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Invited Paper

Three-dimensional memory

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ABSTRACT

Three-dimensional optical memory with photorefractive materials is discussed for ultra-high density/capacity memory exceeding the classical limit of a conventional optical recording system. Bit data are recorded as highly localized refractive index variations in three-dimensional volume using a focused laser beam. We show recording and reading results using various recording materials and optical configurations. A multi-structured optical recording medium using a photoisomerization polymer and a transparent films has been developed for reflection confocal reading. Two-photon recording is also demonstrated.

Keywords: optical memory, confocal microscopy, two-photon absorption, photopolymer, hologram memory

1. INTRODUCTION

Optical memories such as compact disks (CD) and magneto-optical disks (MO) are becoming essential in high technology products such as audio and visual disks, and the external computer memory disk. In these memory devices a laser beam is used to record and read information. Since the laser spot can be focused to within 1 μm scale, optical memory can attain higher density and capacity than those of magnetic memory.

Optical memory is ultimately limited by the diffraction of electro-magnetic waves. Present techniques have almost reached this limit in optical memories that are commercially sold as compact disks or magneto-optic disks. Even with an infinitely large objective lens, the best achievable bit-data resolution distance for recording and reading is never smaller than half the beam wavelength.

To overcome the density limitation, multilayered optical memories have been investigated by many researchers.[1–6] In the memories the z or longitudinal axis is utilized in addition to the surface dimension ($x-y$ space) of conventional optical memory. The data are thus written not on the material surface but within the three-dimensional (3D) thick volume.

This approach requires an optical reading system that is able to read the data from a particular layer without cross talk between adjacent layers. It is also required to a recording technique on which it is possible to write information at a particular layer without erasing or corrupting the data already written at neighboring layers.

In this paper we describe the 3D optics as recording and readout systems of the multilayered optical memories using photorefractive materials. Some configurations of readout system are discussed and the use of two-photon absorption to reduce the cross talk between adjacent layers is also demonstrated.

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2. BIT-ORIENTED THREE-DIMENSIONAL MEMORY

Figure 1 shows a principle of bit-oriented three-dimensional (3D) optical memory. A laser beam is focused into a point in a recording medium. Chemical reactions of the medium should be induced at the spot because extremely high intensity is produced at the focus point. By 3D scanning of the focus spot in the medium we can record bit data in the medium in three dimensions.

Various materials such as photopolymers, photorefractive crystals, photochromic materials, polarization sensitive materials can store data. Among them, photorefractive recording mediums in which data are stored as refractive-index change are most promising materials in 3D memory because they have little absorption so the light penetrates in deep layer in the mediums.

Figure 2 shows a typical readout system of 3D memory. This is basically reflection confocal microscope configuration. Since the pinhole before a detector eliminated scattered light from out of focus region, so the system gives very good axial resolution and high contrast. The optical setup is very simple. We can also use other configurations, which are sensitive refractive-index variations, such as phase contrast microscope, differential phase contrast microscope, differential interference microscope, as readout system.[7,8]

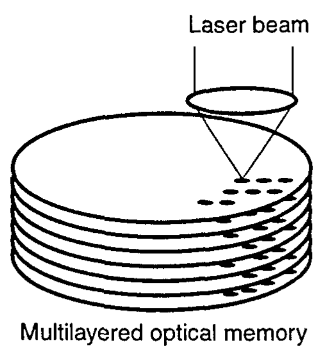

Figure 1. Principle of 3D memory

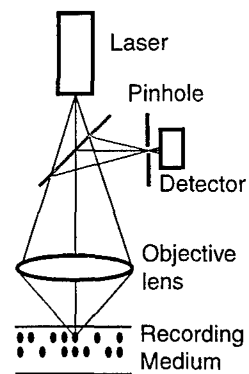

Figure 2. Reflection confocal readout system

3. PHASE-CONTRAST READOUT USING PHOTOPOLYMER MATERIALS

Figure 3 shows an example of 3D optical memory recording and reading using a photopolymer material. Bit data were written every 2 μm$\times$2 μm in a plane, and the longitudinal separation between the data planes was 10 μm. The total number of layers recorded for the optical memory shown in Fig. 3 was 30, which cover a total longitudinal length of 300 μm. The images were obtained using an ordinary phase-contrast microscope.

