Holographic grating fabrication for wide angular bandwidth using polymer thin films^{*}

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To increase the angular bandwidth of volume holographic grating, we fabricate holographic gratings based on grating multiplexing technique by using thin films of photopolymers and polymer dispersed liquid crystals. Experimental results confirm that the liquid crystal materials increase the refractive index modulation of the grating, enabling high diffraction efficiency with wide angular response compared to pure polymer materials. We observe that the fabricated holographic grating has near 80% of diffraction efficiency and about 18° of angular bandwidth, which can be further improved by modifying the liquid crystal/polymer mixtures and the grating multiplexing technique. The grating can be used to fabricate holographic waveguide structures for emerging applications in the near-eye display systems.

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Augmented reality (AR), a new technology that integrates real-world environment with virtual-world information, is receiving much attention in recent years due to its broad applications and significant impact on information technologies. Near-eye display (NED) optics is one of the key components for AR applications, acting as an interactive interface to display a virtual scene into the real world. Many different optical techniques have been exploited for NED applications including combining optics^[1-3], waveguides^[4-8], and retinal projectors^[9,10]. In terms of compactness and light weight, the waveguide-based method is one of the best candidates for implementation of optical see-through head-mounted display applications. Several industrial giants including Sony^[11], Microsoft^[12] and Facebook^[13], have invested significantly in research and development in related technology. However, the small field of view (FOV) has limited the applications of the waveguide-based NEDs. The grating element coupling light into and out of optical waveguides is one of the key elements to determine the FOV in the holographic-waveguide-based AR systems.

In this article, we investigate several factors that affect the diffraction efficiency and angular response of volume holographic gratings. First, we synthesize a new photopolymer material to fabricate gratings and study the effects of monomer concentration on the diffraction properties. Then, we further introduce liquid crystal into the polymer system and fabricate thin films of polymer dispersed liquid crystal (PDLC) for grating multiplexing to improve the angular bandwidth.

Photopolymer consists of several major components including monomers, film-forming resins, initiators, and photosensitizers.

In our work, we use acrylamide (AA) as monomer, N, N'-methylene-bis-acrylamide (BMAA) as a crosslinking monomer, yellowish eosin (YE) as a photosensitizer, triethanolamine (TEA) as an initiator, and polyvinyl alcohol (PVA) as a binder^[14]. In the preparation process of PVA/AA photopolymer materials, it is generally preferred to dissolve the monomer, initiator, photosensitizer in deionized water. The binder needs to be added with water and then dissolved at 80 °C in a magnetic stirrer. The mixture needs to stand for a certain period of time before the film can be prepared. In order to control the thickness of the film, we prepare PVA/AA photopolymer material by spin coating method with different spinning speeds. The prepared mixed viscous solution is spin-coated on a transparent glass substrate, and then placed in a clean and dark room, usually from 12 h to 24 h. After the water in the material evaporates to a certain degree, a photopolymer film sample is formed.

Based on the synthesis of PVA/AA photopolymer material, we further introduce the liquid crystal material to fabricate the films of PDLC composites. For this material preparation, the binder is no longer needed at this time. In the preparation process, the monomer, the initi-

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ator, and the photosensitizer are dissolved in deionized water, filtered, and then added to the liquid crystal according to a mass ratio of 1:1. The PDLC is sandwiched and formed between two transparent glass films. The preparation process is shown in Fig.1.



Fig.1 Preparation procedures of the sample films

The recording process of the grating is shown in Fig.2. The reference light and the object light impinge onto the sample to form an interference pattern. The monomer in the dark fringes moves toward the bright fringes where the polymerization process takes place to produce a holographic grating of refractive index gradient. After the grating is successfully recorded, we use 632-nm laser as the probe light and measure both the transmitted light intensity and the diffracted light intensity of the grating to calculate the diffraction efficiency.



Fig.2 Schematic diagram for recording gratings

According to Kogelnik's law^[15], the period of gratings Λ can be expressed as:

$$\Lambda = \frac{\lambda}{2n\sin\theta},\tag{1}$$

where λ is the wavelength of recording beam, *n* is the average refractive index of the recording material and θ is the half-angle between the two interference beams interacted on the gratings.

The diffraction efficiency is defined as^[16]

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$$\eta = \frac{I_1}{I_0 + I_1},\tag{2}$$

where I_1 and I_0 represent the first-order diffracted and transmitted light intensities, respectively.

The diffraction efficiency is defined as^[17]

$$\Delta n = \frac{\lambda_{\rm p} \cos\theta \sin^{-1}(\sqrt{\eta})_{\rm l}}{\pi d},\tag{3}$$

where λ_p is the wavelength of probing beam, and *d* is the thickness of sample.

Fig.3 shows the experimental setup. A 532 nm laser is used to record the diffraction gratings. The diameter of the laser beams is expended to 2 cm, and then divided into two secondary beams with an intensity ratio of 1:1. The incidence angle of the reference light and the object light can be adjusted by rotating the mirrors and varying the sample location. The time of recording is controlled by the shutter and electronic timer. The shutter can control the opening and closing of the light path. The shutter is connected to electronic timer, which can set the exposure time in advance. We monitor the diffraction grating using a 633 nm laser beam. The photo sensitizer does not absorb light in red spectral band. After the first recording, the sample was rotated to record the second grating. We can change the angle of the probe light by rotating the sample to monitor the angular response around the Bragg condition.



