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Optical phase conjugation from phase-shifted gratings in photorefractive mesogenic composites

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Optical phase conjugation has been observed from non-local gratings generated in a photorefractive mesogenic composite consisting of a low-molar-mass nematic mixture, a functionalized copolymer and a photoconductive sensitizer. The degenerate four-wave mixing experiment enabled measurements of phase conjugate reflectance as a function of the intensity ratio of the forward and backward pump beams. The characteristics were strongly dependent on the spatial phase shift; these results are quantitatively explained by the coupled wave theory in relation to non-local gratings.

1. Introduction

Optical phase conjugation is a fascinating subject that will play a very important role in the forthcoming highly information-oriented society [1]. The optical phase conjugation phenomenon has been investigated in various material systems possessing third order optical non-linearity [1–3]. Among them, photorefractive materials are by far the most efficient non-linear optical media for optical wave mixing and phase conjugation with low light intensity requirements [2, 3]. In photorefractive media an optical periodic pattern inside the material, created by interference between two coherent beams, results in a periodic patterned generation of mobile charges and corresponding charge generation sensitizer ions. Drift of mobile charges under an external d.c. field, and subsequent recombination with sensitizer ions, results in charge separation and a space-charge field. The resulting internal space-charge electric field then modulates the refractive index to create a phase grating that can diffract a light beam. The charge transport in the photorefractive materials gives rise to a spatial phase shift between the incident light intensity pattern and the refractive index modulation. An important consequence of this phase shift is energy transfer, called asymmetric two-beam coupling, between two light beams interfering

in a photorefractive medium. If the coupling is sufficiently strong, the two-beam coupling gain may exceed the losses and optical amplification can occur.

More than 20 years after the photorefractive effect was discovered in inorganic crystals such as LiNbO_3 and BaTiO_3 , research has been focused extensively on these inorganic crystals [2, 3]. Since the discovery of the photorefractive effect in an organic crystal in 1990 [4, 5], searches for new photorefractive materials have been extended to organic materials including organic crystals [4–6], polymers [6–8], organic glass [6] and liquid crystals [9–28]. Among these organic photorefractive materials, liquid crystals show a high diffraction efficiency and fast response under a low operating voltage. The refractive index change due to the electro-optic response in liquid crystals originates in reorientation of the mesogenic molecules. A liquid crystal is peculiar in that it is made of anisotropic molecules and it also keeps anisotropy on a macroscopic scale. Due to this anisotropy, the reorientation of molecules gives rise to a large change in refractive index. In 1994, Khoo and Liang reported that low-molar-mass nematic liquid crystals show high performance photorefractivities with photoconductivity [9]. The mixture of the nematic low-molar-mass liquid crystal doped with a sensitizing dye is homeotropically aligned on indium tin oxide (ITO)-coated glass slides to allow for the application of an external electric field. Reorientation of liquid crystalline

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molecules can occur even in weak optical and electric fields, so a space-charge field, which can be orders of magnitude larger than an optical field, should produce impressive photorefractivity.

The photorefractivity in mesogenic materials is mainly based on orientational birefringence and the effects are named orientational photorefractive effects. One advantage in this system is that the operating voltage required is much lower than for the other organic photorefractive materials including polymer composites, organic crystals and organic glasses. The dye-doped low-molar-mass liquid crystals show high performance photorefractivities; but unfortunately, good photorefractive performance is only observed for large fringe spacing, which is not suitable for high resolution applications. A sophisticated method for improving the characteristics of the photorefractive mesogenic materials is to combine the low-molar-mass liquid crystals with a polymer [12, 13, 16, 17, 20, 22]. 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 [16, 17, 22, 27, 28]. Recently, we observed highly efficient phase conjugation in the photorefractive mesogenic composite in the Bragg regime and the effects of light absorption on the properties were clarified [28]. Since non-local gratings are generated in the photorefractive mesogenic composites, it is expected that the spatial phase shift strongly affects the properties of the optical phase conjugation. The aim of the present article is to demonstrate characteristics of optical phase conjugation in a photorefractive mesogenic composite and to investigate the effects of the non-local gratings with spatial phase shifts.

