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# Non-linear polarization effects in guest–host liquid crystals

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We report on an experimental investigation of the non-linear polarization effect in guest–host nematic liquid crystals (GH-NLCs) with uniaxial alignment. The degree of polarization of a laser beam transmitting the GH-NLCs and an aperture behind them is strongly dependent on the incident beam intensity. The mechanism is discussed on the basis of the laser-induced photothermal self-phase modulation. The origin of the effect is a strong anisotropy of the intensity-induced change or refractive indices.

## 1. Introduction

Spatial self-phase modulation of light is a typical type of non-linear wave propagation and has been studied under various contexts [1–3]. The effect is often responsible for optical damage of transparent materials, is a limiting factor in the design of high power laser systems, and sometimes plays an important role in the occurrence of the other physical processes in optical media. Because of their large refractive index anisotropy, large laser-induced refractive index change and ease of fabrication, nematic liquid crystals (NLCs) are emerging as a very important class of non-linear optical materials for use in the technology of photonics [4–11]. An optical limiting device for CO<sub>2</sub> laser applications using a 100 μm thick DLC film and thermally-induced spatial transverse self-phase modulation has been reported by Durbin *et al.* [1]. We recently showed that guest–host NLCs (GH-NLCs) can be sensitized for photothermal refractive index change at a desired wavelength by doping them with proper sensitizing dyes as guest molecules [12–14]; we then applied GH-NLCs to controllable optical limiting devices for lasers [14].

In this paper we demonstrate the non-linear polarization effect in GH-NLCs. In our materials, the refractive indices were changed by pure photothermal effects without the reorientation of the NLC molecules [12]. The non-linear polarization effect originated in the large photothermal refractive index change and the strong refractive index anisotropy in the GH-NLCs. The degree of polarization of the transmitted laser beam through the GH-NLCs

and an aperture placed behind them depended on the light intensity, a result of the strong anisotropy of photothermal effects in the GH-NLCs.

## 2. Experimental

Poly(vinyl alcohol) (PVA) was used as an alignment layer for the GH-NLCs. The NLC mixture (ZLI-2061) was obtained from Merck Japan Ltd. Commercially available *N,N*-dimethyloaniline (phenol blue) was obtained from Aldrich Co., Ltd. as the guest dye. One mg of phenol blue was dissolved in 1 g of ZLI-2061 at 100°C. The resulting GH-NLCs were sandwiched between two parallel transparent glass substrates coated with PVA films. The inner surfaces of two substrates were unidirectionally rubbed (anti-parallel cell) and the distance of 100 μm between them was maintained by polyester spacers. A monodomain structure of our GH-NLCs was observed using a polarizing microscope. A highly homogeneous alignment of both NLC and dye molecules was achieved as a result of the guest–host effect. Due to this alignment of dye molecules, the cell showed strong dichroism as indicated in figure 1.

Figure 2 shows the experimental set-up for the measurement of non-linear polarization effects in the GH-NLCs. A linearly polarized 632.8 nm He-Ne laser output was passed through a quartz depolarizer (Sigma Koki Co., Ltd.; C-178) to obtain a randomly polarized beam. The laser beam was focused to an  $e^{-2}$  diameter of 760 μm at the GH-NLCs cell. The incident beam intensity was controlled by a variable ND filter and changed between 0 and 19 mW. When the non-linear polarization effect

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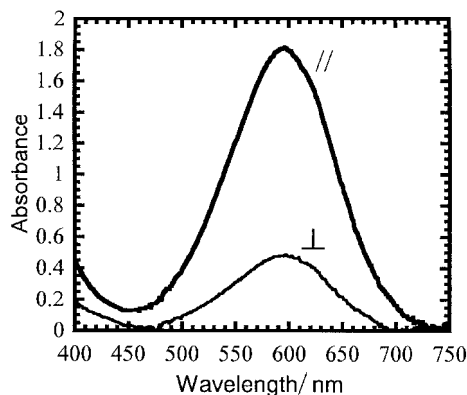


Figure 1. Absorption spectra of the guest–host liquid crystal.  $\parallel$  and  $\perp$  correspond to the polarization directions of the incident light, being parallel and perpendicular to the rubbing direction, respectively.

was observed, the laser beam passed through the GH-NLC, an aperture, and a Glan–Thompson prism placed behind the GH-NLC. The resulting transmitted laser beam was detected by a silicon photodiode. In order to observe the far-field pattern of the transmitted laser beam, the aperture was removed and the transmitted beam pattern was projected onto a white paper; the image was recorded by a CCD camera.

