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Phase grating generation and optical phase conjugation in photorefractive polymer/dissolved liquid crystal composites

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The orientational photorefractive properties of photorefractive mesogenic composites have been investigated by means of two-beam coupling and degenerate four-wave mixing. Photorefractive mesogenic composites, consisting of low molar mass liquid crystals, polymer, and photoconductive sensitizer, constitute novel organic materials possessing high performance photoreactivity. The refractive index change was estimated on the basis of the theory of two-beam coupling and four-wave mixing, and large index modulation of over 0.01 was obtained.

1. Introduction

Optical phase conjugation plays a very important role in photonics [1, 2]. The phenomenon of optical phase conjugation has been investigated in various material systems possessing third-order optical non-linearity. Among them, photorefractive materials are by far the most efficient of non-linear optical media for optical wave mixing and phase conjugation with low light intensity requirements. Among non-linear optical media, organic materials possess some important advantages over inorganics [3, 4]. Several types of photosensitive organic material have been developed into new active devices, including photochromic and photorefractive materials. Since the first report in 1990 [5], many investigations on organic photorefractive materials such as organic crystals, polymer composites, and liquid crystals have contributed to the improvement of photorefractive performance [6, 7]. Among these organic photorefractive materials, photorefractive liquid crystals have several advantages in comparison with other materials [6–11]: (i) a low operating voltage required for the realization of wave mixing, (ii) a large refractive index modulation resulting from the large anisotropy of the mesogenic molecules, and (iii) ease of fabrication, as developed in the display industry. Low molar mass nematic LCs (L-LCs) show high performance photorefractivities deriving from the L-LC property of photoconductivity. Since the discovery of photorefractive LCs, good photorefractive performance has been observed only for large fringe spacing (Raman–Nath regime).

Recently, it has been found that the resolution of a photorefractive LC is improved by combining a L-LC with a polymer [6, 7, 12–15]. Photorefractive Bragg gratings can be recorded in such materials. Polymeric materials in the photorefractive mesogenic composites play a very important role in terms of improving the resolution, stabilizing the homeotropic alignment, and functionalizing the materials. The aim of the present study is to investigate the fabrication of a photorefractive phase grating from a functionalized mesogenic composite, and to demonstrate the optical phase conjugation by means of two-beam coupling and degenerate four-wave mixing.

2. Experimental

2.1. *Materials*

Our photorefractive mesogenic composite was based on a low molar mass nematic mixture, functionalized copolymer, and sensitizer. The low molar mass liquid crystal consisted of the multicomponent nematic mixture given in figure 1 (*a*); the functionalized copolymer, described in figure 1 (*b*), was synthesized by free radical copolymerization of methacrylate monomers containing 4-cyanobenzoate and *N*-ethylcarbazoyl side

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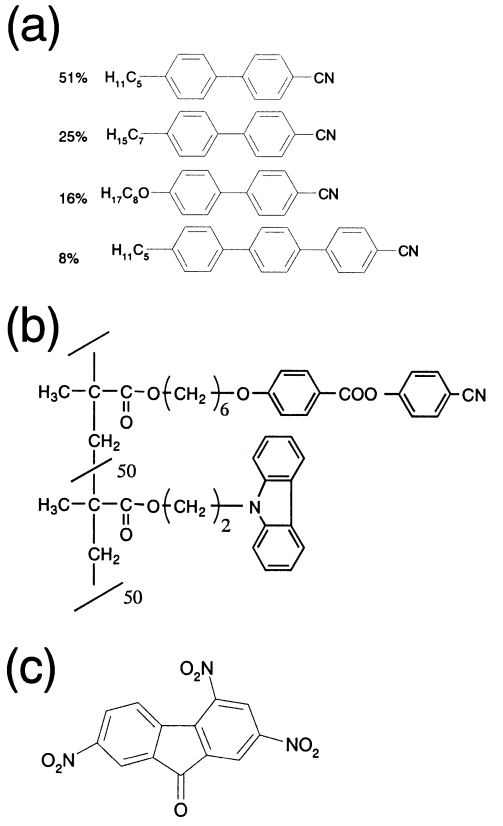

Figure 1. Chemical formulae of the components of the photorefractive mesogenic composite used in the present study. (*a*) Low molar mass liquid crystal mixture, (*b*) functionalized copolymer, and (*c*) photoconductive sensitizer (TNF).

groups; the copolymer makes the charge transfer complexes with the sensitizer 2,4,7-trinitro-9-fruorene (TNF) molecules, figure 1 (*c*), which is favourable for photoconductive effects.

