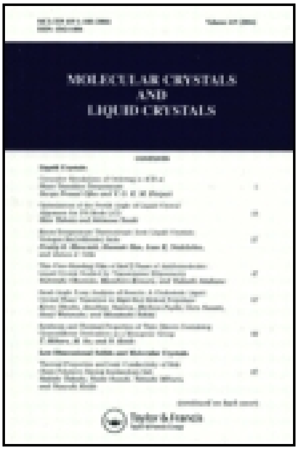

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

Self-diffraction Pattern Formation in Liquid Crystals on Dye-doped Polymer Surfaces

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Version of record first published: 24 Sep 2006

Mol. Cryst. Liq. Cryst., Vol. 325, pp. 137–144
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(Received 15 April 1998)

We report on diffraction rings and a He–Ne laser induced self-phase modulation due to photothermal effects in nematic liquid crystals (NLCs) on dye-doped polymer surfaces. A far-field pattern of the laser beam passed through the NLC cells depended on the polarization of the incident laser beam and the results were explained by sign of the refractive index change. Three kinds of NLCs with different nematic–isotropic transition temperatures (T_{NI}) were used and higher photosensitivity in low T_{NI} of NLCs was obtained than that in high T_{NI} of those.

Keywords: Liquid crystal; self-phase modulation; photothermal effect; dye-doped polymer

1. INTRODUCTION

Optical-field-induced refractive-index changes can have an appreciable effect on laser-beam propagation in a nonlinear medium [1]. This leads to such well-known phenomena as self-focusing and self-phase modulation. Self-phase modulation is typical of the type of nonlinear wave propagation that depends critically on the transverse profile of the beam. In several publications, the results of studies on self-phase modulation in organic materials have been presented in the last years [2–5].

Nematic liquid crystals (NLCs) in the mesophase have been found to exhibit extremely large laser-induced refractive index changes [2, 3, 6–10]. Such self-diffraction phenomena originating in self-phase modulation are

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readily observed through the thermally induced refractive index change in the NLCs and have been reported by several authors. Durbin *et al.*, reported that multiple diffraction rings originating in the self-phase modulation in the NLCs were formed by irradiation with a high-power Ar laser beam [2]. Khoo *et al.*, reported on thermally induced spatial transverse self-phase modulation and optical limiting of a CO_2 laser with a 100 μm NLC (E7) film [3]. Recently we demonstrated that high photosensitivity to a desired wavelength was obtained by doping with a small amount of dyes, which absorbed the laser beam [10]. The multiple diffraction ring pattern of He–Ne laser beam passed through the host–guest liquid crystals was observed since the maximum phase increment was much larger than 2π.

In the present study, we present our observation results of diffraction rings from the NLCs on dye-doped polymer surfaces and show that the results can be explained by the spatial self-phase modulation due to a photothermal refractive index change.

2. EXPERIMENTS

In order to investigate effects upon nematic–isotropic transition temperature (T_{NI}), three kinds of NLCs were used in this study. Three kinds of NLCs, *i.e.*, 5CB, E7 and ZLI2061, with different T_{NI}, were obtained from Merck Japan Ltd. The T_{NI} of the three kinds of corresponding NLCs, 5CB, E7 and ZLI2061, are 36, 59 and 95°C, respectively, according to the NLC physical characteristic data provided by the manufacture. Commercially available poly(methyl methacrylate) (PMMA) was obtained from Kodak Co., Ltd. and used as matrix polymer without further purification. *N*,*N*-dimethylindoaniline [phenol blue (PB)] was purchased from Aldrich Co., Ltd. and used as a doped dye. Poly(vinyl alcohol) (PVA) was used as a NLC alignment polymer layer.