The photopolymer memory medium we used was monomer mixture composed of a methacryl compound and an allyl compound with benzil as an initiator and michler's ketone as a dye sensitizer.[9] The metacryl compound polymerizes faster than the allyl compound when illuminated with light. This causes the refractive index to increase from $n \simeq 1.5$ to $n \simeq 1.6$.

The photopolymerizable solution was sandwiched between a microscope slide and a microscope cover glass (thickness 170 μm) to prevent oxygenation of the photopolymer. The spacers between the slide and the cover glass was also made with cover slips of about 1 mm thickness.

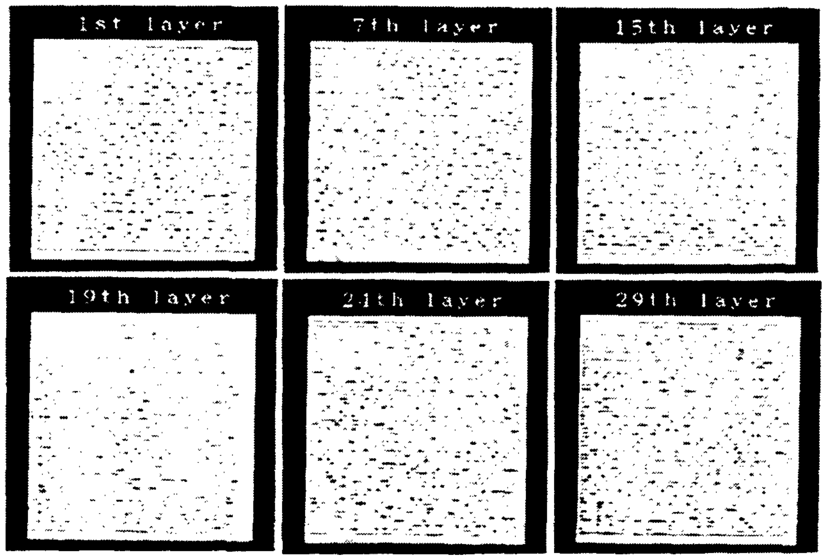

Figure 3. 30 layers recording and reading using photopolymer material

In order to write the data, the photopolymer is placed on a computer-controlled $x - y - z$ microscope stage. A 5 mW argon-ion laser, 488 nm wavelength, was used as the light source, which was focused onto the photopolymer by an objective lens (Carl Zeiss Axiophoto, NA=1.0, oil immersion, 40× magnification). An exposure time of 60 ms per point was used. At the focused spot, methacryl compounds photopolymerize because of their fast photopolymerization property. Allyl compounds, on the other hand, do not photopolymerize because the speed of photopolymerization of allyl compounds is slower than that of methacryle compounds.

As a result, only methacryl compounds are polymerized at the focused spot and the allyl compounds are pushed away, leaving a high-refractive-index region. We then record the data in the medium by scanning a focused spot in three dimensions with an exposure time of 60 ms at each data point.

We also developed a confocal type phase-contrast readout system. Figure 4 shows an optical setup of confocal configurations. This system is a laser-scanning confocal microscope appropriate for phase-contrast imaging. Point light source illumination reduces unnecessary scattered light because the point detector of a confocal microscope only detects the light intensity from a specific point of interest in the thick sample and rejects the scattered light produced from other non-focused points. High contrast images are therefore observed, and unwanted cross talk between planes is low. Better performance is obtained in comparison with the images produced using a conventional optical microscope. Spatial resolution is also improved because of the nonlinear frequency-response of the photorefractive materials.

