band, and has an absorption shoulder in the vicinity of 530 nm. This result is different from the absorption spectrum of 4,6-dimethyl-pyrimidine in a liquid. In the liquid state, 4,6-dimethyl-pyrimidine has only one absorption peak. At the liquid state, 4,6-dimethyl-pyrimidine are free molecules, the absorption peak is due to the absorption of the molecule itself. We make polymeric films using in situ polymerization method. When PMMA monomer is polymerized, the state of two branches of 4,6-dimethyl-pyrimidine molecule may be changed, so the torsion or bending happens, which lead to absence of the absorbent in other wavelengths. The fluorescence peak of 4,6-dimethyl-pyrimidine polymer film is at about 540 nm. Before and after exposure, absorption intensity and fluorescence intensity of the polymer film is difference in large, this property can be used for photobleaching storage.

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