2. Experimental

2.1. Materials

Our photorefractive mesogenic composite consists of a low-molar-mass nematic mixture (E7), copolymer (figure 1) and a photoconductive sensitizer. One of the merits of the photorefractive mesogenic composite is the use of the highly functionalized copolymer. The copolymer consists of mesogenic and photoconductive functions in the polymer side chains. Since mesogenic molecules are incorporated into the polymer side chain, the low-molar-mass nematic mixture can easily dissolve such a copolymer without phase separation. Since phase separation in the mesogenic composite does not occur at room temperature, the composite in the mesophase shows good transparency without scattering. The copolymer described in figure 1 forms charge transfer complexes on adding sensitizing dye molecules such as 2,4,7-trinitro-9-fluorenone (TNF), which promotes photoconductive effects in the visible regime.

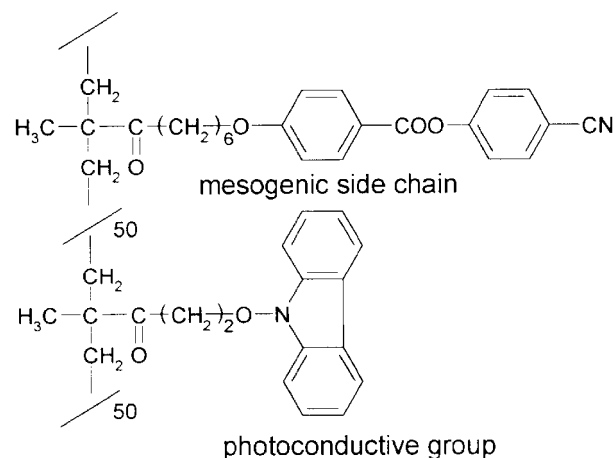


Figure 1. Chemical structure of the functionalized copolymer for the photorefractive mesogenic composite. The copolymer has both mesogenic and photoconductive group side chains.

The photorefractive mesogenic composite sample was prepared by mixing the copolymer, E7 and TNF (20.00:79.65:0.35 wt %) at about 140°C and stirring until the mixture became transparent. The homogeneous solution was sandwiched between two ITO-coated glass substrates with a 50 μm thick polyester film as the spacer. Since the mesogenic composite shows the mesophase at room temperature, an opaque film was obtained when the mixture was cooled below the clearing temperature. When a d.c. voltage was applied to the sample, the opaque mesogenic composite film became completely transparent and this state was maintained under no electric field due to the polymer-stabilized effect. Figure 2

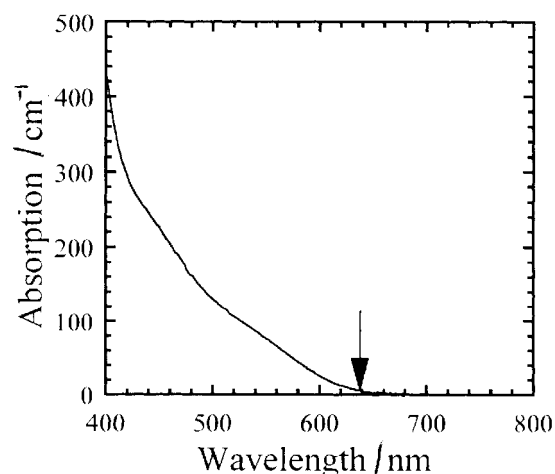


Figure 2. Absorption spectrum of the photorefractive mesogenic composite. A He-Ne laser was used as the pump light source and the wavelength (633 nm) is shown as an arrow in the figure.

shows absorption spectrum of the photorefractive mesogenic composite film. The copolymer and TNF form charge transfer complexes and the absorption coefficients in the visible region increase on adding the TNF.

