Direct Fabrication of Surface Relief Holographic Diffusers in Azobenzene Polymer Films

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We propose a direct fabrication of light diffusers using azobenzene polymer films. Holographic recordings of surface relief diffusers are formed on the polymer film by exposure to an Nd:YAG laser (532 nm) through source diffusion materials. No post-treatment is needed, and it can be erased by heating or irradiating uniform laser beam. Diffusion pattern can be controlled by the polarization of the laser. Transmittance of over 88% is obtained at the wavelength of 633 nm. © 2005 The Optical Society of Japan

1. Introduction

Light diffusers have many applications, including computer displays, signs, and lighting systems to increase contrast ratio or to control an illumination pattern of a light source. A ground glass and a plastic containing a particulate scatterer are often used as light diffusers. Several studies of such diffusion have been conducted.[1,2] It is difficult to obtain a uniform scattering intensity over a defined viewing angle. Holographic diffusers are useful to make desired scattering patterns.[3] It is possible to control diffusion angle by selection of the properties of the source diffuser, by control of its subtended angle, by selection of the holographic medium and so on. Evaluations of holographic diffusers made of various photoinduction materials, for example, silver halide material, are reported by Kim *et al.*[4] Silver halide materials and photopolymers are often used as holographic mediums.[5] No pre or post-treatment is needed, and the hologram can be rewritten.

Azobenzene functionalized polymers are one of the most promising materials for holographic medium. The recording of polarization holographic gratings using azobenzene-containing polymer film as photoanisotropic materials has been reported.[6] The photoinduced anisotropic effect is due to *trans–cis–trans* isomerization and the orientational effects of the azo-dye chromophore. To record a polarization holographic grating, two mutually orthogonal linear polarization beams and a relatively thick film of over 10 μm is necessary. On the other hand, the direct recording method of surface relief structures on thin azo polymers has been reported[7,8] in which the structure is recorded by means of a spatially varying intensity distribution combined with a spatially varying polarization distribution.[9] There have been many discussions about the mechanism of this phenomenon, and some models have been suggested.[10–13] The mechanism of this process involves repeated *trans–cis–trans* photoisomerization and large-scale molecular motions and volume changes caused by the orientation of the azobenzene

moieties occur simultaneously. Large-amplitude and a stable surface relief structure were produced upon exposure to an interference pattern of polarized laser beams at modest intensities without any subsequent process. The fabrication beam power is a few tens mW/cm^2 and the fabrication temperature is below the glass transition temperature (T_g). The structure could be erased by heating the polymer film above its T_g, and the writing and erasing cycles could be repeated. Application for a grating coupler,[14] an electrically addressed diffraction device[15] and a guided mode resonance filter[16] were reported. From above-mentioned reasons, we chose to use the azobenzene polymer film to fabricate a holographic diffuser.

In the present letter, we propose a direct fabrication of surface relief holographic diffusers. Holographic recordings of these diffusers are formed on the polymer film by exposure to Nd:YAG laser (532 nm) through source diffusion materials.

2. Experimental Procedure

The side-chain azo-polymer, poly-orange tom-1 isophoronediisocyanate is used in this study. Figure 1 shows the chemical structure and absorption spectrum of the material. The concentration of dye is over 50 wt% and T_g is 136°C. The absorption peak and the cut off wavelength of the dye are 440 and 560 nm, respectively. This polymer is dissolved in cyclohexanone. Samples of 3 μm thickness are prepared by spin-coating on a slide glass plate. Refractive index of the film is measured as 1.65 at the wavelength of 633 nm by m-line technique.

The experimental setup for making a holographic diffuser is shown in Fig. 2. An Nd:YAG laser at the wavelength of 532 nm is used as a laser source. The laser beam is collimated to 1 cm in diameter, and input to a source diffuser. In this experiment, light diffuser LSD20PC10 (Physical Optics Corp.) is used as a source diffuser. Speckle pattern of the diffused light is recorded on the polymer film. Control of the angular range of diffusion is achieved by
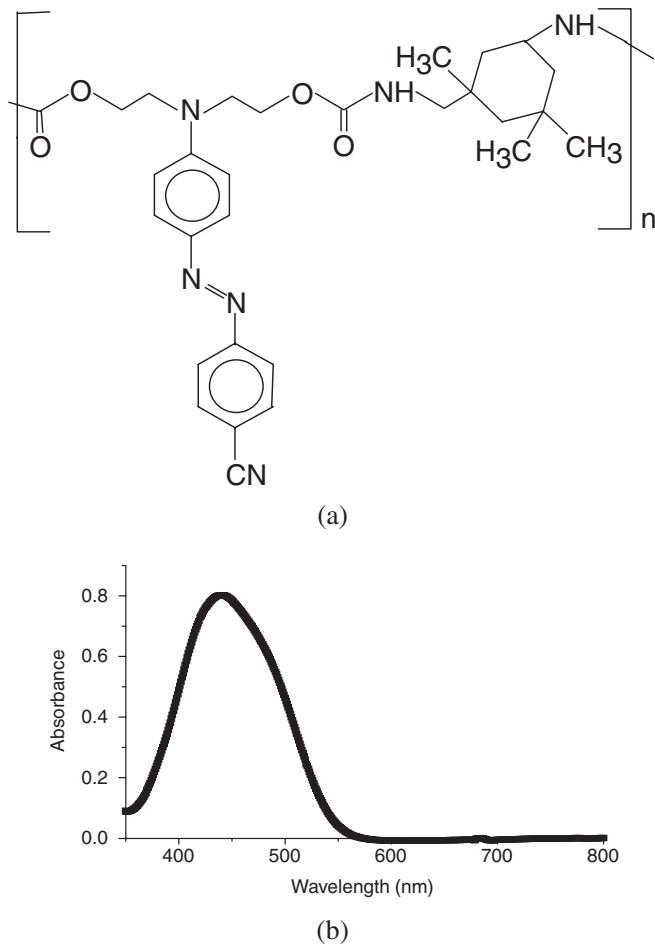
(a)

