

## Application to Optically Controlled Spatial Light Modulator Using Organic Photorefractive Composite

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**ABSTRACT:** Optically controlled spatial light modulator was demonstrated as application of photorefractive polymeric material. Photorefractive composites based on photoconducting carbazole-substituted polysiloxane matrix (PSX-Cz) show an excellent stability against phase separation, compared to poly(*N*-vinyl carbazole) based composite. 2-[3-[(*E*)-2-(piperidino)-1-ethenyl]-5,5-dimethyl-2-cyclohexenylidene] malononitrile (P-IP-DC) has large dipole moment and large polarizability anisotropy. A photorefractive sample in which a chromophore (P-IP-DC) was dispersed showed a high diffraction efficiency of 85% at an external field of 28 V/ $\mu\text{m}$ . Application of photorefractive material for the optically controlled spatial light modulator which can convert incoherent optical image imposed in Xe-lamp light into coherent image in He–Ne laser wavelength is demonstrated using this composite. [DOI 10.1295/polymj.36.674]

**KEY WORDS** Photorefractive Polymer / Spatial Light Modulator (O-SLM) / Two-Beam Coupling /

Photorefractive effect refers to a spatial modulation of refractive index of materials which possess both photoconductivity and optical nonlinearity simultaneously.<sup>1,2</sup> As two coherent beams intersect a photorefractive material, the internal space-charge field is formed due to redistribution of photogenerated charges, which subsequently modulate refractive index of the material *via* an electro-optic effect. The polymeric photorefractive materials with low glass transition temperature exhibit two attractive features, the large refractive index modulation (up to the order of  $10^{-2}$ ) and the optical erasability of recorded holographic information. These advantages make the organic photorefractive material to become one of the most promising candidates for optical information processing, *e.g.*, pattern recognition and fingerprint verification.<sup>3–5</sup>

Especially, photorefractive polymeric composites with carbazole-substituted polysiloxane matrix (PSX-Cz) show low glass transition temperature ( $T_g$ ) and excellent stability against phase separation, comparing to poly(*N*-vinyl carbazole) (PVK) that has been most widely used as photoconducting matrix in the photorefractive composite. The former reduces amount of the plasticizer used for lowering the glass transition temperature of the composite to the room temperature. The latter enables to use almost all kinds of chromophore in the preparation of photorefractive composite and enhances long term stability.

There are two representative factors, the magnitude of space-charge field and the electro-optic coefficient, in the steady state performance of the photorefractive

material. To achieve the higher diffraction efficiency or gain coefficient of the material, the higher magnitude of space-charge field and electro-optic coefficient are necessary. The measurement method for magnitude of the space-charge field was proposed for the first time by Moerner *et al.* in 1994.<sup>8</sup> So far the diffraction efficiency (or gain coefficient) was mainly discussed to evaluate the steady state performance of the photorefractive material, whereas the magnitude of space-charge field was not often used in spite of its importance. It is due to the fact that measurement method is not simple and requires lots of time. In 2002, we proposed a new characterization method for the space-charge field that is more simple and reliable. In contrast to the previous method where the electro-optic coefficient,  $r_{13}$  and  $r_{33}$ , should be independently determined, the new method requires only the birefringence at various poling angles and poling fields so that the measurement can be simply carried out. From the magnitude of space-charge field, we can estimate the trap density in a sample, which is significant information.

Spatial light modulators (SLMs) are devices that impose a given image on an optical wave by spatially modulating its intensity and/or phase distribution.<sup>6</sup> There are two classes in SLMs; one is electrically controlled SLMs, and the other is optically controlled SLMs. They are classified by the way the image is provided to the device. In the former case, electrical signals, representing the information to be input to the device, controls the spatial distribution of transmission or phase shift of the device. In the latter case,

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the information to be input to the device is imposed on the optical wave instead of electric signal. Using optically controlled SLMs, incoherent images can be converted into coherent images, and images in the infrared can be switched into images in the visible. Since it is difficult to carry out optical data processing such as holographic recording and pattern recognition with incoherent image data, optically controlled SLMs are very useful in various optical application fields.<sup>6</sup>

In this work, we presented the steady state performance of our photorefractive material with the diffraction efficiency. Two factors, a magnitude of space-charge field and a field-induced birefringence, dominantly determining the steady state performance of sample were measured at various applied fields. To demonstrate optically controlled SLMs using our material, an incoherent image imposed in Xe-Lamp light was converted into coherent image in the coherent light from He-Ne laser.