2.2. Measurements

2.2.1. Photorefractive phase shift

We can use the conventional theory for photorefractive two- and four-wave mixing because all experiments described here were performed for the thick grating (Bragg) regime. Clear evidence of photorefractivity is provided by demonstration of the phase-shifted refractive index grating [2, 3]. The refractive index modulation is out-of-phase with the optical interference pattern, and this phase shift can induce an energy exchange between two coherent beams, which strongly affect the characteristics of the optical phase conjugation. In order to clarify the nature of optical phase conjugation in the mesogenic composite, the value of the spatial phase shift must be determined. The photorefractive phase shift was measured by the translational method with a two-beam coupling configuration as shown in figure 3. A continuous wave He-Ne laser beam with p-polarization was split into two beams which were made to overlap in the photorefractive mesogenic composite mounted on a translational stage. The index of refraction including the fundamental component of the intensity-induced gratings can be written as

$$n = n_0 \left[\frac{n_1}{2} \exp(i\phi) \exp[-i\mathbf{K} \cdot \mathbf{r}] \frac{A_1^* A_2}{I_0} + \text{c.c.} \right] \quad (1)$$

where \mathbf{K} is the grating vector, I_0 is the total intensity of the two beams, n_1 is the index modulation depth,

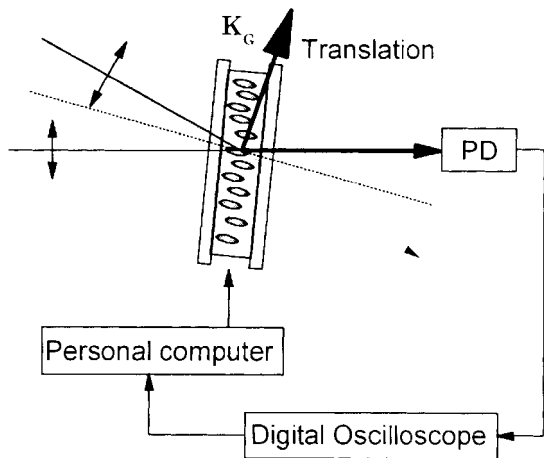


Figure 3. Schematic of the two-beam coupling experimental geometry for measuring the spatial phase shift. The two writing beams are p-polarized; PD is a photodiode.

and ϕ is the spatial phase shift. By solving the coupling equations, including the photorefractive index of refraction, the gain g can be expressed as [2, 3]

$$g = \frac{1 + m}{1 + m \exp(-\Gamma d)} \quad (2)$$

where m is the ratio of the input beam intensities, d is the interaction length, and Γ is the exponential gain coefficient, which is expressed for a codirectional two-wave mixing by

$$\Gamma = \frac{2\pi n_1}{\lambda \cos \frac{\beta}{2}} \sin \phi \quad (3)$$

where β is the incident angle between the two writing beams. The two-beam coupling gain coefficient strongly depends on the polarization direction of the two writing beams. We confirmed that the two-beam coupling effects can be ignored when the two writing beams were of the s-polarization state. Therefore, polarization directions of the two beams were set to be p-directions as shown in figure 3. A simple experimental technique was performed for measuring ϕ that is based on translating the grating with respect to the interference pattern at a rate much faster than the response of the photorefractive mesogenic composite. This can be accomplished by the direct movement of the sample. The fringe spacing was set to be $3.0 \mu\text{m}$ by controlling the incident angle θ between the writing beams. A d.c. field was applied and the sample was tilted 30° from the bisector of the two writing beams to permit charge migration along the grating wave vector.

2.2.2. Degenerate four-wave mixing

Optical phase conjugation in the photorefractive mesogenic composite has been measured with the degenerate four-wave mixing configuration shown schematically in figure 4. Optical phase conjugation experiments were carried out using a He-Ne laser at 633 nm. As shown in figure 2, the mesogenic film was transparent and the absorption coefficient at 633 nm was less than 1 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 three beams: two formed the counter-propagating pump fields E_1 and E_2 ; the third 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 a fourth beam of field amplitude E_3 , which is phase conjugate to the probe beam E_4 . The beams 1, 2, 3 and 4 are of the same angular frequency ω . The field amplitude $E_j(r, t)$ associated with the j -th beam are