A PB-doped PMMA film in this study was prepared by a spin-coating method using a chloroform solution at a weight ratio of PB and PMMA of 5:95. The solution was spin-coated on glass substrates and the film thickness was set to be about 4 μm. Figure 1 shows visible absorption spectra for the PB-doped PMMA film. PVA was suitable to the NLC alignment layer since PVA was dissolved in hot water while the PB-doped PMMA was not dissolved in water. This means that the polymer-layered structure is easily fabricated by spin-coating a PVA aqueous solution on the PB-doped PMMA film. In addition, it is well known that the NLC molecules are aligned on the rubbed PVA surfaces. PVA was dissolved in hot water and

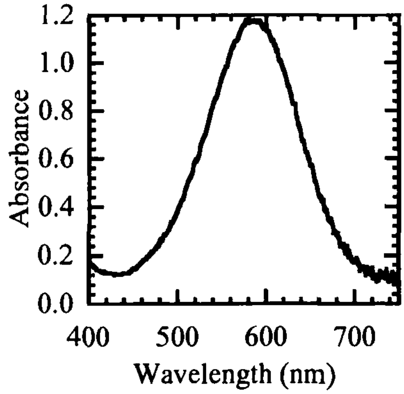

FIGURE 1 Visible absorption spectra for the dye-doped polymer film.

the solution was spun on the PB-doped PMMA film, forming an about 0.7 μm thick film. The resulting layered films were dried and two polymer coated glass substrates were unidirectionally rubbed with a silk cloth in the horizontal direction. A cell was mounted with anti-parallel rubbing directions and the NLCs were sandwiched with a 100 μm thickness polyester spacer. The test sample structure is shown in Figure 2 schematically. A monodomain structure of three kinds of our resultant NLC cells was observed by polarized microscope, and highly homogeneous alignment was obtained.

A linearly polarized He–Ne laser (632.8 nm) was used for exciting spatial self-phase modulation in the NLCs on the PB-doped PMMA films. The laser beam was focused to an e^{-2} diameter of 250 μm at NLC cells. The polarization direction of the laser beam was controlled by a half-wave plate.

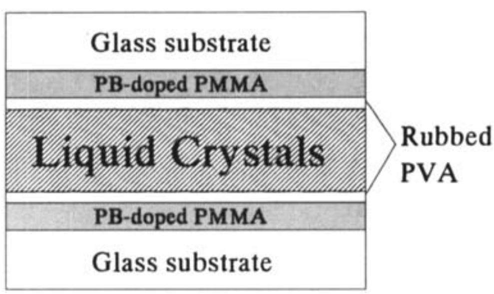

FIGURE 2 Schematic structure of the sample cell.

The polarization direction was parallel (extraordinary) or perpendicular (ordinary) to the NLC director. The laser beam was passed through the sample cell perpendicular to the glass substrate. Then the transmitted beam pattern (far-field pattern) was projected on a white paper and the image was recorded by a CCD camera.

3. RESULTS AND DISCUSSION

Figures 3(a) – 3(f) show the far-field patterns of the parallel [3(a) – 3(c)] and perpendicularly [3(d) – 3(f)] polarized beams passed through the 5CB-cell.

