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Observation of spatial distribution of laser-induced refractive index change in dye-doped liquid crystals

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Spatial distributions of photothermal refractive index changes in dye-doped liquid crystals were determined by an optical interferometric method. The refractive index change of the order of 10^{-1}, including spatial distribution, was estimated by the described experimental technique. The absolute value of the refractive index change was proportional to the pump beam power, and the diameter of the index distribution was slightly larger than that of the pump laser beam due to heat conduction.

1. Introduction

A wide range of applications has arisen that benefit from the ability of liquid crystals to change their birefringence in an electric field [1]. In recent years this range has been extended by the discovery of new phenomena: the orientational photorefractive effects [2–4], photothermal refractive index change [5–7], and photoinduced phase transition [8–10]. Dye-doped liquid crystals exhibit a myriad of optical effects which are intrinsic to their unique nature. These useful non-linear optical properties in the dye-doped liquid crystals will play an important role from the viewpoint of photonics applications. In order to realize these applications, it is important to investigate the mechanism of laser-induced refractive index change in dye-doped liquid crystals. For this purpose, several types of experiments have been performed in previous work. One sophisticated method is to observe laser-induced birefringence by using a linear probe beam with crossed polarizer configuration [8–10]. Although laser-induced birefringence has been roughly estimated by means of this configuration, the spatial distribution of laser-induced refractive index change has not been characterized. Another method is to characterize the self-diffraction rings caused by extremely large self-phase modulation of the dye-doped liquid crystal [5–7]. When the refractive index change is negative and the maximum phase increment $[\Delta\phi]_{\max}$ is much larger than 2π, the number of bright rings is approximately given by the integer closest to but smaller than $[\Delta\phi]_{\max}/2\pi$, and the diameter of the outermost ring is determined from the maximum slope of $\Delta\phi$ at the inflection point. The spatial distribution of refractive index change was estimated by solving Kirchoff's diffraction integral including non-linear phase distribution. However, a large phase increment over 2π is necessary for accurate determination of the spatial distribution of the refractive index change, because the experimental diffraction patterns have to be theoretically fitted with Kirchoff's diffraction integral.

In the present work, by means of a Mach–Zehnder interferometer, we have investigated the spatial distributions of laser-induced refractive index change in a dye-doped liquid crystal. By using the optical interferometer, the spatial distributions of the laser-induced refractive index change were observed with high sensitivity and accuracy.

2. The method

In order to prove the validity of the characterization method described here, 4-pentyl-4′-cyanobiphenyl (5CB) was used as the host liquid crystal and 1-(*N*-methylamino)anthraquinone as the guest dye. The guest dye (2 mg) was dissolved in 5CB (1 g) at a temperature above the nematic–isotropic transition temperature (~35°C). The resulting dye-doped liquid crystal was sandwiched between two parallel transparent glass substrates. The inner surfaces of the two substrates were coated with poly(vinyl alcohol) films and rubbed unidirectionally. The space between the films was maintained by 50 μm thick polyester films, and homogeneous structure obtained. Strong anisotropy existed because the dye-doped liquid crystal was well aligned.

The experimental arrangement is schematically shown in figure 1. The major elements of our experimental apparatus are a Mach–Zehnder interferometer working with one collimated He-Ne laser beam (632.8 nm) as a

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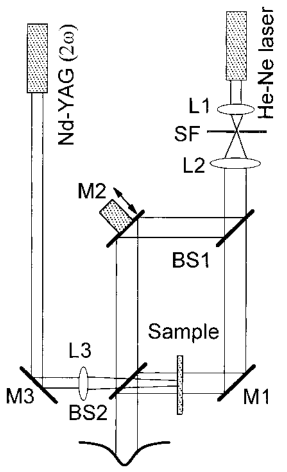

Figure 1. Experimental set-up for laser-induced refractive index change measurements, where M1, M2, and M3 denote mirrors, BS1 and BS2 beam splitters, L1, L2 and L3 lenses, and SF is a spatial filter.