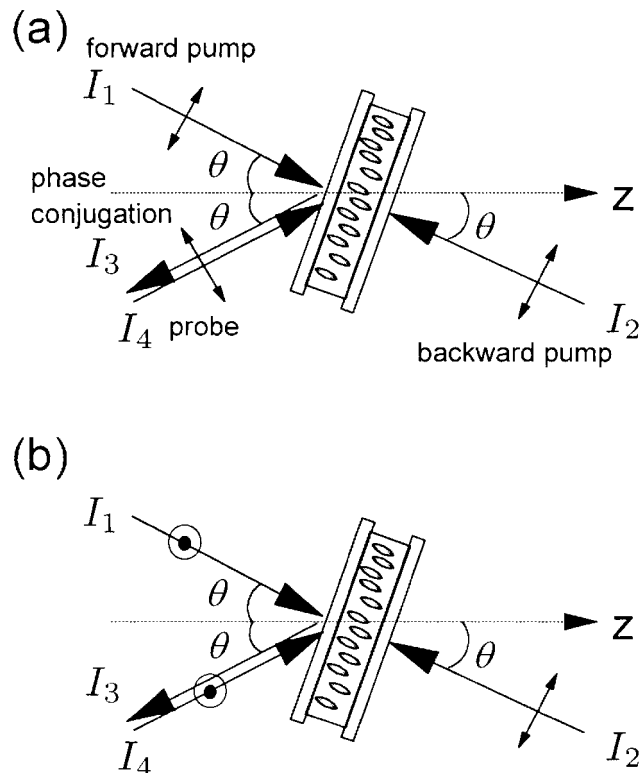


Figure 4. Geometry of the degenerate four-wave mixing experiments. I_1 and I_2 denote the forward and backward pump beams, respectively. I_4 represents the probe beam and I_3 the phase conjugate signal beam. (a) Beams 1, 2 and 4 were of the same polarization state (p-polarization); (b) beams 1 and 4 were of the same polarization state (s-polarization) and the polarization direction of beam 2 was perpendicular to that of the other two beams.

assumed to be

$$E_j(r, t) = E_j(r) \exp[i(k_j r - \omega t)] + \text{c.c.} \quad (4)$$

where k_j is the j -th wave vector. Since the spatial coherence is kept 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 the properties of optical phase conjugation in the photorefractive mesogenic composites strongly depend on the efficiency of 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 very important to observe the dependence of the polarization directions in order to clarify the optical phase conjugation from the non-local gratings. In the present study, the polarization directions of the three beams

were independently controlled by three half-wave plates. In the configuration shown in figure 4(a), beams 1, 2 and 4 were of the same polarization state (p-polarization) in order to permit the two-beam coupling between beams 1 and 4. On the other hand, in the configuration shown in figure 4(b), beams 1 and 4 were of the same polarization state (s-polarization) in order to prevent two-beam coupling, and the polarization direction of beam 2 was perpendicular to that of the other two beams (p-polarization).

3. Results and discussion

The photorefractive phase shift was measured by the translational method with two-beam coupling configuration as described in figure 3. Since the two-beam coupling gain coefficient can be expressed by equation (3), when the grating is translated from the initial writing, Γ becomes

$$\Gamma(\xi) = \frac{2\pi n_1}{\lambda \cos \frac{\beta}{2}} \sin(\phi + 2\pi\xi/\Lambda) \quad (5)$$

where ξ is the grating translation in the direction of the grating vector and Λ is the grating constant. Since the translational signal is obtained as a function of moving time of the stage, ξ should be expressed as a function f of time.

$$\Gamma(t) = \frac{2\pi n_1}{\lambda \cos \frac{\beta}{2}} \sin[\phi + 2\pi f(t)/\Lambda] \quad (6)$$

where the function f includes the velocity and acceleration of the translational stage. Figure 5 shows a typical example