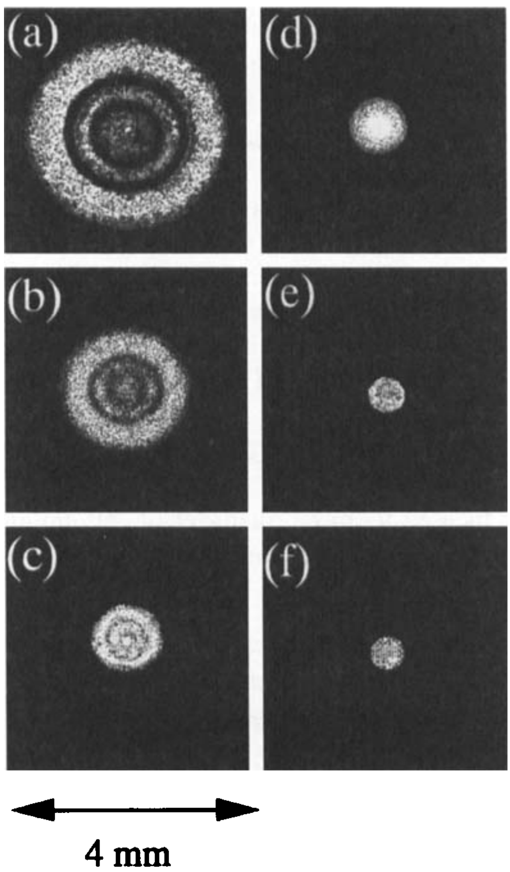

FIGURE 3 Far-field patterns in passage of the beam through the 5CB-cell. The observation was taken at a distance of 75, 50 and 25 mm, corresponding to [(a), (d)], [(b), (e)] and [(c), (f)], respectively. The beam intensity was 8 mW and the polarization direction of the laser beam was parallel [(a) – (c)] and perpendicular [(d) – (f)] to the NLC director.

The observation was taken at a distance of 25, 50 and 75 mm from the sample cell, respectively. The NLCs on the dye-doped polymer surfaces show large nonlinear effects and the beam characteristics were changed drastically in passage of the beam through the NLC cells. Since PB does not show any photochemical and/or photophysical changes to our knowledge and the NLC molecules are separated in space from the dye molecules by the PVA layer, we attributed the experimental results to temperature dependence of the refractive indices of the NLCs. It is well known that the gradient $\partial n_e/\partial T$ is negative, while $\partial n_o/\partial T$ is positive, where n_e and n_o are the extraordinary and ordinary refractive indices, respectively [10]. In case of a beam polarized parallel to the NLC director, numbers of the diffraction rings were observed. Since the refractive index change is negative in this case and the maximum phase increment $[\Delta\phi(r)]_{\max}$ is much larger than 2π, the number of the bright rings is approximately given by the integer closest to but smaller than $[\Delta\phi(r)]_{\max}/2\pi$ and the diameter of the outermost ring is determined from the maximum slope of $\Delta\phi(r)$. The diffraction half-cone angle is approximately given as [3]

$$\theta = [\Delta\phi(r)]_{\max}/(2\pi/\lambda) \tag{1}$$

Figures 4(a)–4(f) show the far-field patterns of the parallel and perpendicular polarized beam with several intensities passed through the E44-cell. The diameter of the outermost ring increased with increasing the light intensities. The results suggest that the value of $[\Delta\ \phi(r)]_{\max}$ increased with increasing the light intensity.

On the other hand, when the polarization direction of the incident beam was perpendicular to the NLC director, the far-field patterns of the transmitted laser beam can be explained by self-focusing and self-defocusing due to the positive refractive index change. In case of the positive refractive index change, the central part of the beam having a higher refractive index than the edge and the self-focusing and self-defocusing processes are caused by "a positive lens". Since the incident laser beam has the Gaussian profile, the central part of transmitted laser beam has a higher intensity than the edge as shown in Figures 3(d)–3(f).

Figures 5(a)–5(f) show the far-field patterns of the parallel and perpendicularly polarized beams passed through three kinds of NLCs, respectively. The NLCs with low T_{NI} show larger nonlinear effects than those with high T_{NI}. The beam characteristics were changed drastically in passage of the beam through the NLCs with low T_{NI} (5CB). Both the absolute value of

