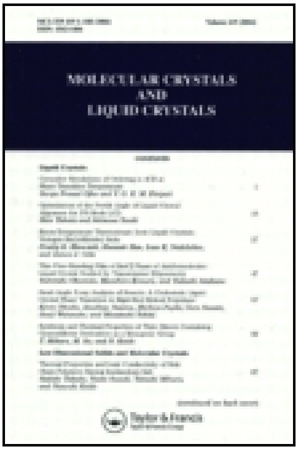

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Photorefractive Effect of Ferroelectric Liquid Crystals with Application of a Biased Alternating Electric Field

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The photorefractive effect of a ferroelectric liquid crystal (FLC) was investigated with application of a biased alternating electric field. A two-beam coupling gain coefficient of 96 cm^{-1} was obtained for a 10 μm-thick sample. The gain coefficient obtained was found to be much larger than those obtained with application of a DC electric field.

Keywords: biased AC field; ferroelectric liquid crystals; photorefractive effect; two-beam coupling

INTRODUCTION

Photorefractive materials have been attracting a great deal of interest, as they are potential candidate materials for devices producing dynamic holograms [1–6]. The photorefractive effect is defined as the optical modulation of the refractive index of a material. It has been known that organic materials show large photorefractivity. An organic photorefractive material is comprised of a photoconductive compound and a compound that exhibits an electro-optic effect. When laser beams interfere in a photorefractive material, a refractive index grating is formed. The mechanism responsible for the formation of this refractive index grating is the generation of a space-charge field (internal electric field) due to charge separation between the light and dark positions of the interference fringe and the subsequent change in the refractive index via an electro-optic effect (Fig. 1). A characteristic of

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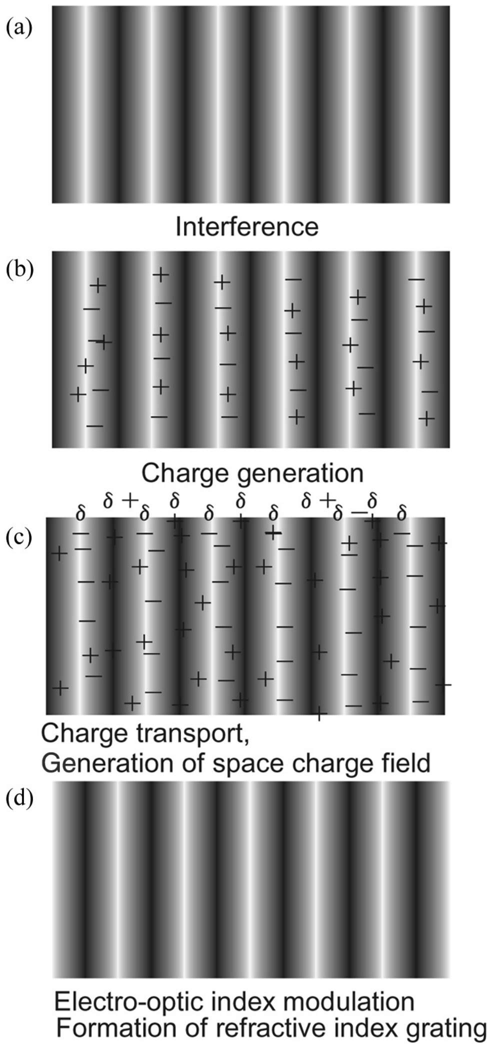


FIGURE 1 Schematic illustration of the mechanism of the photorefractive effect: (a) two laser beams interfere in the photorefractive material; (b) charge generation occurs at the light area of the interference; (c) while the electrons are trapped at the trap site in the light area, holes migrate by diffusion or drift in the presence of an external electric field and generates an internal electric field between the light and the dark positions; (d) the refractive index of the corresponding area is altered by the internal electric field.

the photorefractive effect is that the phase of the refractive index grating is shifted from the interference fringe. In such a phase-shifted index grating, laser beams undergo a unique mode of propagation. The interfering laser beams are energetically coupled in the photorefractive material through the phase-shifted index grating. The transmitted intensity of one beam through the material appears to increase, while that of the other appears to decrease. This phenomenon is