(b)

Fig. 1. (a) Chemical structure and (b) absorption spectrum of poly-orange tom-1.
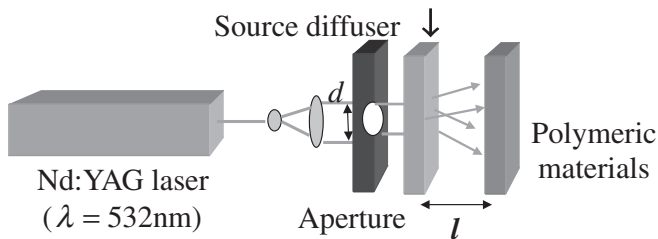

Fig. 2. Experimental setup for making a holographic diffuser.

placing the polymer substrate some distance back from the holographic plate. The distance was fixed to 1 cm in this study. Holographic diffusers can be recorded in proportion to the gradient of the light intensity as surface relief images without any subsequent process. We do not need a darkroom to record surface relief structures because the image is not exposed in visible light.

Figure 3 shows the time dependence of zero order beam ratio when a circularly polarized laser beam at 532 nm of 200 mW/cm^2 is irradiated through the source diffuser. The zero order beam ratio is a reverse measure of diffusion efficiency,[5] diffusers differ in their ability to diffuse light as
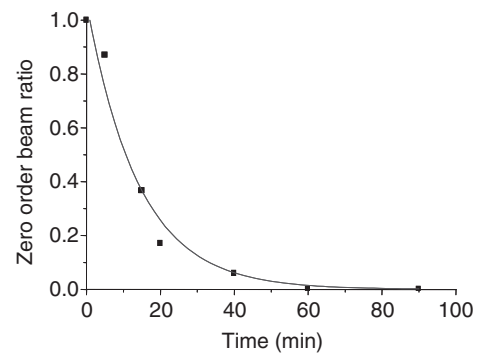

Fig. 3.   Time dependence of zero order beam ratio.

quantified by this ratio. Diffusers with a high zero order beam ratio are partially transparent, whereas those with a small zero order beam ratio appear white. The ratio decreases exponentially, and a zero-order beam ratio of less than 0.001 at the wavelength of 633 nm is obtained after 90 min. This value is much smaller than that of a silver halide material (zero-order beam ratio; 0.05 – 0.5), and is as small as that of ground glass and a photopolymer. Total transmittance over 88% is obtained at the wavelength of 633 nm.

We measured the diffuse transmittance of the polymer film. A He–Ne laser of 633 nm is input normal to the sample. Diameter of the laser beam is about 1 mm. A laser power meter is used to measure the intensity of the diffused light, and it is placed 3 cm behind the sample. Figure 4(a) shows the diffuse transmittance versus angle for the source diffuser at the wavelength of 633 nm. It is a circular diffuser, and the diffusion angle of two orthogonal directions is about 20°. The diffusion angle is defined as the angle of half the maximum intensity. Figure 4(b) shows the diffuse transmittance versus angle for an azobenzene light diffuser. A solid line shows the diffuse transmittance in the horizontal direction, and the dotted line shows that in the vertical direction. Diffusion patterns for the horizontal and vertical directions are almost the same and a diffuse angle of about 26° is obtained. Compared with the source diffuser, the diffuse angle is increased in the recording process.