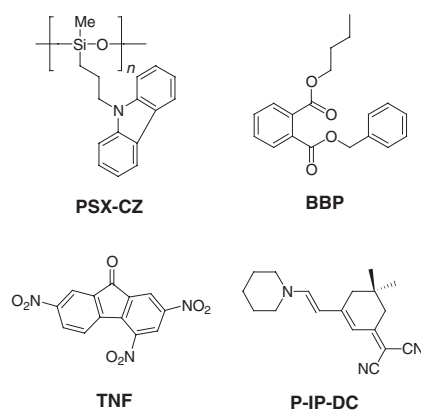
## EXPERIMENTS

### Materials and Sample Preparation

2-[3-[(*E*)-2-(Piperidino)-1-ethenyl]-5,5-dimethyl-2-cyclohexenylidene] malononitrile (P-IP-DC) was synthesized as a nonlinear optical chromophore with large dipole moment and polarizability anisotropy. 2-(3,5,5-Trimethyl-2-cyclohexenylidene) malonitrile (10 g, 0.0536 mol) was dissolved in 1-(dimethoxymethyl)-piperidine (20 mL).<sup>7</sup> The mixture was stirred at ambient temperature for 30 min and then 110 °C for 3 h under nitrogen atmosphere. After the reaction mixture was cooled to room temperature, the volatile components were removed using a rotary evaporator and the dark-red residue was obtained. The reaction product was dissolved in a mixed solvent of methylene chloride (10 mL) and methanol (150 mL). After the solution was concentrated and then cooled to 5 °C, the reaction product was precipitated on standing. The crude product filtered and then recrystallized from hexane-ethanol.

Polysiloxane with carbazole pendant (PSX-CZ) sensitized by 2,4,7-trinitro-9-fluorenone (TNF) was used as a photoconducting matrix due to its excellent physical properties. PSX-CZ shows good compatibility with organic materials and high optical clarity.<sup>7</sup> Its  $T_g$  is around 51 °C and  $T_g$  of the composite can be lowered to room temperature by adding 10 wt % of liquid plasticizer, butyl benzyl phthalate (BBP). The chemical structures of the molecules of which the photorefractive polymer composites are consisted are shown in Figure 1.

Since electro-optic (EO) behavior of the photorefractive polymer system is dominantly caused by the reorientation of chromophore under a given electric



**Figure 1.** Chemical structures of components of photorefractive composites.

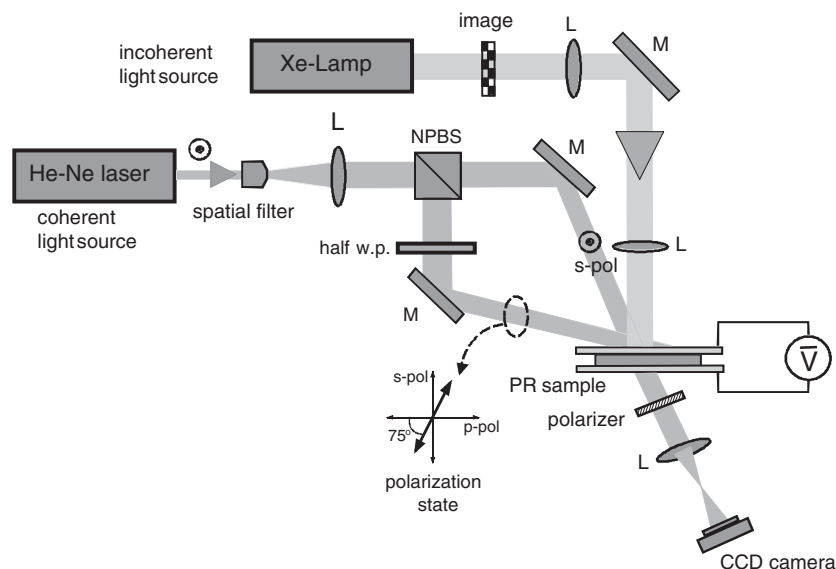
field,  $T_g$  of the system should be lowered below the room temperature to enhance its EO properties, index modulation, and response time.<sup>8</sup> The photorefractive composite was prepared by mixing PSX-CZ:P-IP-DC:BBP:TNF in the ratio of 63:27:9:1 wt %. The  $T_g$  of the composite was 22 °C by differential scanning calorimeter (Perkin Elmer DSC7) at the heating rate of 10 °C/min.