The photorefractive mesogenic composite was prepared by mixing the nematic mixture, copolymer and TNF (20:78:2 wt %) at about 150°C and stirring until the mixture became transparent. The colour of the solution of the nematic mixture with TNF was yellow. Once the copolymer was added to this solution, the colour was changed to red by the generation of charge transfer complexes. The homogeneous solution was sandwiched between two ITO-coated glasses substrates with a 50-μm thick polyester film as the spacer. When the mixture was cooled below 58°C, it assumed the mesogenic phase without phase separation. Since the resulting mesogenic composite revealed a nematic liquid crystalline phase, the opaque film became completely transparent on application of an electric field, due to the homeotropic alignment of the mesogen. Once the mesogenic groups in the photorefractive mesogenic material aligned homeotropically, this homeotropic

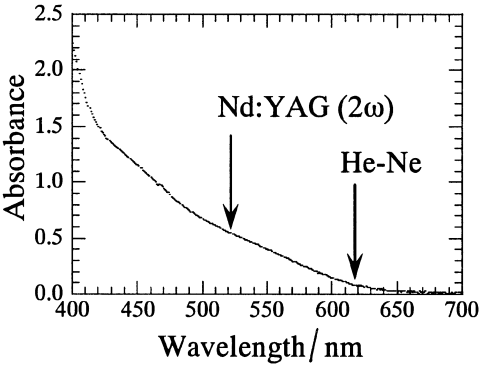

Figure 2. Absorption spectrum of the photorefractive mesogenic composite. A frequency-doubled Nd-YAG laser (532 nm) or He-Ne laser (633 nm) was used as the pumping beam.

state was maintained under zero electric field, even at room temperature, due to the polymer-stabilizing effects. Figure 2 shows the absorption spectrum of the photorefractive mesogenic composite. Because of the charge transfer complexes, the light in the visible region was absorbed, which is desirable for photorefractive applications.

2.2. *Measurements*

To investigate the photorefractive properties in our sample we performed two-beam coupling and four-wave mixing experiments in the tilted geometry, as shown in figure 3. In the two-beam coupling configuration described in figure 3 (*a*), a linearly polarized beam from a continuous-wave He-Ne laser (633 nm) was divided into two beams of equal intensity by a polarizing beam splitter. A half-wave plate controlled the polarization directions of the beam. Two coherent *p*-polarized writing beams were superposed onto the sample on the same spot.

Phase conjugation was measured with a degenerate four-wave mixing configuration shown schematically in figure 3 (*b*). Optical phase conjugation experiments were carried out using a 633 nm He-Ne laser. The electronic transition of the photorefractive mesogenic composite is off-resonant, with a wavelength of 633 nm as shown in figure 2. The output laser beam was divided into two beams, which were directed onto the mesogenic composite possessing orientational photorefractive properties. One of the two beams formed the counter propagating pump fields E_1, E_2, and the other was used as the probe beam with electric field amplitude E_4. Interaction of these beams in the photorefractive polymer composite generated the fourth beam of field amplitude E_3, which is phase conjugate to the probe beam E_4. The polarization directions were