### 3. Results and discussion

Figure 3 shows a typical plot for transmitted light intensity versus incident light intensity.  $\parallel$  denotes the transmitted light intensity for the laser polarization parallel to the nematic director, and  $\perp$  denotes perpendicular polarization. At low intensity the transmitted light intensity for both parallel ( $I_{\parallel}$ ) and perpendicular polarization ( $I_{\perp}$ ) were proportional to the incident light intensity. However the transmitted light intensity was not proportional to the incident light intensity in the high incident power region. Figure 4 shows degree of polarization versus incident light intensity, where the percentage degree of polarization is defined by,

$$P = \frac{I_{\perp} - I_{\parallel}}{I_{\perp} + I_{\parallel}} \times 100. \quad (1)$$

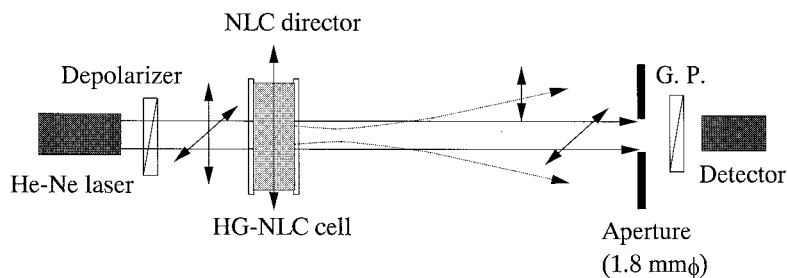


Figure 2. Experimental set-up for observation of the non-linear polarization effect. G.P. = Glan–Thompson prisms.

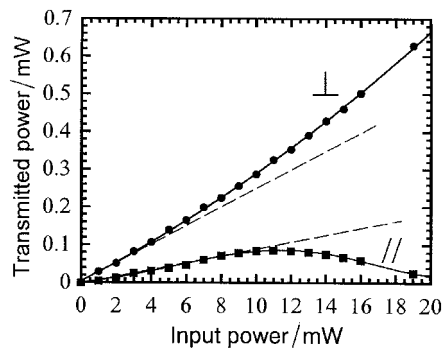


Figure 3. Transmitted light intensity versus incident light intensity.  $\parallel$  and  $\perp$  denote the transmitted light intensity for the parallel and perpendicular polarization direction of Glan–Thompson prism to the nematic director, respectively.

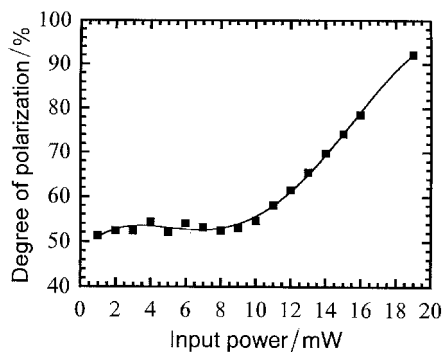


Figure 4. Degree of polarization of transmitted light versus incident light intensity.

The polarization direction of the incident laser beam was nearly random ( $P \cong 0$ ) because the beam was passed through the depolarizer. Since the cell showed strong dichroism (as shown in figure 1) due to the alignment of dye molecules, once the beam passed through the GH-NLC, the degree of polarization was about 50% in the linear region (0–6 mW) at the beginning of the curve. The degree of polarization increased with increasing input laser power due to the non-linear polarization effect; we obtained close to 100% degree of polarization in the case of 19 mW of incident laser power.

The non-linear polarization effect can be explained by the complex non-linear susceptibilities originating in the laser-induced photothermal effects in the GH-NLC. In a recent study, we determined a complex photothermal refractive index change in ZLI-2061 doped with phenol blue using a Mach–Zehnder interferometer [15]. In principle, both the real and imaginary parts of the non-linear susceptibilities can modify the transmitted light intensity distribution in space. However, according to our experimental results, the absolute value of the real part was about ten times that of the imaginary one. Consequently, we attribute the non-linear polarization effects to the real part of the refractive index change originating in the photothermal effects. The real part of the extraordinary non-linear susceptibility was negative, while the ordinary one was positive [15]. Since the refractive index change was proportional to the incident beam intensity [15] and the incident beam showed a Gaussian intensity profile, the phase increment  $[\Delta\phi(r)]$  should start to appear at the inflection point in the transmitted intensity.