Figure 5(a) presents an example of bit data which were read out using a confocal microscope, together with Carl-Zeiss Axiophoto. A He-Ne laser (632.8 nm) was used together with a phase-contrast objective and an annular pupil for phase-contrast (dark field) imaging. For comparison the same segment of the data was read using a conventional microscope with the same objective lens, and is shown in Fig. 5(b). The results demonstrate the advantages of confocal microscopy for high-contrast and high-resolution imaging of 3D structures. Since only the light intensity in the conjugate pair of the point of interest in the thick sample volume (or the focused point of the laser beam in the volume) is detected in a confocal microscope, scattered light produced by other non-focused points does not

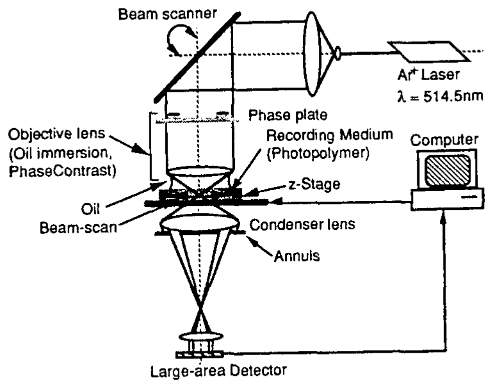

Figure 4. Optical setup of confocal type phase-contrast readout system.

contribute to the detected signal. Hence, the signal contrast of the images is excellent and the cross-talk between planes is negligible compared with images obtained using a non confocal microscope. Spatial resolution is also better because of the nonlinear spatial response (product of illumination point-spread functions and the detection amplitude point-spread function).

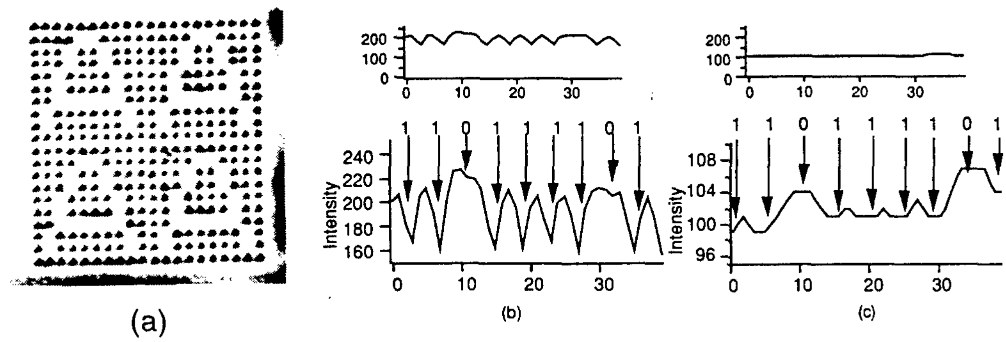

Figure 5. Readout result by using confocal phase-contrast microscope.

4. MULTI-STRUCTURED RECORDING MEDIUM FOR READING WITH REFLECTION CONFOCAL MICROSCOPE

Reflection-type confocal microscope has very high axial resolution, so it is a very attractive configuration as the readout system of multilayered optical memories. However, it is difficult to use the reflection-type confocal configuration as the readout system, because an extremely high numerical aperture (NA) lens is required for recording and reading.[10]

Some techniques have been proposed in order to read out data in multilayered memories with the reflection confocal configuration. Wilson et al. proposed the recording with 4Pi confocal configuration,[10] while Toriumi et al. proposed the use of two-photon recording and reading with longer wavelength light.[11]

We achieved the reflection confocal readout by using a recording medium in which photosensitive thin films and non-photosensitive films are piled up alternately.[12] We used urethane-urea copolymer film as the photosensitive material.

Figure 6 shows the spatial frequency distributions of bit-datum recorded with focused laser beam and coherent optical transfer function (CTF) of reflection type confocal microscope.[13,14] Figure 6(a) shows a spatial frequency distribution of bit datum recorded in very thick medium. This distribution coincides with the spatial frequency distribution of the focused light to record the bit data, because the bit is recorded with the focused beam. It is assumed that the NA of the objective lens is given by $n \sin \alpha$ and $k = 2\pi/\lambda$, where λ denotes the wavelength.

Figure 6(b) shows the spatial frequency distribution of bit-datum recorded in the thin layer of which thickness is same as the wavelength of recording light. Since the extension of the recorded bit in the axial direction is limited by the thickness of the photosensitive film, the spatial frequency distribution of the bit datum is much extended in the axial direction. The distribution is calculated by the convolution between the spatial frequency distribution of focused spot and the distribution of thin photosensitive film.