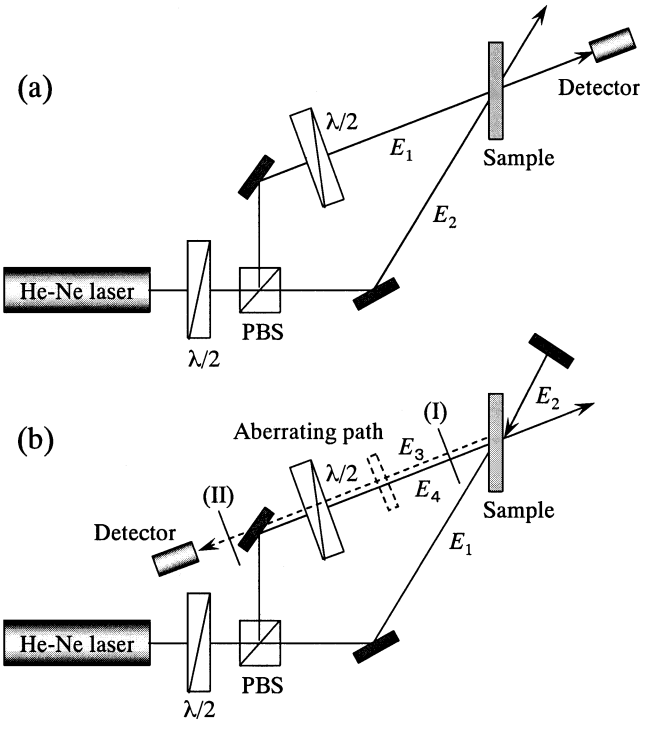

Figure 3. Geometry of (*a*) two-beam coupling and (*b*) degenerate four-wave mixing experiments. $\lambda/2$ = half-wave plate; PBS = polarizing beam splitter.

independently controlled by half-wave plates. Beams 1 and 4 were of the same polarization state (s-polarization) and the polarization direction of beam 3 was perpendicular to that of the other two (p-polarization), in order to prevent two-beam coupling. In fact, it was confirmed that the gain coefficients of two-beam coupling between any of the beams were negligibly small under the experimental conditions described here. The grating period was varied by controlling the incident angle between beams E_1 and E_4 and set to be about 3.0 μm.

3. Results and discussion

The electric fields of the two light waves may be written

$$E_j = A_j \exp\left[\mathrm{i}\left(\omega t - k_j \cdot \mathbf{r}\right)\right], j = 1, 2 \tag{1}$$

where A_1, A_2 are the wave amplitudes, ω is the angular frequency, and $\mathbf{k}_1$, $\mathbf{k}_2$ are the wave vectors. The index of refraction including the fundamental component of the intensity-induced gratings can be written

$$n = n_0 \left[\frac{n_1}{2}\exp(\mathrm{i}\phi)\exp[-\mathrm{i}K\cdot r]\frac{A_1^* A_2}{I_0} + \text{c.c.}\right] \tag{2}$$

where c.c. represents complex conjugation, ϕ is a phase shift and

$$\mathbf{K} = k_2 - k_1 \tag{3}$$

$$I_0 = |A_1|^2 + |A_2|^2. \tag{4}$$

Here n_0 is the index of refraction without irradiation and n_1 is a real number. By using the coupling equations including photorefractive index of refraction described in equation (2), the solutions for I_1, I_2 corresponding to the transmitted light intensities are obtained as

$$I_1 = I_1(0)\frac{1+\xi^{-1}}{1+\xi^{-1}\exp(\Gamma d)} \tag{5}$$

$$I_2 = I_2(0)\frac{1+\xi}{1+\xi\exp(-\Gamma d)} \tag{6}$$

where ξ is the input intensity ratio,

$$\xi = \frac{I_1(0)}{I_2(0)} \tag{7}$$

Γ is the gain coefficient,

$$\Gamma = \frac{2\pi n_1}{\lambda \cos\frac{\theta}{2}}\sin\phi \tag{8}$$

and d is the thickness of the gratings and is rewritten as

$$d = L/\cos\beta \tag{9}$$

where θ is the incident angle of the two beams and L is the thickness of the photorefractive mesogenic composite. When a phase shift taken place between the recorded grating and the light fringe pattern, the energy is transferred between them according to equations (5) and (6). If there is no phase shift ($\phi = 0$), Γ is equal to zero according to equation (8) and the energy transfer cannot be observed as understood by equations (5) and (6). In other words, observation of energy transfer in the thick grating condition is considered as firm proof of the photorefractive nature of the recorded grating.

Optical phase conjugation in the photorefractive mesogenic composite was measured with the degenerate four-wave mixing configuration shown schematically in figure 3 (*b*); a He-Ne laser of 633 nm was used. As shown in figure 2, the mesogenic film was transparent and the absorption coefficient at 633 nm was less than $1\,\mathrm{cm}^{-1}$. This means that the absorption loss of our photorefractive mesogenic composite is sufficiently low at 633 nm. The output laser beam was divided into two beams forming the counter propagating pump fields E_1, E_2; another beam was used as the probe beam with electric field amplitude E_4. Interaction of these beams in the photorefractive mesogenic composite leads to generation of the fourth beam of field amplitude E_3, which is phase conjugate to the probe beam E_4. The beams 1, 2, 3 and 4 have the same angular frequency ω.