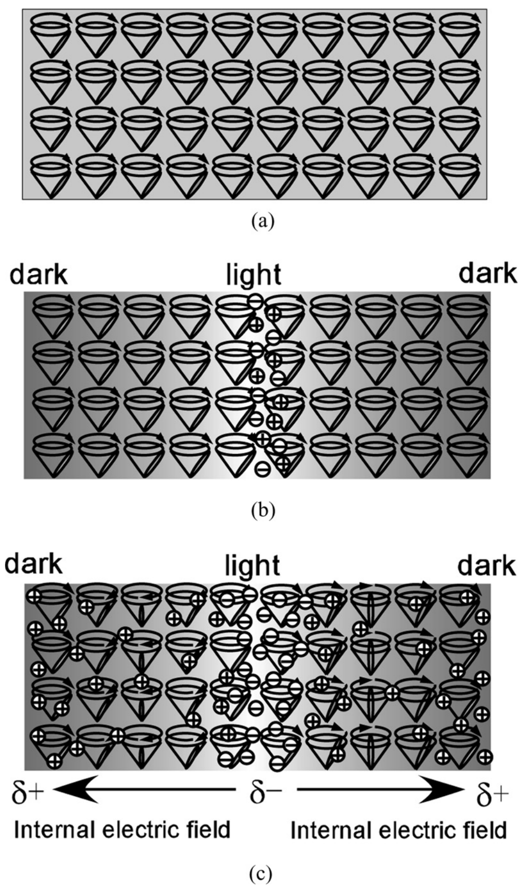


FIGURE 2 Schematic illustration of the mechanism of the photorefractive effect in FLCs upon application of an AC electric field: (a) positive and negative charges appear at the light positions of the interference fringe; (b) an internal electric field develops in the area between the light and dark positions of the interference fringe; (c) the rotational motion of FLC molecules in the corresponding area is biased by the internal electric field.

known as asymmetric energy exchange in photorefractive two-beam coupling. The photorefractive effect of ferroelectric liquid crystals (FLCs) has been investigated. FLCs belong to the family of chiral smectic C (SmC*) liquid crystals and have a layered structure [7]. When FLCs that are several microns thick are sandwiched between two glass substrates, they exhibit spontaneous polarization. FLCs in this state are called surface-stabilized ferroelectric liquid crystals (SS-FLCs). Since the switching of SS-FLCs originates from their bulk polarization, the switching response is extremely fast. The photorefractive effect of SS-FLCs under application of an alternating electric field (AC field) has been reported [8,9]. The rotational motion of FLC molecules under the influence of an AC field is modulated by an additional photo-induced electric field built at the interference fringe of two-laser beams (Fig. 2). This results in the formation of a hologram comprised of the periodic difference in the molecular motion of the FLC molecules. An asymmetric energy exchange is observed upon application of the AC field. This grating was interpreted to be based on the spatial difference in the molecular motion of the FLC molecules. The response time was in the order of a few tens of milliseconds, and was dominated by the formation of the internal electric field.

The photorefractivity of FLCs is accomplished through charge generation and diffusion. The application of an AC field on the FLC results in a very stable photorefractive response. However, it is obvious that an AC field is not advantageous for charge separation. In the current study, the effect of a biased AC electric field on the photorefractivity of SS-FLCs was investigated. Figure 3 shows the concept of applying a biased AC field.

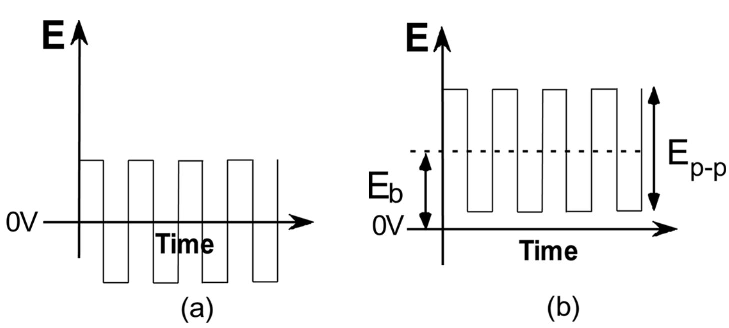


FIGURE 3 Concept of biased alternating electric field application: (a) alternating electric field; (b) biased alternating electric field.

EXPERIMENTAL

The FLC mixture used in this study was obtained from Clariant Japan (Felix SCE8). The physical properties of SCE8 are listed in Table 1. The FLC was doped with 2 wt% 9-ethyl-3-carbazolcarboxyaldehyde diphenylhydrazone (CDH) and 0.1 wt% 2,4,7-trinitro-9-fluorenone (TNF) as a sensitizer. The samples were injected into a glass cell having transparent electrodes and a polyimide alignment layer (LX-1400, Hitachi Chemicals Co., parallel rubbing). The thickness of the sample was controlled by the cell gap and was set to 10 μm. In order to form a highly homogeneous surface-stabilized state, the samples were heated to the isotropic phase temperature and deliberately cooled to the SmC* phase at a rate of 0.1°C/min. A uniformly aligned SS-state with few zig-zag defects was observed by examination under a polarizing microscope.