Figure 6(c) shows the CTF of reflection confocal microscope. As Wilson et al. pointed out,[10] the spatial distribution shown in Fig. 6(a) has no overlap with CTF of reflection confocal microscope unless we use extremely high NA lens.

The spatial distribution shown in Fig. 6(b) easily has overlapping area with the CTF, if we carefully select the thickness of the recording layer. As a conclusion, we can read the data with the reflection confocal configuration by using the medium piled up the thin recording layers and non-photosensitive transparent layers alternately.

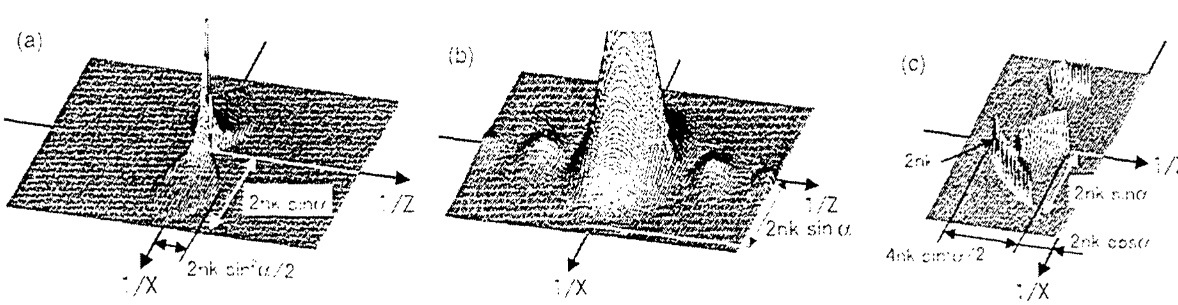

Figure 6. Spatial frequency distribution of the recorded bit datum recorded (a) in a thick medium and (b) in a thin layer. The thickness of the recording layer is the same as the wavelength of the recording light. (a) and (b) are truncated at an arbitrary value to show the structure in the high spatial frequency region. (c)CTF of the reflection confocal microscope.

We developed a recording medium in which photosensitive films and non-photosensitive films were coated on a glass substrate alternately. Urethane-urea copolymer was selected for photosensitive layers and polyvinyl alcohol (PVA) was used for transparent layers.

Figure 7(a) shows the chemical structure of urethane-urea copolymer.[15,16] The urethane-urea copolymer was originally developed for nonlinear optical waveguide. The copolymer has comparatively high optical nonlinearity and its stability at the room temperature. The copolymer has a side chain structure to semi-fix chromophores as a photosensitizer for optical memory use.

The absorption spectrum of the copolymer shows a maximum at 476.3 nm and little absorption in the region longer than 600 nm. By illumination of blue light, the azo-dye induces cis-trans isomerization producing refractive-index change. The recording process is photon-mode, so the recording time is expected as very fast response.

In generally, it is difficult to pile up two organic films alternately without influence each other. We select PVA as non-photosensitive transparent film, because water and pyridine as solvents for PVA and the urethane-urea do not dissolve the urethane-urea film and the PVA film, respectively. Figure 7(b) shows the multilayered recording medium we developed. The process of making the medium is explained as follows. The urethane-urea copolymer was solved in pyridine and spin-coated on a glass substrate. The thickness was less than 1 μm. After evaporation of pyridine solvent in an oven the PVA film was also spin-coated on the urethane-urea copolymer film. The thickness of the PVA film was about 8 μm. The urethane-urea copolymer was spin-coated again on the PVA film.