Fig.3 Experimental setup for recording holographic gratings

As we have known, the diffraction efficiency and angular bandwidth of the grating are primarily related to the thickness of the sample and the degree of refractive index modulation^[18]. Theoretically speaking, the larger the refractive index modulation results in the higher the diffraction efficiency and the wider the angular range. The thicker the sample also causes the higher the diffraction efficiency but the narrower the angular range. In the photopolymer system, we investigated the effect of monomer concentration in the polymer on the grating quality. In this study, we prepared photopolymers with different monomer concentrations. In the film-coating process, the spin coating is set to the same speed and acceleration. Also, the gratings are recorded with the same experimental condition. The recording time of every grating is 10 s, and the diffracted light intensity and the transmitted light intensity satisfying the Bragg WU et al.

condition are measured by a photodetector. As for single-recording grating, the time of 10 s is enough to consume most of the monomers. If the time is longer, the diffraction efficiency of the grating is not improved. But if the time is too short, the monomers cannot be sufficiently polymerized, which will reduce the diffraction efficiency. Tab.1 shows a list of ingredients for different material composites.

Tab.1 Composition table of different concentrations of monomers (Unit: g)

Material Proportion	AA	TEA	BMAA	YE	PVA
18.6%	1.623	1.721	0.373	0.01	5
21.8%	1.980	1.721	0.373	0.01	5
24.1%	2.256	1.721	0.373	0.01	5
27.8%	2.735	1.721	0.373	0.01	5
30.1%	3.059	1.721	0.373	0.01	5
32.9%	3.483	1.721	0.373	0.01	5
35.2%	3.859	1.721	0.373	0.01	5

As displayed in Fig.4, within a certain range, we observe that the diffraction efficiency of the grating increases with the increase of the monomer concentration. An increase in the monomer concentration results in an increase in the polymer concentration in the bright region. As such, the refractive index gradients in the bright and dark regions are enhanced, thereby the diffraction efficiency of the grating is increased. However, when the monomer concentration exceeds 30.1%, the diffraction efficiency does not increase further. This is because the monomer in the photopolymer has reached the maximum of its concentration. If more monomer is added, the monomer will be precipitated after the water evaporates from the film sample. In this situation, the sample is no longer transparent, and the grating is difficult to be formed. Based on our study, the optimum monomer concentration of the polymer is about 30%.



Fig.4 Diffraction efficiency of different monomer concentrations

From the above experiments, even with the monomer concentration increased, we found only 28.1% of the diffraction efficiency can be obtained. We found that the diffraction efficiency of PDLC grating is greatly improved by adding liquid crystal into the polymer through increasing the refractive index modulation. The liquid crystal can be dissolved in the polymer without affecting the properties of the polymer. After the exposure, refractive index difference is formed between the polymer aggregate and the liquid crystal aggregate. And because of the electro-optical effect of liquid crystal, we can control the orientation of the liquid crystal molecules and change the refractive index by applying a driving voltage, so as to control the state of the grating. The electrical properties of PDLC gratings will be our next research and will not be discussed in this article. Therefore, we add liquid crystal to the polymer systems and fabricate the PDLC gratings, to further increase the refractive index modulation and thereby the diffraction efficiency and the angle selective range. The liquid crystal used in this experiment is BHR40300, which is produced by Beijing Bayi Space LCD Technology Company. The temperature of clearing point is 81 °C. During the entire experiment, the temperature was lower than 81 °C, so it was always in the liquid crystal state.

After the addition of the liquid crystal, the quality of the grating is significantly improved, the diffraction efficiency is up to 78%, and the full width half maximum (*FWHM*) of the first-order Bragg diffraction peak is also significantly increased. The *FWHM* of the photopolymer grating is about 5°, and the *FWHM* of the PDLC grating is about 10°. According to Eq.(3), the refractive index modulation of the photopolymer grating is about 0.018, and the refractive index modulation of the PDLC grating is about 0.023.



Fig.5 Experimental results of diffraction efficiency as a function of angular bandwidth

In our study, we also conduct experiments to form multiplexing gratings by using multiple-exposure method. After the first exposure, we rotated the sample to 5° and then record the second one. In order to make full use of the monomer, we still control the total exposure time to 10 s. We tried to set the every exposure time is 5 s, and we found that the diffraction efficiency of the second grating is significantly lower than that of the first grating. Because the monomer concentration at the second exposure was significantly lower than that at the first expo• 0004 •

sure. Therefore, we reduce the first exposure time, we set the first exposure time as 4 s and the next exposure time as 6 s.

The Bragg angles of the two gratings are different. When the diffraction peaks of the two gratings partially overlap, the angular response range of the grating is extended. As shown in Fig.6, the multiplexing grating has much wide *FWHM* compared to the single-exposure grating. The *FWHM* of the photopolymer grating is about 9°, and the *FWHM* of the PDLC grating is about 18° for multiplexing only two gratings.



Fig.6 Experimental results of diffraction efficiency as a function of angular bandwidth by multiplexing two gratings

In experiments, we have also recorded an image of a Chinese letter by using our sample films to investigate the quality of the image reconstruction from the holographic grating. The diffracted image and the transmitted image of the grating are shown in Fig.7. From the figure we can see that the grating has good image quality, which is important for the NED applications.



Fig.7 Image reconstruction from recorded holograms in a PDLC film

In summary, we have synthesized the PVA/AA pho-topolymers and the PDLC composites for holo

graphic recording applications. We have investigated the diffraction efficiency and angular bandwidth by using these sample films. It is found that the addition of the liquid crystal enables improvements of not only the diffraction efficiencies but also the FWHM of the grating angular response due to the enhancement of refractive index modulation. In addition, we have further performed the grating multiplexing experiments by using multiple exposure method. Experimental results show that the FWHM of both photopolymer gratings and PDLC grating can be significantly improved.

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