The photorefractive effect was evaluated using a two-beam coupling experiment. A p-polarized beam from an Ar^+ laser (488 nm, continuous wave) was divided into two by a beam splitter and interfered in the sample film. The intensity of the laser was 2.66 mW for each beam (1 mm diameter). An AC square-waveform electric field of $2.0\,V_{p\text{-}p}\,\mu m^{-1}$ with a bias field of 0 to $1.2\,V\,\mu m^{-1}$ and frequency of 0 to 10 kHz was applied to the sample using a function generator. The change in the transmitted beam intensities was monitored using photodetectors. The intersection angle ϕ of the two laser beams and the sample angle α (see Fig. 4a) were set to 20° and 50°, respectively. The signal arising from the diffracted beam was fitted using the square of a single exponential function [3],

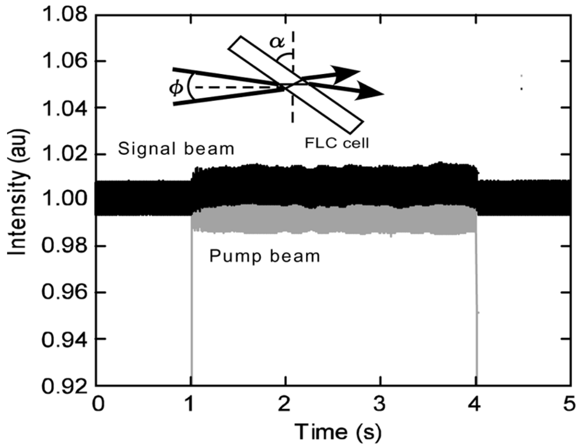
$$\gamma(t) - 1 = (\gamma - 1)[1 - \exp(-t/\tau)]^2 \tag{1}$$

where $\gamma(t)$ represents the transmitted beam intensity at time t divided by the initial intensity ($\gamma(t) = I(t)/I_0$), γ represents $\gamma(t)$ at the stationary state, and τ is the formation time. The gain coefficient was calculated from the γ value obtained from the fitted curve. The two-beam coupling gain coefficient, Γ, was calculated assuming Bragg diffraction,

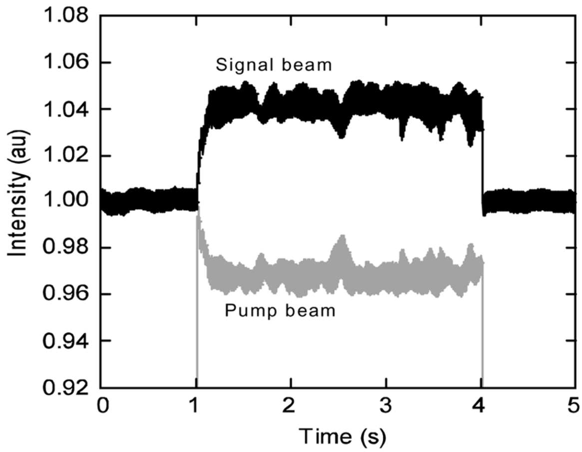
TABLE 1 Physical Properties of FLC

FLC	Ps at 25°C (nC/cm^2)	Phase transition temperature[a] (°C)	Response time[b] (μS)	Rotational viscosity (mPas)	Tilt angle (deg.)
SCE8	−4.5	−Sc* 60 S_A 80 N* 104 I	50	76	20

[a]C = crystal; Sc* = chiral smectic C phase; S_A = smectic A phase; N* = chiral nematic phase; I = isotropic phase.
[b]Response time to a 10 V/μm electric field at 25°C in a 2 μm cell.



(a)



(b)

FIGURE 4 Asymmetric energy exchange with a square waveform of $2.0\,\mathrm{V_{p\text{-}p}}\,\mu\mathrm{m}^{-1}$, 100 Hz; (a) without bias, and (b) with a bias of $0.5\,\mathrm{V}\,\mu\mathrm{m}^{-1}$. The intersection angle ϕ was 20°, and the sample angle α was 50°.

as follows [1,2],

$$\Gamma = \frac{1}{L}\ln\left(\frac{gm}{1+m-g}\right) \tag{2}$$

where g is the ratio of the intensities of the signal beam behind the sample with and without a pump beam, and m is the ratio of the beam intensities (pump/signal) in front of the sample.