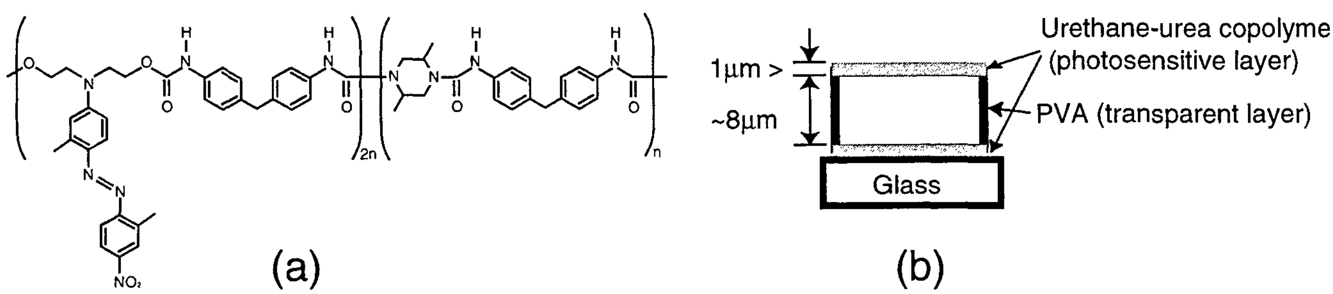

Figure 7. (a)Chemical structure of the urethane-urea copolymers and (b) the recording medium that we developed.

Figure 8 shows the optical configuration of recording and reading data system. An argon-ion laser (Ar^+) is used as a light source for recording. The wavelength of the laser is 488 nm and the output power is about 30 mW. The light is focused with an objective lens (NA: 0.65, 40×) into the recording medium. The medium is scanned with a computer-controlled three-axes stage. A shutter is also computer controlled in order to record the bit sequence in the medium. The power of laser light is adjusted with a neutral density filter. A white light source and a CCD camera is used to observe the recording process.

The readout system is a reflection-type confocal microscope. For reading a helium-neon (He-Ne) laser is used as a light source, because the urethane-urea copolymer has no absorption for red light. The scattered light at the recorded bit data is detected with a photo-multiplier tube (PMT). A pinhole of 30 μm diameter before the PMT eliminates the scattered light from the out of focus of the objective lens.

Figure 9 shows the axial distribution of the recorded data. The figure is reconstructed form a set of many images captured when the focus plane was changed. The three cross sections along the optical axis and along the first and second layers, respectively, are also shown in Fig. 9. The two recording layers are clearly detected. The bit data

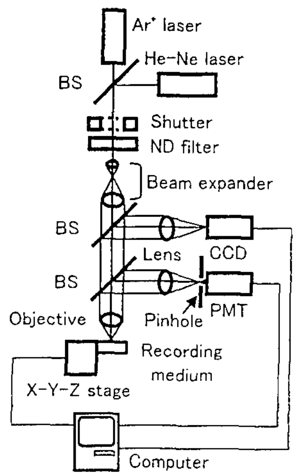

Figure 8. Optical system for recording and reading data of multilayered optical memory.

are also clearly recognized. The side-lobes in the cross-section along the optical axis is due to the aberrations of the objective. We may say that the reflection type confocal microscope configuration can be used as readout system of multilayered optical memory by using the recording medium in which the photosensitive thin films and transparent films are piled up alternately.

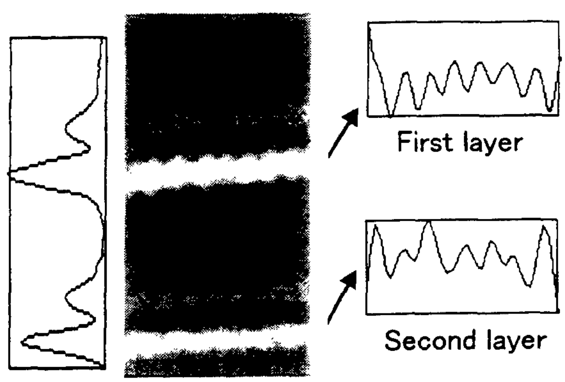

Figure 9. Axial distribution of the two-layer recorded data.

5. TWO-PHOTON RECORDING IN MULTI-STRUCTURED MEDIUM

Two-photon excitation is preferable in 3D optical memory because the crosstalk between adjacent layers is much reduced. Another advantage of two-photon excitation is reduction in multiple scattering. This reduction occurs because of the utilization of an illumination beam at infrared wavelength. A Ti:sapphire laser at 760 nm in mode-locked pulsed laser operation was employed as the recording light source. Since the probability of two-photon absorption is proportional to the squared intensity of the incident light,[17,18] photoisomerization can be induced only at the focal spot, where the intensity is very high. This technique is attractive for recording data in an erasable medium, because it is possible to write information onto a particular layer without erasing the data already written on neighboring layers.

