Mapping the Distribution of Refractive Index in Photopolymer Film with Scanning Near-field Optical Microscopy

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1. Introduction

Blue-Ray Disk is standardized as a new generation optical disc format with the lowest achievable wavelength and the highest NA that is achievable under industrial conditions. Future increases in density are possible by taking advantage of shorter wavelength lasers, higher lens numerical aperture. However, the method of shortening the wavelength of light reaches the limit.

Recently developed near-field optical techniques, the solid immersion lens (SIL)¹⁾ and the super-RENS²⁾ are utilized in a realistic demonstration of near-field optical data storage. These techniques have an advantage in the place which can use a present light source, but the improvement in read-out speed is not expectable. Several approaches for volumetric optical data storage have been researched. Three dimensional optical storage opens up another dimension to increase the capacity of storage media.

The hologram known as photograph technology for many years attracts attention as a bearer of high-density optical storage, because:

1. offers the possibility of storing 1 terabyte (TB) of data in a disk as CD and DVD.
2. As many as million bits can be read in parallel, compared to only one bit in conventional optical storage.
3. An illegal copy is not carried out, because a duplicate is difficult.
4. Compatibility may be able to be carried out with a pickup of the present CD and DVD.

It is realized by performing hologram record for a small volume or a multilayered waveguide. Former, volume hologram, greatly has an advantage, because tracking mechanism does not change in present CD and DVD. Besides media fabrication process is simple

compared to multilayered waveguide which needs UV embossing process³⁾ and inclination accuracy of core layer. We have developed photopolymerizable materials for recording phase holograms. It satisfies the many stringent criteria for a viable storage material including high dynamic range, high photosensitivity, dimensional stability, optical clarity, manufacturability, nondestructive readout, thickness, and environmental and thermal stability. The obtained holograms were colorless and the transmittances of holograms were about 90%⁴⁾. About the system of hologram record, the collinear system which OPTWARE Co.,⁵⁾ developed attracts attention. Unlike the conventional system which divides a beam into two, the collinear system has a big meaning in the place which simplified optical equipment composition and it can be miniaturized in the future. However, it needs for special optical set up and have to be evaluate the same way. In generally, we don't know the resulting difference between conventional two beam recording system and collinear recording system.

In this paper, we demonstrate a hologram recording to a small volume using microscopic reduced optical system and evaluate a refractive-index modulation produced in photograph polymer by scanning near-field optical microscope (SNOM).

2. Experiments

Table 1 shows compositions of photopolymerizable materials in this experiment. We have developed a diallyl phthalate prepolymer (DAPP) as prepolymer, and 9,9'-bis[4-(3-acryloyloxy 2-hydroxy) propoxyphenyl] fluorine (FDA) which has diphenyl fluorine form as a monomer, neopentyl glycol

Table 1. Compositions of the photosensitive mixtures

Function	Compound	Type-A /Wt%	Type-B /Wt%
Low photopolymerizable	DAPP :Diallyl Phthalate Prepolymer	47.78	57.33
High photopolymerizable	FDA :9,9'-bis[4-(3-acryloyloxy 2-hydroxy)propoxyphenyl] Fluorine	19.11	-
	A-NPG :Neopentyl Glycol Diacrylate	-	38.23
Plasticizer	DES : Diethylsebacate	28.67	-
Photoinitiator	HBI: Hexaryl bisimidazole derivatives	2.87	2.87
Photoinitiator assistance	Thiol-type compound	1.43	1.43
Photosensitive dye	CY : cyanine Dye	0.14	0.14

diacrylate as a monomer, diethyl sebacate (DES) as plasticizer, photoinitiator (HBI) that has imidazole form, and cyanine dye (CY) as photosensitizer. This photopolymer is advanced in transparency, and transmittance from 400-800 nm is more than 90%.

Sample holograms procedure is divided into three major efforts: (1) compounds solution was applied on PET films using applicator, (2) dried 10 minutes at room temperature under low vacuum oven with 100~300 Pa, and (3) it sticks to glass substrate using roller. The thickness of the photopolymer film is measured using a surface profiler (KLA Tencor :P-15 profiler) and determined to be $8\ \mu\text{m}$.

3. Results and Discussion

Figure 1 shows an optical set up for a hologram recording to a small volume using microscopic reduced optical system. Second harmonic generation of Nd:YAG laser (532 nm) is divided into two beams with $7\ \mu\text{W}/\text{cm}^2$. Both beams are led to an objective lens (Olympus : LMPlanFl 50X/0.50) attached to an inverted microscope. It being required for an optical system is so that the both beams completely overlap a single beam at the focus point. Transmission hologram was formed at focus point by scale of 1/50 of the beam size.