probe beam and one frequency-doubled Nd-YAG laser beam (532 nm) as a pump beam. The dye-doped liquid crystal film was placed in one arm of the interferometer. The pump beam was focused to an e^{-2} diameter of 490 µm at the sample cell. Since the two beams (He-Ne laser beam) in the Mach–Zehnder interferometer are of the same frequency, a stationary interference pattern is formed and the electrical field of the two waves can be written as

$$E_1(x,y) = A_1(x,y)\exp[i\phi_1(x,y)]$$
$$E_2(x,y) = A_2(x,y)\exp[i\phi_2(x,y)] \qquad (1)$$

where A_1 and A_2 are the wave amplitudes, and ϕ_1 and ϕ_2 are the phases. The intensity of the electromagnetic radiation after interference can be written

$$I(x,y) = |E_1(x,y) + E_2(x,y)|^2. \qquad (2)$$

Using equation (1) for the electric field, the intensity can be written

$$I(x,y) = |E_1(x,y)|^2 + |E_2(x,y)|^2 + 2A_1(x,y)A_2(x,y)\cos\Phi(x,y) \qquad (3)$$

where

$$\Phi(x,y) = \phi_1(x,y) - \phi_2(x,y). \qquad (4)$$

The intensity of the light leaving the interferometer is a function of the phase difference between two arms, and the maximum value is obtained at $\Phi = 2m\pi$ (m = integer). The laser-induced change in the refractive index of the dye-doped liquid crystal is related to the change in phase ($\Delta\Phi$) as follows:

$$\Delta\Phi(x,y) = \frac{2\pi}{\lambda} l \Delta n(x,y) \qquad (5)$$

where λ is the wavelength of the probe beam and l is the thickness of the dye-doped liquid crystal film. After the phase difference between two arms of the interferometer was set to be $2m\pi$, the refractive index of the dye-doped liquid crystal was spatially modulated by the frequency-doubled Nd-YAG laser with a Gaussian profile. The intensity of the light leaving the interferometer can then be written

$$I(x,y) = |E_1|^2 + |E_2|^2 + 2A_1A_2\cos[\Delta\Phi(x,y)]. \qquad (6)$$

3. Results

Figure 2 shows the fringe patterns from the optical interferometer without the pump, (*a*), and with the pump beam, (*b*). Since the two beams were adjusted to follow the same course at an exit of the interferometer, and the phase difference between two arms was set to be $2m\pi$ by adjusting the position of M2, only one bright spot was observed in the fringe pattern as shown in

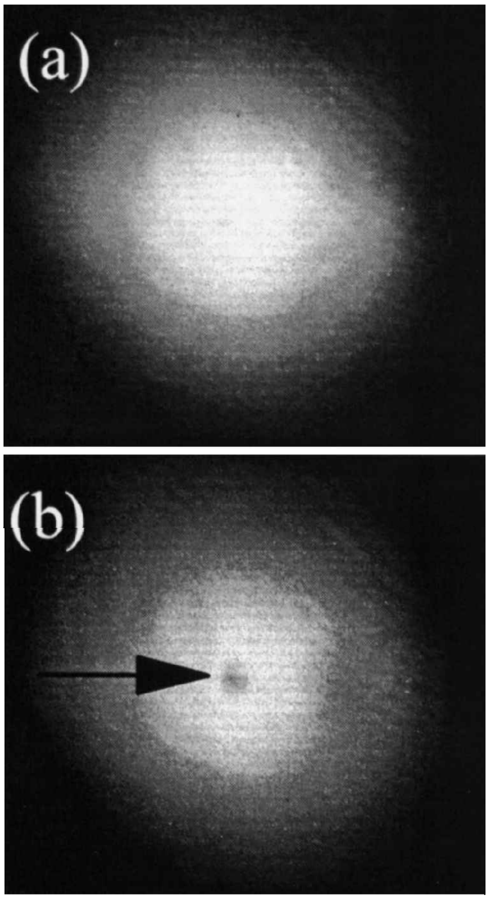

Figure 2. Interferometer fringe patterns (*a*) without pump and (*b*) with pump beam. The dark spot originating in a photothermal refractive index change is indicated by the arrow.

figure 2(*a*). When the pump beam was irradiated at the sample, according to equation (6), a dark spot appeared in the fringe pattern as shown in figure 2(*b*).

In order to characterize the distribution of the refractive index change quantitatively, the intensity distribution of the fringe pattern was measured as follows. The fringe pattern was cut by a pinhole and a guide to the photomultiplier by an optical fibre. The pinhole was set on the pulse stage, and one-dimensional scans of the intensity distribution were obtained by changing the pinhole position. Figure 3(*a*) shows the measured fringe pattern from the optical interferometer without the pump and with the pump beam. The intensity of the light after interference decreased under irradiation by the pump beam. Using equation (6), the spatial distribution of the laser-induced refractive index change was estimated as described in figure 3(*b*). Although the signal-to-noise ratio became worse for small index changes ($<10^{-3}$) due to instabilities of the optical interferometer and/or the limit of the performance of the detector, the distribution was close to Gaussian. As shown in figure 3(*b*), a refractive index change of the order 10^{-3}, including spatial distribution, was estimated by means of the experimental technique here described.