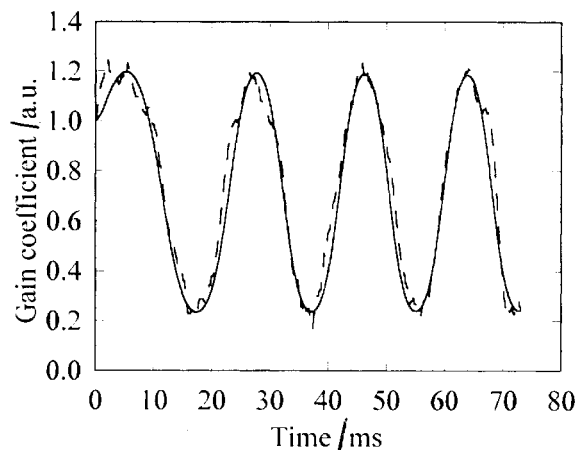


Figure 5. Beam intensity modulation for the photorefractive mesogenic composite with applied electric field of $0.45 \text{ V } \mu\text{m}^{-1}$, by use of the two-beam coupling translation technique. The dashed line denotes experimental data, and the solid line denotes theoretical fitting results.

of signals for measuring the photorefractive phase shift. The experimental curve (dashed line) was in good agreement with the theoretical curve (solid line) obtained from equation (6). The absolute value of the spatial phase shift is strongly dependent on the magnitude of the applied electric field. When the applied electric field is set to be 0.2, 0.3 and 0.4 V μm^{-1} , the spatial phase shift can be estimated as 43°, 53°, and 61°, respectively.

Under the degenerate four-wave mixing configurations, in general, 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. Among the six sets of the 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 [22, 28]. If the transmission grating is dominant, the four beams can write the following grating, whose vector is given by

$$\mathbf{K} = k_1 - k_3 = k_4 - k_2. \quad (7)$$

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

$$n = n_0 + \frac{n_1 \exp(i\phi)}{2} \frac{E_1 E_3^* + E_2^* E_4}{I_0} \exp(i\mathbf{K} \cdot \mathbf{r}) + \text{c.c.} \quad (8)$$

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

$$(\nabla^2 + \omega^2 \mu \epsilon) E = 0. \quad (9)$$

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:

$$\left| \frac{d^2}{dz^2} E_j \right| \ll \left| \beta_j \frac{d}{dz} E_j \right| \quad (10)$$

where

$$\beta_j = k_j \cos \theta. \quad (11)$$

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

coupled equations:

$$\frac{dE_1}{dr} = -\frac{\alpha}{2} E_1 \quad (12)$$

$$\frac{dE_2}{dr} = \frac{\alpha}{2} E_2 \quad (13)$$

$$\frac{dE_3}{dr} = \frac{\alpha}{2} E_3 + i\pi n_1 \frac{\exp(-i\phi)}{\lambda} \frac{E_1 E_4^* + E_2^* E_3}{I_0} E_2 \quad (14)$$

$$\frac{dE_4^*}{dr} = \frac{\alpha}{2} E_4^* + i\pi n_1 \frac{\exp(-i\phi)}{\lambda} \frac{E_1 E_4^* + E_2^* E_3}{I_0} E_1^* \quad (15)$$

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

$$E_3 = i c_\gamma E_1(0) \int_r^d \frac{\exp[(\gamma + \alpha/2)(r - r')]}{I_0(r')} dr' \equiv c_\gamma E_1(0)/J(r) \quad (16)$$

where

$$\gamma = \frac{i\pi n_1 \exp(-i\phi)}{\lambda} \quad (17)$$

and

$$c = \frac{E_2(d) E_4^*(0) \exp(-\alpha d/2)}{\gamma I_1(0)/J(0) - 1}. \quad (18)$$