Figure 5 shows a typical example of the three dimensional view of the surface relief diffuser observed by an atomic force microscope. A random surface relief structure with large modulation depths of over 2 μm is observed. There is large polarization dependence when we make a surface relief structure. When the film is exposed to p-polarized light, significantly larger surface relief is fabricated than when the film is exposed to an s-polarized beam. Figure 6 shows the diffuse transmittance versus angle for a polymeric diffuser at the wavelength of 633 nm using a linear polarization (horizontal direction) for the recording laser beam. All the experimental conditions are the same as the previous one except the laser polarization. The diffusion angle is wider in the direction of writing polarization of the laser. A different diffusion pattern can be made by using a different polarization for the recording laser beam. Diffusion patterns are
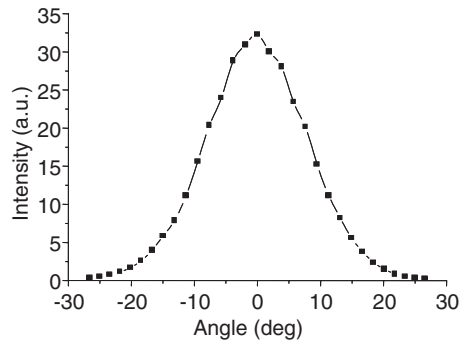
(a)
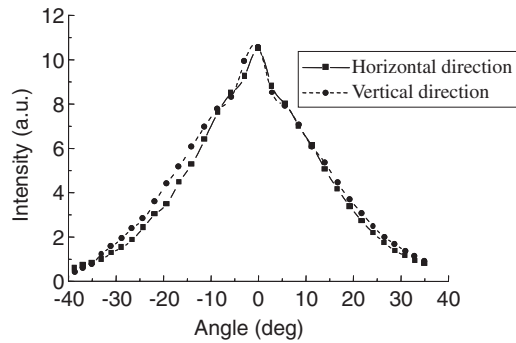
(b)

Fig. 4.　Diffuse transmittance versus angle for (a) source diffuser and (b) polymeric diffuser.
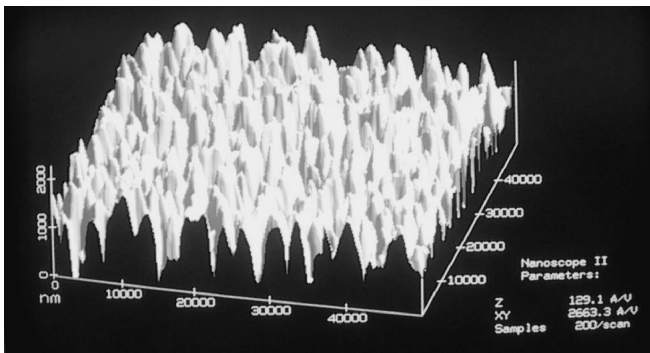

Fig. 5.　Three-dimensional view of the surface relief diffuser.

also controlled by its subtended angle, by the aperture size, and the source diffusion materials.

These surface relief structures are erased by heating above T_g or irradiating a uniform laser beam. Figure 7 shows a photo of diffusion patterns (a) before recording, (b) after recording and (c) after erasure when the He–Ne laser is incident to the sample. The recording pattern is erased after heating at 140°C for 30 min. The writing and erasing cycles can be repeated.

3.　Conclusions

We have proposed a direct fabrication of light diffusers using azobenzene polymer film. Holographic recordings of surface relief diffusers are formed on the polymer film by
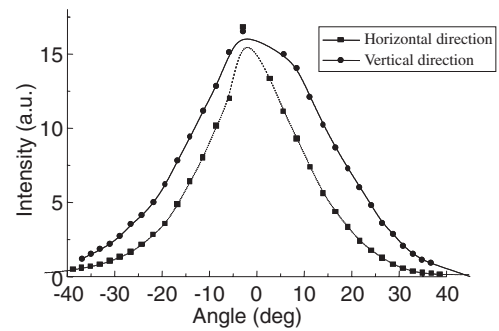

Fig. 6.　Diffuse transmittance versus angle for polymeric diffuser at $\lambda = 633$ nm using a linear polarization as a recording laser beam.
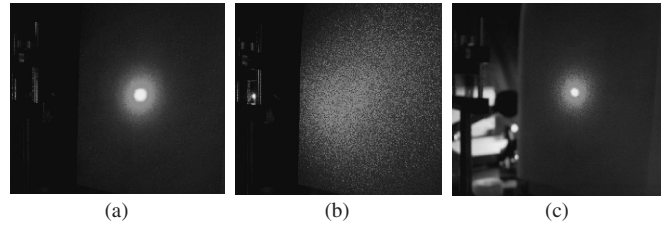

Fig. 7.　Diffusion patterns (a) before recording, (b) after recording and (c) after erasure.

exposure to Nd:YAG laser (532 nm) through source diffusion materials without any subsequent processes. A random surface relief structure with large modulation depth of over 2 μm is fabricated. This diffuser has strong absorption in the visible region shorter than 600 nm, but the zero-order beam ratio of less than 0.001 is obtained at the wavelength of 633 nm. Transmittance of over 88% is obtained at the wavelength of 633 nm. Higher transmittance can be expected with an antireflection coating on glass plates. Diffusion patterns are controlled by the polarization of the writing laser beam; they are also controlled by its subtended angle, the aperture size and the source diffusion materials. The relief structure can be erased by heating or irradiating a uniform absorption laser beam. We have already proposed a replication of holograms using azobenzene polymers.[17] By using the technique, it is expected that the transparent holographic diffuser in visible region is fabricated. We have also reported the diffraction efficiency increase by corona charging in photoinduced surface relief gratings on an azo polymer film.[18] By charging the material, an electrically induced change of diffusion pattern is expected. This is the easiest way to make holographic diffusers. They can be applied as new types of functional holographic diffusers.

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