For device preparation, the mixture was dissolved in dichloromethane and filtered through a 0.2 μm filter. The composite was cast on an indium tin oxide (ITO) glass plate with an etched electrode pattern, dried slowly for 6 h at ambient temperature and subsequently heated in an oven at 90 °C for 24 h to remove the residual solvent. Then, the composite was softened by placing it on a hot plate at 100 °C, followed by sandwiching between ITO coated glasses under light pressure to yield a film with uniform thickness. The thickness of the film was controlled by a Teflon spacer between two ITO glass plates.

### Measurements

The diffraction efficiency of photorefractive material was determined by the degenerate four-wave mixing (DFWM) experiment. Two coherent laser beams at the wavelength of 632.8 nm were irradiated on the sample in the tilted geometry with the incident angle of 30° and 60° with respect to sample normal. Both of the writing beams were s-polarized and had the equal intensity of 60 mW/cm<sup>2</sup>. The photorefractive grating recorded was read out by a p-polarized counter-propagating reading beam. The intensity of reading beam was attenuated to 0.1 mW/cm<sup>2</sup> which is very weak compared with writing beam intensity in order not to affect the formation of photorefractive grating. The thickness of the sample prepared for DFWM measurement 100 μm.

The four-wave mixing setup was modified for the purpose of determining the space-charge field in pho-



**Figure 2.** Schematic diagram of optical setup for optically controlled spatial light modulator. half w.p.: half waveplate, PR sample: photorefractive sample, NPBS: non-polarizing beam splitter, M: mirror, L: lens, and CCD: charge-coupled-device.

photorefractive polymeric composites, which was described in detail in the previous paper.<sup>9</sup> Simply by adding a pair of crossed polarizers unit to a conventional degenerated four-wave mixing setup, we could measure the birefringence of the photorefractive materials induced by a newly formed space-charge field. Consequently, the magnitude of the space-charge field can be determined quantitatively from the variation of the birefringence induced by the space-charge field. Under a given external field applied, the birefringence of the photorefractive polymer can be calculated on the basis of the oriented gas model and the index ellipsoid method.<sup>10,11</sup> Oriented gas model is good for explaining the poling behavior of anisotropic molecules with permanent dipole under a given external field.

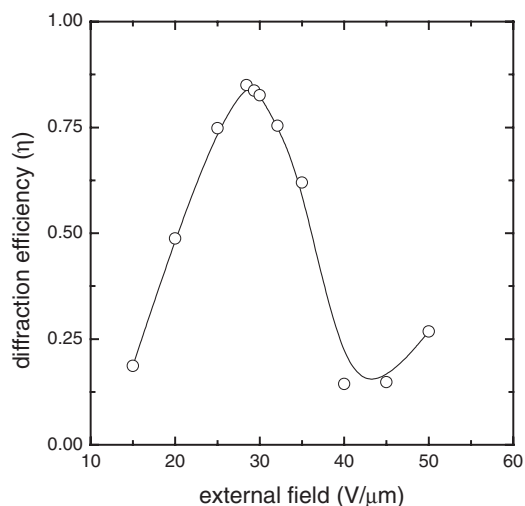
The index ellipsoid method allows estimation of the effective birefringence of the sample at various poling angles.

Optically controlled SLM (OCSLM) was demonstrated at the wavelength of 632.8 nm using our photorefractive sample and its schematic diagram was shown in Figure 2. We used Xe-lamp as incoherent light source and He-Ne laser at 633 nm as coherent one. The setup was based on the two-beam coupling. The s-polarized He-Ne laser beam was spatially filtered, expanded up to 30 mm in beam diameter, and split into two writing beams with a non-polarizing beam splitter. The polarization state of a writing beam could be controlled with a half-wave plate. We modified the polarization states of two writing beams in two-beam coupling setup in order to simplify the OCSLM setup. One of the writing beams is s-polarized and the other is linearly polarized at 75°. The latter play a role of the reading beam with its p-polariza-

tion component ( $I_0 \times \cos 75^\circ$ ) in addition to the writing beam with its s-polarization component ( $I_0 \times \sin 75^\circ$ ). That is, the interference pattern was made by the s-polarized writing beam and the s-polarization component of the other writing beam. Since the reading beam intensity was determined by the polarization direction, the angle should be adjusted experimentally on the basis of the photorefractive properties of the sample. In the case of our sample and geometric configuration, OCSLM showed best performance at the angle of 75°. The polarizer was placed between the sample and CCD camera to pass the only p-polarized diffracted beam, except the s-polarization components of the writing beams. Consequently the experimental setup could be simplified. Additional reading beam was not necessary for the readout of recorded information because of the linearly polarized writing beam at 75°. In order to provide the incoherent two-dimensional optical information to OCSLM, Xe-Lamp light was passed through the mask pattern. This incoherent image shined onto the photorefractive sample might destroy the grating generated by two coherent beams. Then the coherent reading beam shined onto the incoherent image would not be diffracted. So the coherent image converted from the incoherent image could be captured by the CCD camera.