The field amplitude $E_j(r, t)$ associated with the j-th beam are assumed to be

$$E_j(r, t) = E_j(r) \exp\left[\mathrm{i}\left(k_j r - \omega t\right)\right] + \text{c.c.} \tag{10}$$

where k_j is the j-th wave vector. Since spatial coherency is maintained under our experimental conditions, the interference between the beams gives rise to six sets of spatial intensity modulations in the photorefractive mesogenic composite, each with a different fringe spacing. Two sets, between E_1 and E_4, and between E_2 and E_3, form transmission gratings, while the other four form reflection gratings.

It is expected that optical phase conjugation in the photorefractive mesogenic composites strongly depends on the efficiency of the beam coupling through the non-local grating. Since the beam coupling coefficients depend on the polarization directions of the pump and probe beams, it is important to observe the dependence of the polarization directions, in order to clarify the optical phase conjugation from non-local gratings in the photorefractive mesogenic material. In the present study, the polarization directions of the three beams were controlled by three half-wave plates and beams 1 and 4 were of the same polarization state (s-polarization) in order to prevent two-beam coupling, the polarization direction of beam 2 was perpendicular to that of the other two beams (p-polarization). Under degenerate four-wave mixing configurations, in general, interference between the beams gives rise to six sets of spatial intensity modulations in the composite, each with a different fringe spacing. Among the six sets of gratings, the grating period of the reflection grating is shorter than the resolution limit of our photorefractive material; the coupled equations can be simplified by assuming that the phase-conjugated response is mostly dependent on the transmission grating. If the transmission grating is dominant, the four beams can write the following grating, whose grating vector is given by

$$\mathbf{K} = k_1 - k_3 = k_4 - k_2. \tag{11}$$

The resulting space charge field-induced refractive index can be written as consisting of spatial Fourier components:

$$n = n_0 + \frac{n_1 \exp(\mathrm{i}\phi)}{2} \frac{E_1 E_3^* + E_2^* E_4}{I_0} \exp(\mathrm{i}\mathbf{K}\cdot\mathbf{r}) + \text{c.c} \ldots \tag{12}$$

The following Helmholtz equation must be solved by substituting the refraction index into the Helmholtz wave equation:

$$\left(\nabla^2 + \omega^2 \mu\varepsilon\right) E = 0. \tag{13}$$

In the first approximation, the beam amplitudes may be assumed to obey the slowly varying envelope approximation, i.e. they are assumed to vary by only a small fraction over the distance of one optical wavelength. Mathematically, the second-order spatial derivative is neglected relative to the optical wave vector times the first derivatives as follows:

$$\left|\frac{\mathrm{d}^2}{\mathrm{d}z^2} E_j\right| << \left|\beta_j \frac{\mathrm{d}}{\mathrm{d}z} E_j\right| \tag{14}$$

where

$$\beta_j = k_j \cos\theta. \tag{15}$$

Solving the Helmholtz wave equation under the slowly varying approximation, we arrive at the following coupled equations:

$$\frac{\mathrm{d}E_1}{\mathrm{d}r} = -\frac{\alpha}{2} E_1 \tag{16}$$

$$\frac{\mathrm{d}E_2}{\mathrm{d}r} = \frac{\alpha}{2} E_2 \tag{17}$$

$$\frac{\mathrm{d}E_3}{\mathrm{d}r} = \frac{\alpha}{2} E_3 + \mathrm{i}\pi n_1 \frac{\exp(-\mathrm{i}\phi)}{\lambda} \frac{E_1 E_4^* + E_2^* E_3}{I_0} E_2 \tag{18}$$

$$\frac{\mathrm{d}E_4^*}{\mathrm{d}r} = \frac{\alpha}{2} E_4^* + \mathrm{i}\pi n_1 \frac{\exp(-\mathrm{i}\phi)}{\lambda} \frac{E_1 E_4^* + E_2^* E_3}{I_0} E_1^* \tag{19}$$