Figure 2 shows a CCD image of two-beam

Fig. 1. Configuration of a microscopic reduced optical system for hologram recording.

Fig. 2. CCD image of an interference pattern created from mutually coherent beams.

superposition by expansion observation. To show a coherence of the beam, it was observed in the top more slightly than a focus. Mutually coherent light from two beams creates an interference pattern. While photopolymer was photopolymerize within the area of beam spot, interference pattern forms a small structure of refractive index modulation on the photopolymer. The angle of two beams was about 3 degrees, so the period of interference fringes should appear in $10\ \mu\text{m}$ with parallel optical system. In this case, somewhat decreased period may be recorded with microscopic reduced optical system.

Figure 3 shows the image of volume hologram observed by differential interference microscope technique. About $40\ \mu\text{m}$ hologram recording spots were observed. It is assumed that distorted beam

Fig. 3. Differential interference microscope image of recorded hologram spots.

profiles caused hollow shape in each spot. Though Our photopolymer is able to produce $\Delta n=0.02$, it is not enough to observe in detail structure by differential interference microscope. We evaluate of this refractive index modulation by SNOM that allow us to observe it.

Fig. 4. Schematic diagram of SNOM system. Probe-sample distance control and sample scanning were carried out using an AFM controller. PD: photodiode, DL: Diode laser.

forms a surface relief after the PET film was removed in Type-A composition. As photopolymerization generates a stress in film, not a little, the stress eases after the PET film was removed. In the case of Type-B composition, the PET film is successfully removed from photopolymer without forming surface relief. This composition does not contain a plasticizer, so the film becomes hard firmly. Although Photopolymerization speed is a little inferior because of the lack of mobility, it is enough sensitivity as photopolymer.

Fig. 5. Topographic (left) and transmission SNOM (right) images of a recorded hologram spot.

Figure 4 shows a block diagrams of experimental set up for SNOM⁶⁾. Light from an Nd:YAG laser (532 nm) was coupled into the near-field probe for the same condition of hologram recording. Polarization of light can be controlled by combination of wave plate, and the light from aperture illuminate a sample surface. Then transmitted light through the sample is gathered by objective lens and is focused to photomultiplier. Circularly polarization used in this experiments. Optical fiber was sharpened up by chemical etching, and was coated by aluminum metal films with 350 nm. At the end of this fiber, optical aperture was formed. This small light source was rastered scanned by SPM technique, the light intensity through the sample forms an optical image (transmission SNOM image). The average aperture size of probe was 110 nm in these experiments. Evanescent field from the aperture decreases exponentially to distance. To maintain an aperture-sample distance is strictly constant, PET film on recorded photopolymer was removed. And the surface roughness of the sample influence to the SNOM image, because thick SNOM probe cannot trace to the rough surface. In some case, SNOM image contains actually optical property and topographic artifact. It turned out that photopolymer

Figure 5 shows AFM and transmission SNOM images of recorded volume hologram (showed in Fig 4) observed at the same time. AFM image describe that the surface is smoothly flat without some dusts and dips. The difference of optical property from material was not appearing in topographically. SNOM image shows bright circle shape. Transmission SNOM image was consists of light intensity at each pixel. If there are absorbing components on/in the film, such area becomes dark in the image. A very small amount of dye was completely breached by UV light after recording, so the image shows slight difference of refractive index after recorded. The aperture size is enough small to the period of interference fringes, periodic structure of refractive index modulation does not work as a grating for the small light source. If the probe is located on near boundary between higher refractive index and lower refractive index, the light wave slightly bends toward the higher refractive index (higher density). High N.A. objective lens in front of the photomultiplier focuses on the aperture, so the loss amount of light makes contrast in SNOM image. Since a clear boundary does not necessarily exist in the film, it can be expected that bright and dark do not appear clearly, either. The reflection light produced by the interface of glass and air

is considered to make the complicated pattern in the SNOM image.

4. Conclusion

We demonstrated a hologram recording to a small volume using microscopic reduced optical system for a high-density optical memory. Transmission hologram was formed in a spot size with $40\ \mu\text{m}$ in diameter. Our photopolymer is mainly consists of diallyl phthalate prepolymer and forms refractive index modulation with $\Delta n=0.02$ in films. Detail structure of Interference pattern is difficult to observe in conventional microscopic technique, so we evaluate it using near-field optical microscopy (SNOM). Δn imaging of photopolymer for recording holograms was obtained from the spot.

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