## RESULTS AND DISCUSSION

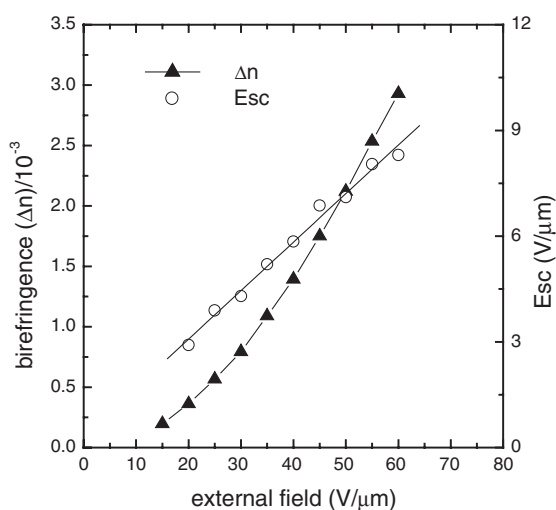
Diffraction efficiency is one of the most important factors determining the performance of holographic materials. Figure 3 shows the diffraction efficiency of the photorefractive composite measured by degenerate four-wave mixing method (DFWM) at various



**Figure 3.** Diffraction efficiency of the photorefractive composite at different external fields.

applied electric fields. As the applied field increases, the diffraction efficiency increases gradually up to 85% until *ca.* 30 V/μm and then decreases to near zero. This sinusoidal behavior is attributed to the property of diffraction represented by the equation:  $\eta = \sin^2(C \cdot \Delta n)$ , where  $\Delta n$  is index contrast of photorefractive grating and  $C$  is a constant depending on optical wavelength, sample thickness, etc. Our photorefractive material exhibits a good performance, *i.e.*, high diffraction efficiency of 85% at an applied field of 28 V/μm.

Figure 4 shows the field-induced birefringence of the photorefractive sample and the magnitude of internal space-charge field formed by the exposure of two writing beams. The electro-optic property was characterized by the transmission ellipsometric measurement at various applied fields. As can be seen in Figure 4,



**Figure 4.** (a) Photoinduced birefringence ( $\Delta n$ ) and (b) magnitude of the space-charge field ( $E_{sc}$ ) at different external fields.

the field-induced birefringence of the sample exhibited quadratic growth with increasing the applied field. In the case of the photorefractive polymeric material with low  $T_g$ , it has been generally accepted that the field-induced birefringence is mainly attributed to the molecular reorientation under a given applied field. The large orientational birefringence of current polymeric composite, which is associated with the large  $\mu^2 \Delta\alpha$  of P-IP-DC chromophore where  $\Delta\alpha$  is polarizability anisotropy and  $\mu$  is dipole moment, may lead to the high diffraction efficiency at a moderate electric field of 30 V/μm according to the results of the semi-empirical quantum calculation method (MOPAC 6).

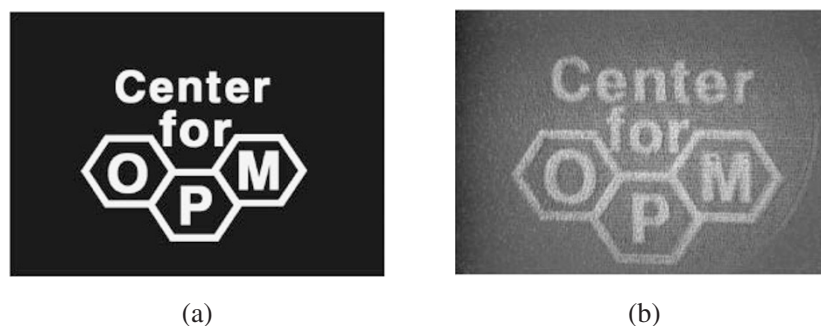
The magnitude of the space-charge field was measured at various applied fields using the modified four-wave mixing setup previously mentioned in the measurement section. The magnitude of space-charge field increases linearly with external electric field, which indicates that the grating vector component of the external field is much smaller than the saturation field,  $E_q$ .<sup>12</sup> According to the results, the magnitude of space-charge field obtained from our characterization method was *ca.* 70% of the grating vector component of the external field. Our polymer sample shows the good phase stability and optical clarity over several months when stored at room temperature, which was confirmed by an UV spectrometer and a polarizing microscope.