Figure 10 shows a new type multi-structured recording medium. We have succeeded in developing four recording layers. This is very similar with the medium shown in Fig. 7(b) except for transparent layers. We used poly(methyl methacrylate) (PMMA) material with PVA as transparent layers, because it is difficult to coat thick (about 8 μm as shown in Fig. ??(b)) layer of PVA. PMMA is much easier to produce thick layer but it interferes with urethane-urea copolymer layer when it is spin-coated on the layer directly. To avoid such interfere we coated PVA thin film on urethane-urea copolymer layer in order to protect from PMMA.

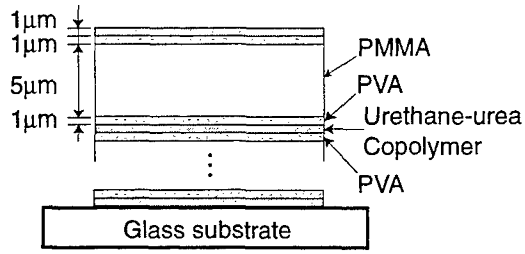

Figure 10. Multi-structured recording medium.

A Ti:sapphire laser was used for a light source of two-photon recording. The wavelength of laser light was 800 nm. Since urethane-urea copolymer has little absorption at the wavelength region longer than 600 nm, bit data should be recorded with two-photon process.

Figure 11 shows readout results of first, third, and fourth layers. Each layer were clearly read out with little crosstalk. In second layer, we failed to record data. We believe that the layer was damaged in the process of pile up other layers.

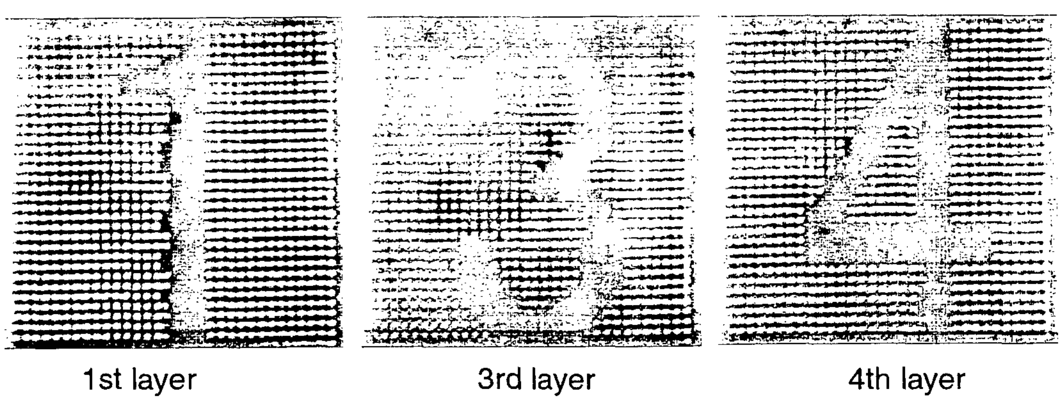

Figure 11. Recording results using two-photon absorption.

6. DISCUSSION AND CONCLUSION

We demonstrated multilayered optical memories in which three-dimensional optics was introduced into the readout system and writing system. The multilayered optical memories have clear advantages over holographic memory recording.[19] In terms of the signal-to-noise ratio, holographic memory is not robust to speckle noise unlike multilayered memory recording by the focused-beam scanning which is essentially imaging using spatially-incoherent illumination. Moreover, signal contrast in images using confocal imaging is better than that obtained by the hologram read-out because pinhole detection eliminates the scattered light that causes background noise. Moreover, random access bit recording and reading is possible only when using scanning optics.

The high density memory using three-dimensional optics does not conflict with other techniques for achieve high density: they are combined with each other. The multilayered memory is a just extension of z-direction of conventional optical memories, so scanning, tracking, and auto-focusing techniques in the conventional memory systems can be useful with some modifications.

We also presented the confocal readout of multilayered optical memory. For this purpose, the recording medium piled up the thin urethane-urea copolymer films and non-photosensitive transparent films was developed. We need more discussion on the optimization of the thickness of the recording layers and the transparent layers. The thinner film of recording layer much extends the spatial frequency distribution of bit in the axial direction, but it reduces contrast of bit data.

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