where α is the absorption coefficient and $r = z/\cos\theta$. Using the boundary condition $E_3(d) = 0$, the equation for E_3 can be written

$$E_3 = \mathrm{i}c\gamma E_1(0) \int_{\mathrm{r}}^{\mathrm{d}} \frac{\exp\left[(\gamma + \alpha/2)(r - r')\right]}{I_0(r')} \mathrm{d}r' \equiv c\gamma E_1(0)/J(r) \tag{20}$$

where

$$\gamma = \frac{\mathrm{i}\pi n_1 \exp(-\mathrm{i}\phi)}{\lambda} \tag{21}$$

and

$$c = \frac{E_2(d) E_4^*(0) \exp(-\alpha d/2)}{\gamma I_1(0)/J(0) - 1}. \tag{22}$$

Taking the square of both sides of equation (20) one obtains the intensity of the phase conjugation beam. The phase conjugate reflectance is defined as the intensity ratio of probe and phase conjugate beams. To confirm that the generated light is a phase conjugate beam under our experimental conditions, we observed that phase aberrations were compensated by letting the wave-front retrace its path through the aberrating medium, following wave-front reversal via a phase-conjugated mirror, which is undoubted proof of the phase conjugation. In figure 4 we show photographs taken in the far field of (*a*) the now distorted beam [corresponding to plane (I) illustrated in figure 3 (*b*)] after passage through the inhomogeneous polymer-coated glass plate; and (*b*) the exciting beam [corresponding to plane (II) in figure 3 (*b*)] after conjugation and subsequent traversal back through the distorter. The near-perfect recovery of a diffraction-limited focal spot using non-linear optical phase conjugation is thus demonstrated.

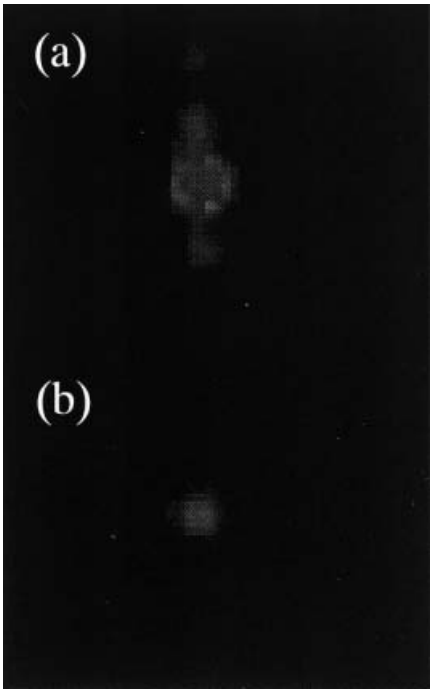

Figure 4. Results of an aberration correction demonstration with the configuration of figure 3 (*b*). Photograph (*a*) shows the beam after it has passed through a phase aberrator; (*b*) shows the beam after a phase conjugate reflection and a second pass through the same aberrator.

Figure 5 is a comparison between the refractive index changes estimated from the two-beam coupling experiments and those from the four-wave mixing. Similar results were obtained from the two-beam coupling and four-wave mixing experiments although the small discrepancy in the region of large light intensity cannot be ignored. We think that this discrepancy resulted from the existence of the backward reading beam in the degenerate four-wave mixing. In order to confirm this consideration, in the two-beam coupling experiments, the photorefractive mesogenic composite was irradiated with an additional incoherent laser beam and the effects were investigated. The two beam coupling properties were disturbed by irradiating with the additional laser beam as shown in figure 6. This means that the additional photogenerated charges with uniform distribution disturbed the spatially modulated charge generated by the interference light. This annihilation of the two-beam coupling was caused more effectively by using the frequency-doubled Nd-YAG laser beam as the additional incoherent laser beam. The frequency-doubled Nd-YAG laser beam irradiation effectively generated space charges because its wavelength of 532 nm is on-resonant as shown in figure 2. Figure 7 shows the gain coefficients versus