In Figure 5, the optically controlled SLM was demonstrated using our photorefractive sample. The OCSLM setup was constructed based on the two-beam coupling setup. Its basic principle can be described as follows: two laser beams intersected the photorefractive sample and formed the photorefractive grating in the sample. Then, incoherent light imposed with an input image illuminated on the sample and erased locally the photorefractive grating in accordance with the input image. When the reading beam was illuminated, the transmitted beam had the same image with what was imposed in the incoherent beam. Figure 5a and b exhibited the incoherent image and the coherent image converted, respectively.

The incoherent image was successfully converted into coherent image using this OCSLM at the applied field of 30 V/μm. The high-contrast coherent image could be obtained at a relatively low applied field because of the excellent steady-state photorefractive performance of our sample. The thickness of the active layer in the sample was 50 μm.

## CONCLUSIONS

We prepared photorefractive polymeric composite consisting of carbazole-substituted polysiloxane ma-



**Figure 5.** Photographs of (a) incoherent image to be input on optically controlled SLM and (b) converted coherent image imposed on He–Ne laser light.

trix, P-IP-DC chromophore, BBP plasticizer, and TNF photosensitizer. Our sample showed high diffraction efficiency of 85% even at the applied field of 28 V/ $\mu\text{m}$ , which provided substantial advantage for the demonstration of holographic application. Also, we experimentally determined two factors in the composite, effective electro-optic coefficient and internal space-charge field under different external fields, which govern the efficiency of photorefractivity. At an applied field of 30 V/ $\mu\text{m}$ , the birefringence and the space-charge field were *ca.*  $8 \times 10^{-4}$  and 4.5 V/ $\mu\text{m}$ , respectively. Optically controlled spatial light modulator was successfully demonstrated using our photorefractive composite. The incoherent image imposed in Xe-Lamp light was converted into coherent image with good contrast in the coherent light from He–Ne laser. We expect it to be used for optical data processing such as holographic recording, pattern recognition, and novelty filter.

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#### REFERENCES

1. P. Günter and J.-P. Huignard, "Photorefractive Materials and Their Applications," Springer-Verlag, Berlin, 1988, vol. 1, p 7.
2. W. E. Moerner and S. M. Silence, *Chem. Rev.*, **94**, 127 (1994).
3. W.-J. Joo, N.-J. Kim, H. Chun, I. K. Moon, and N. Kim, *Polymer*, **43**, 9863 (2001).
4. B. L. Volodin, B. Kippelen, K. Meerholz, B. Javidi, and N. Peyghambarian, *Nature*, **383**, 58 (1996).
5. C. Poga, P. M. Lundquist, V. Lee, R. M. Shelby, R. J. Twieg, and D. M. Burland, *Appl. Phys. Lett.*, **69**, 1047 (1996).
6. J. W. Goodman, "Introduction to Fourier Optics," McGraw-Hill, Singapore, 1966, pp 243–246.
7. H. Chun, I. K. Moon, D.-H. Shin, and N. Kim, *Chem. Mater.*, **13**, 2813 (2001).
8. W. E. Moerner, S. M. Silence, F. Hache, and G. C. Bjorklund, *J. Opt. Soc. Am. B*, **11**, 320 (1994).
9. W.-J. Joo, N.-J. Kim, H. Chun, I. K. Moon, N. Kim, and C.-H. Oh, *J. Appl. Phys.*, **91**, 6471 (2002).
10. A. Yariv, "Optical Electronics," 4th ed., Saunders College Publishing, Philadelphia, PA, 1991, chapt. 1.
11. H. S. Nalwa and S. Miyata, Ed., "Nonlinear Optics of Organic Molecules and Polymer," CRC Press, Boca Raton, FL, 1997, p 465.
12. P. Yeh, "Introduction to Photorefractive Nonlinear Optics," Wiley, New York, N.Y., 1993, chapt. 2 and 3.

1. P. Günter and J.-P. Huignard, "Photorefractive Materials