Taking the square of both sides of equation (16) one gets the intensity of the phase conjugation beam. The phase conjugate reflectance is defined as the intensity ratio of probe and phase conjugate beams. According to equation (16), the phase conjugate reflectance is strongly dependent on both the spatial phase shift (ϕ) and the index modulation depth (n_1). In figures 6(a) and 6(b) the phase conjugate reflectivities, which can be calculated from equation (16), are plotted versus the pump beam intensity ratio. The experimental data described in figure 6(a) were obtained when beams 1, 2 and 4 were of the same polarization state (p-polarization), [see figure 4(a)], while those in figure 6(b) were obtained when beams 1 and 4 were of the same polarization state (s-polarization) and the polarization direction of beam 2 was perpendicular to that of the other two beams, [see figure 4(b)]. In the case of the p-polarization state, the two-beam coupling between beams 1 and 4 were observed as shown in figure 5, while the two-beam coupling effects can be ignored when the beams 1 and 4 are of the s-polarization state. In the case of the s-polarization state, one may assume that the response is local and in the first approximation one may put $\phi = 0$ because the two-beam coupling effects can be ignored. In the case of the p-polarization state, the phase conjugate reflection must be calculated from equation (16)

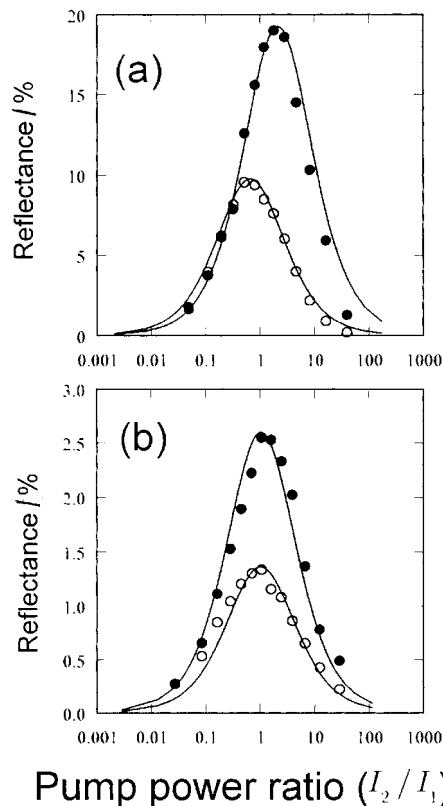


Figure 6. Phase conjugate reflectance versus pump power ratio. The probe beam intensity was 5.0 mW. The applied d.c. field was $0.4 \text{ V } \mu\text{m}^{-1}$ (filled circles) and $-0.4 \text{ V } \mu\text{m}^{-1}$ (open circles). (a) Beams 1, 2 and 4 were of the same polarization state (p-polarization); (b) beams 1 and 4 were of the same polarization state (s-polarization) and the polarization direction of beam 2 was perpendicular to that of the other two beams. The solid lines denote theoretical fitting results.

including the spatial phase shift obtained from the two-beam coupling experiments as shown in figure 3. According to equation (16), the dependence of phase conjugate reflectivity on the pump ratio is a manifestation of the same phase which produced the directionality in two-wave mixing. Since the sign of the spatial phase shift is reversed by changing the direction of the applied d.c. electric field, the photorefractive media with negative and positive phase shift display an optimum pump ratio of over [filled circles in figure 6(a)] and under unity [open circles in figure 6(a)], respectively. In contrast, as shown in figure 6(b), in the case of the s-polarization state, the photorefractive mesogenic composite displays an optimum pump ratio of around unity because the two-beam coupling effects can be ignored.

Both spatial phase shift ϕ and index modulation depth n_1 are dependent on the magnitude of the applied d.c. electric field. Figure 7 shows the phase conjugate reflectivity versus the pump ratio (I_2/I_1) for various