The spatial self-phase modulation is caused by  $\Delta\phi$  and the diffracted beam originating in the self-phase modulation is theoretically explained using Kirchoff's diffraction integral [16, 17]. According to the theory, if the non-linear susceptibility is negative (extraordinary refractive index change case) and the maximum phase increment  $[\Delta\phi(r)]_{\max}$  is much larger than  $2\pi$ , a diffraction ring pattern is observed and the number of bright rings is approximately given by the integer closest to but smaller than  $[\Delta\phi(r)]_{\max}/2\pi$ . The diameter of the outermost ring in the far-field pattern is determined from the maximum slope of  $\Delta\phi$  at the inflection point. According to these considerations, in the case of negative non-linear susceptibility, the on-axis region is darkened and a doughnut-shaped pattern is obtained. In addition, since the diameter of the outermost ring depends on the maximum slope of  $\Delta\phi$ , the diameter increases with increasing input of laser power. On the other hand, if the non-linear susceptibility is positive (ordinary refractive index change case), the self-focusing processes are caused by a 'positive lens' originating in the positive non-linear susceptibility. In this case, the higher intensity central part of the beam should experience a larger refractive index than the edge, and thermal self-focusing can occur in the GH-NLC cell.

Figure 5(a) shows a typical far-field pattern seen when the aperture, Glan–Thompson prisms and silicon photodiode are removed. Both the Gaussian-like (the central part in the far-field pattern) and doughnut-shaped (the surrounding part in the far-field pattern) patterns were observed. The two types of patterns were related to the polarization direction. The far-field pattern in the case of polarization parallel to the NLC director, and

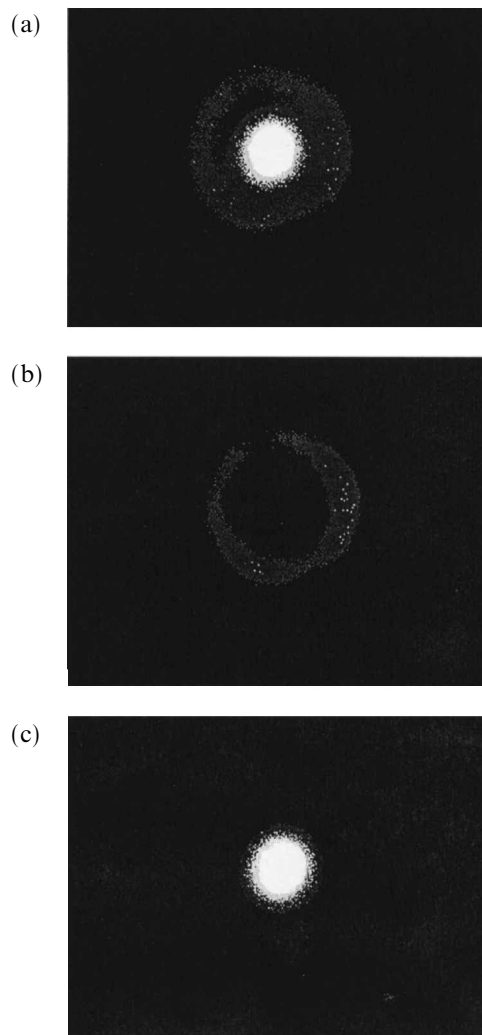


Figure 5. Far-field patterns without the aperture and Glan–Thompson prisms (a), and with Glan–Thompson prisms, (b) and (c). In the case of (b), the polarization direction of the Glan–Thompson prisms is parallel to the nematic director, while in the case of (c) it is perpendicular.

that in the perpendicular case are shown in figures 5(b) and 5(c), respectively. We attribute this behaviour to the different sign of the non-linear susceptibilities of the extraordinary and ordinary refractive index change. Since the non-linear susceptibility for the extraordinary refractive index change is negative, incident laser light having its electric field component parallel to the NLC director is diffracted to the doughnut-shaped pattern. In contrast, inasmuch as the non-linear susceptibility for the ordinary refractive index change is positive, incident laser light having its electric-field component perpendicular to the NLC director is self-focused. If the central part of the far-field pattern is separated in space by the aperture, the beam polarized parallel to the NLC director

is isolated and only the perpendicular polarized component can pass through the aperture in the case of an incident beam with high intensity, as shown in figures 3 and 4.

#### 4. Conclusion

In conclusion, we observed the non-linear polarization effect in guest–host nematic liquid crystals (GH-NLCs) with a uniaxial alignment. The degree of polarization of a laser beam passing through the GH-NLC cell and an aperture strongly depended on the incident beam intensity. The mechanism is discussed on the basis of photothermal spatial self-phase modulation with strong anisotropy. Due to the anisotropy, the far-field pattern from the GH-NLC cell consisted of two kinds of polarized light: doughnut-shaped and Gaussian-like patterns. The polarization direction of the doughnut-shaped beam was polarized parallel to the NLC director, while that of the Gaussian-like beam was perpendicular. The origin of this behaviour was a strong anisotropy of the linear and non-linear indices of GH-NLCs and the nonlinear polarization effects resulting from the anisotropy.

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