RESULTS AND DISCUSSION

Two-Beam Coupling Experiment with Application of Biased AC Field

A typical example of the two-beam coupling signal observed with application of a $2.0\,V_{p\text{-}p}\,\mu m^{-1}$, 100 Hz square waveform electric field without a bias field ($E_b = 0$) is shown in Figure 4a. The transmitted intensities of the laser beams through the FLC/CDH/TNF mixture upon application of an alternating electric field as a function of time. Interference of the divided beams in the sample resulted in increased transmittance of one of the beams and decreased transmittance of the other. Although the transmitted intensity of the laser beam oscillates due to the switching motion of the FLC molecules, the average intensities of the beams were symmetrically changed, as shown in Figure 4a. This indicates that a grating based on the spatial difference in the rotational switching motion of FLC molecules was formed, and acted as a diffraction grating. The asymmetric energy exchange between the signal and pump beams proves the formation of a diffraction grating phase-shifted from the interference fringe. The grating is considered to be based on the spatial difference in the rotational switching motion of FLC molecules. When the bias electric field was set to $E_b = 0.5\,V\,\mu m^{-1}$, the two-beam coupling signal was dramatically enhanced (Fig. 4b). The gain coefficient increased from $10\,cm^{-1}$ ($E_b = 0$) to $70\,cm^{-1}$ ($E_b = 0.5\,V\,\mu m^{-1}$). Obviously, the bias field contributes to the formation of the diffraction grating. The internal electric field is thought to be difficult to form under an AC electric field. It has been reported that the ionic conduction is the major contributor to the formation of the space-charge field in the photorefractive effect of FLC [10]. The anisotropical mobility of ionic species in the LC medium affects the formation of the field. In the present experimental condition, the interference fringe is formed across the smectic layer, so that migration of ionic species occurs in the interlayer direction. The asymmetric structure of the surface stabilized state of the FLC may lead to asymmetric mobility of cations and anions. It was considered that the charge separated state is more effectively formed under the biased AC filed.

Influence of the Frequency

The influence of frequency was investigated using a square waveform electric field. The strength of the applied AC field was set to $2.0\,V_{p\text{-}p}\,\mu m^{-1}$ and the bias electric field was varied from 0 to $1.2\,V\,\mu m^{-1}$. The magnitude of the gain coefficient was measured by varying the frequency from 70 Hz to 10 kHz. Figure 5 shows the magnitude of

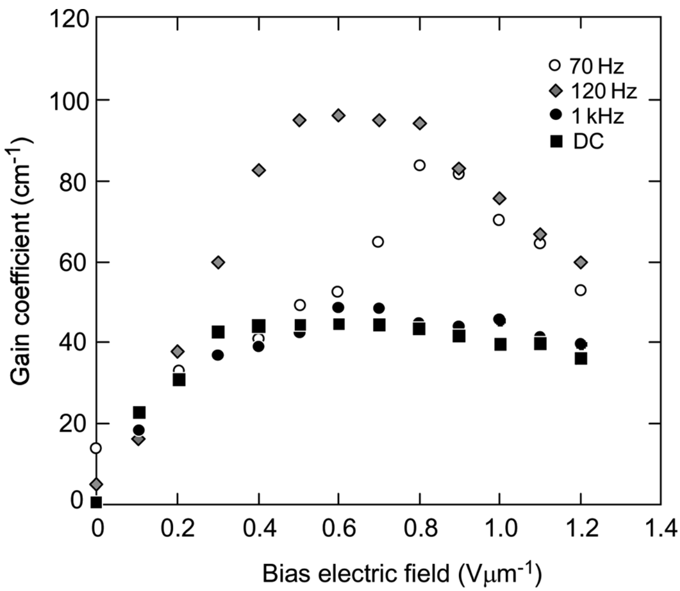


FIGURE 5 Gain as a function of the bias (DC) electric field. The frequency was varied from 70 Hz to 10 kHz.

the gain coefficient as a function of the bias field strength. The magnitude of the gain coefficient was dependent on both E_b and the frequency. A maximum gain coefficient of 96 cm^{-1} was obtained with application of a 120 Hz, 0.6 V μm^{-1} bias field. The gain coefficients obtained under a biased AC field were much larger than those obtained under application of a DC field (Fig. 5(■)). The frequency dependence of the gain coefficient can be attributed to the frequency dependence of the switching motion of the FLC. At frequencies of 70–120 Hz, the FLC molecules can respond to the applied AC field and exhibit the complete switching motion. However, when the frequency is higher than 1 kHz, the FLC molecules cannot switch completely and take on a vibrational motion. The gain coefficient increased as the bias field increased from 0 to 0.8 V μm^{-1}, and decreased when the bias field exceeded 0.8 V μm^{-1}. The bias electric field, in its lower range, can promote charge separation, which accelerates the grating formation. However, the bias electric field also has a simultaneous binding effect on the FLC molecules. At higher bias field, this binding effect becomes dominant and restricts the switching motion of the FLC molecules. The grating formation time had a minimum value of 18 ms at a frequency of 70 Hz and bias field of $1.2\,\mathrm{V\,\mu m^{-1}}$.

CONCLUSION

The photorefractive effect of a ferroelectric liquid crystal upon application of a biased AC field was investigated. The rotational motion of FLC molecules under the influence of an alternating electric field was modulated by an additional photo-induced electric field built at the interference fringe of two laser beams. Asymmetric energy exchange was dramatically enhanced by the application of biased AC field. It was considered that the biased AC field promoted the formation of the internal electric field. In comparison with the photorefractive effect of organic materials, a large two-beam coupling gain and fast response can be easily obtained for FLCs with application of a biased AC field.

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