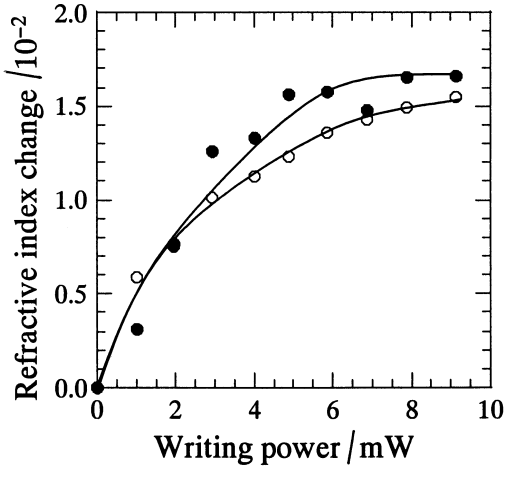

Figure 5. Refractive index change versus writing power. Filled circles denote the refractive index changes estimated from two-beam coupling experiments, open circles are those from degenerate four-wave mixing.

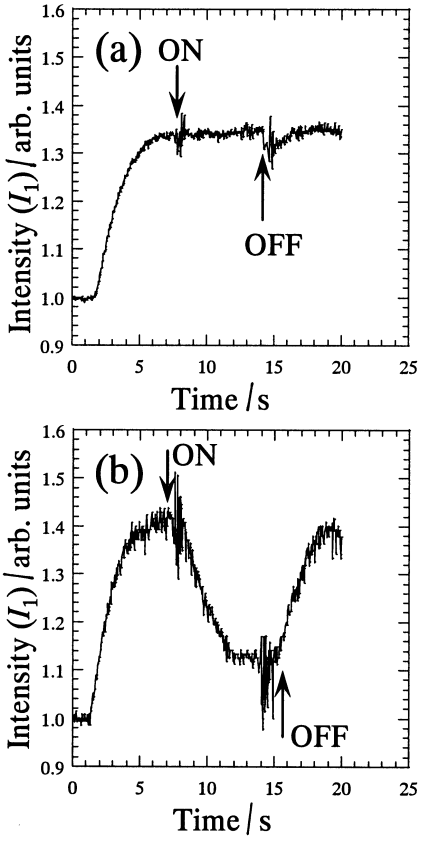

Figure 6. The time dependence of light intensity in the two-beam coupling experiments. The intensity of one beam was monitored while an additional incoherent light was opened (ON) and closed (OFF). The additional incoherent beam was emitted from (*a*) a He-Ne laser and (*b*) a frequency-doubled Nd-YAG laser.

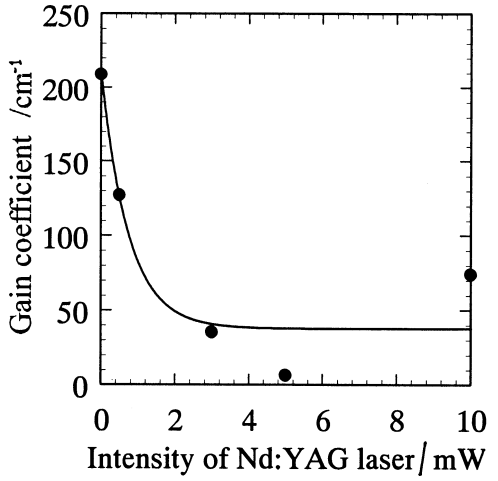

Figure 7. Gain coefficient in the two-beam coupling experiments versus intensity of the Nd-YAG laser.

intensity of the frequency-doubled Nd-YAG laser. The photorefractive gratings can be erased by irradiating with the frequency-doubled Nd-YAG laser, which means that the modulation depth of the photorefractive gratings in mesogenic composites can be controlled by the intensity of the additional incoherent light. Further investigation is necessary to clarify the detailed mechanism, although we offer the above-mentioned qualitative mechanism.

4. Conclusions

The orientational photorefractive properties of mesogenic composite have been investigated by two-beam coupling and degenerate four-wave mixing. The photorefractive mesogenic composites, consisting of a low molar mass liquid crystal, polymer, and photoconductive sensitizer, constitute novel organic materials possessing high performance photorefactivity. The refractive index change was estimated on the basis of the theory of two-beam coupling and four-wave mixing; similar results were obtained from both experiments although there is small discrepancy. This small discrepancy has been attributed to the effect of the backward reading beam in the degenerate four-wave mixing.

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