We previously reported that the laser-induced refractive index change could be estimated from the self-diffraction pattern due to photothermal self-phase modulation by means of Kirchhoff's diffraction theory, including non-linear phase change [6, 7]. In order to estimate the refractive index change with high accuracy, a large phase increment over 2π, or a refractive index change of the order 10^{-2} ($l = 50\,\mu\mathrm{m}$, $\lambda = 0.633\,\mu\mathrm{m}$), is necessary.

Figure 4(*a*) shows peak values of the refractive index change versus pump beam power. The absolute value of the refractive index change was proportional to the pump beam powder under our experimental conditions. In our previous publication [6], self-diffraction patterns originating from photothermal self-phase modulation were in good agreement with calculated results when the refractive index changes were assumed to be described by the equation $\Delta n = n_2 I + n_4 I^2$, where n_2 and n_4 are non-linear coefficients, and I is the light intensity. However, under the experimental conditions described in the present paper, since the pump beam intensity was weak, the higher order optical non-linearity could be ignored. The e^{-2} diameter versus pump beam intensity is summarized in figure 4(*b*). The diameter of the distribution of the refractive index change was slightly larger than that of the pump laser beam. Since the refractive index changes were caused by photothermal effects in the dye-doped liquid crystal, the generated heat was conveyed in the liquid crystal layer according to its thermal conductivity. It has already been reported that the photothermal phenomenon could be characterized

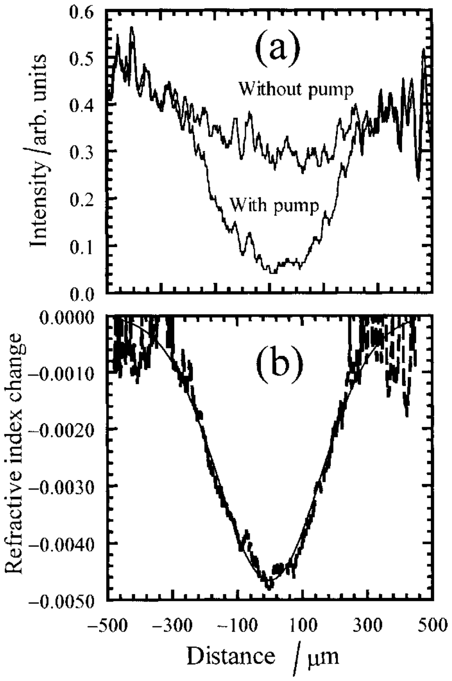

Figure 3. (*a*) Interferometer fringe patterns without pump and with pump beam. (*b*) Distribution of photothermal refractive index change in the dye-doped liquid crystal; the dotted curve denotes experimental data and the solid line represents the theoretical fitting curve.

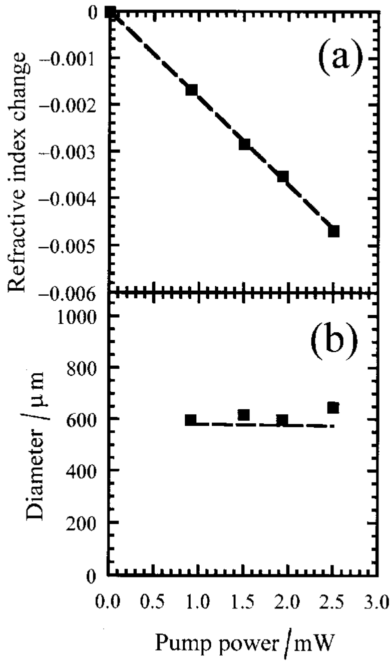

Figure 4. (*a*) Dependence of the refractive index change on the pump beam intensity. (*b*) Dependence of e^{-2} diameter of the index distribution on the pump beam intensity. The filled squares denote experimental data; the dotted lines represent theoretical curves obtained by the use of heat-conduction analysis.

by the use of heat-conduction analysis [7]. We confirmed that the experimentally determined diameter of the distribution of the refractive index change could be explained by the calculated results obtained from heat-conduction analysis as shown in figure 4.

4. Conclusion

The spatial distribution of photothermal refractive index change in a dye-doped liquid crystal has been determined by an optical interferometric method. For the realization of non-linear liquid crystal optical devices, it is important to discriminate between the different mechanisms, including photothermal, photoinduced reorientation, and photoinduced phase transition. The measurement technique described here should be applicable to other materials with different mechanisms.

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