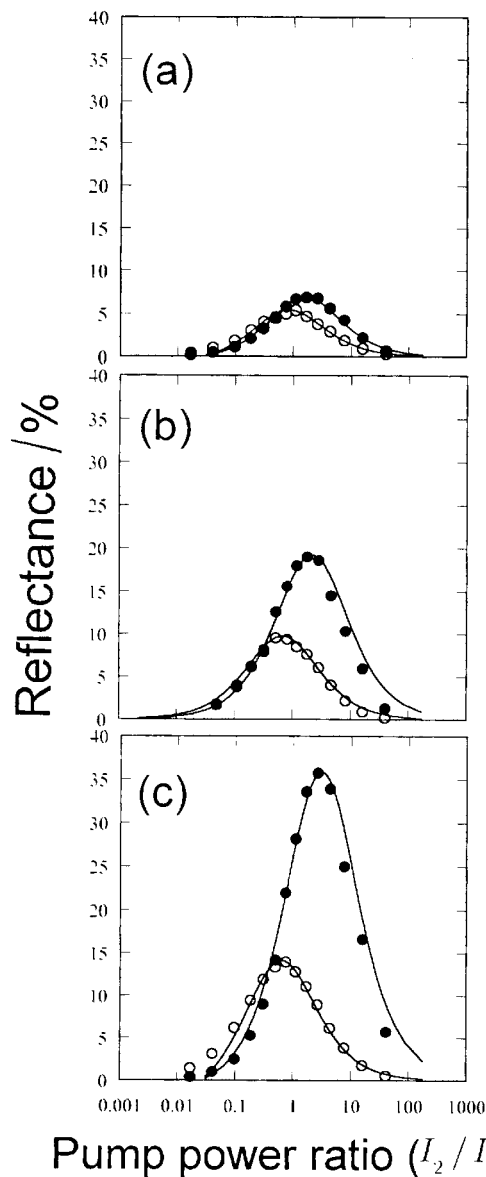


Figure 7. Phase conjugate reflectance versus pump power ratio. The probe beam intensity was 5.0 mW. The applied d.c. field was (a) $\pm 0.2 \text{ V } \mu\text{m}^{-1}$, (b) $\pm 0.3 \text{ V } \mu\text{m}^{-1}$ and (c) $\pm 0.4 \text{ V } \mu\text{m}^{-1}$. Beams 1, 2 and 4 were of the same polarization state (p-polarization). The solid lines denote theoretical fitting results.

magnitudes of the applied electric field. The solid curves show the calculated results according to equation (16) in the undeleted pumps approximation. Experimental data were fitted with the theoretical equation by using the observed spatial phase shift with good accuracy. The amplitude of the modulation depth increased as the applied electric field increased and these results are summarized in figure 8, which shows that the resultant phase conjugate reflectance increased as the applied electric field increased. We attribute the antisymmetric

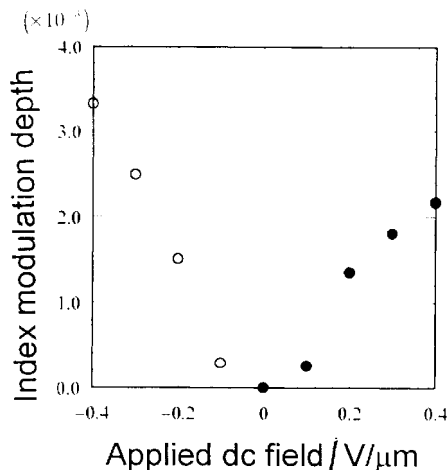


Figure 8. Refractive index modulation depth, which can be estimated from the phase conjugate reflectance, versus applied d.c. field.

electric field dependence to the antisymmetric experimental conditions. Under our experimental conditions shown in figure 4, since the sample cell was tilted from the bisector of the writing beams in order to provide a projection of the grating wave vector along the direction of the applied electric field, the effective value of the electric field should depend on its direction.

4. Conclusions

We have observed an optical phase conjugation effect from phase-shifted gratings in a photorefractive mesogenic composite. An efficient optical conjugated wave has been generated from spatial phase-shifted gratings linked with photoconductivity-enhanced molecular reorientations, as it is present only under the action of an externally applied electric field (orientational photorefractive effect). Degenerate four-wave mixing experiments with different polarization state were performed in order to differentiate the optical phase conjugation from the non-local gratings in the photorefractive mesogenic composite. Since the energy coupling in the media was strongly dependent on the polarization state, the characteristics of the optical phase conjugation were also dependent on the polarization state in these experiments. These results were quantitatively explained by spatial phase shifts, which were estimated from two-beam coupling experiments, and the coupled wave theory considering the non-local gratings.

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