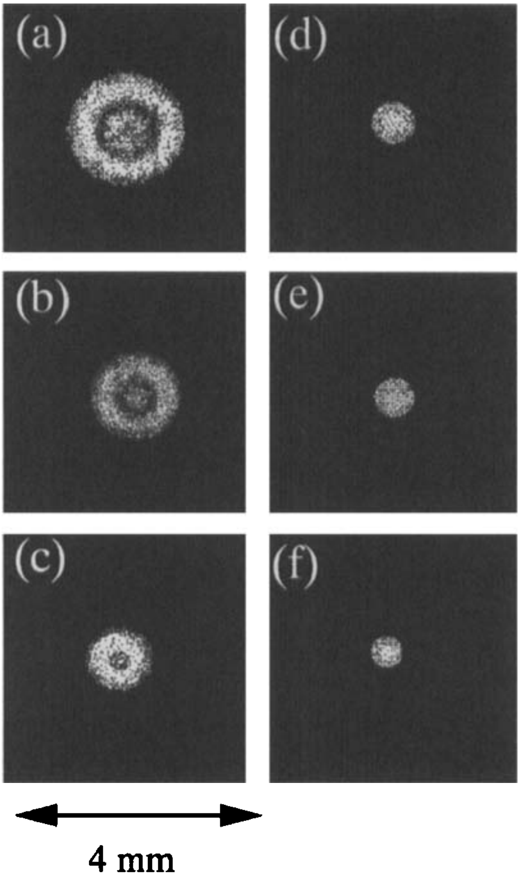

FIGURE 4 Far-field patterns in passage of the beam through the E7-cell. The beam intensity is 11, 8 and 5 mW, corresponding to [(a), (d)], [(b), (e)] and [(c), (f)], respectively. The polarization direction of the laser beam was parallel [(a)–(c)] and perpendicular [(d)–(f)] to the NLC director. The observation was taken at a distance of 75 mm from the sample.

$\partial n_e/\partial T$ and $\partial n_o/\partial T$ increase with decreasing $T - T_{NI}$ and change drastically with approach to T_{NI}. We attribute the large nonlinearity in the 5CB cell to the larger gradient $\partial n_e/\partial T$ compared with that in the ZLI2061 cell. In case of the negative refractive index change, the third order nonlinear refractive coefficient (n_2) can be roughly estimated as [11]

$$n_2 = \frac{N n_0 \lambda}{l E_0^2}, \tag{2}$$

where E_0^2 is peak intensity of the incident laser beam, l is thickness of the nonlinear medium, n_0 is the linear refractive index, and N is the total

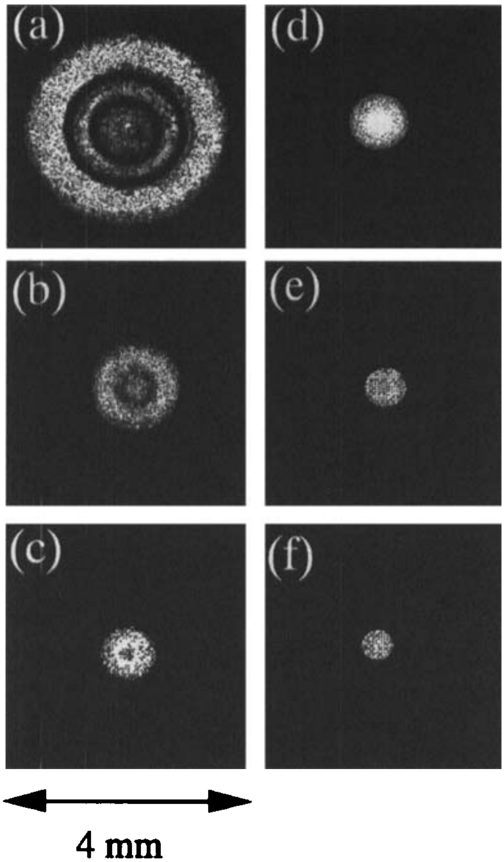

FIGURE 5 Profiles of beams passed through three kinds of NLCs *i.e.*, 5CB, E7 and ZLI2061, corresponding to [(a), (d)], [(b), (e)] and [(c), (f)], respectively. The beam intensity was 8 mW and the polarization direction of the laser beam was parallel [(a) – (c)] and perpendicular [(d) – (f)] to the NLC director. The observation was taken at a distance of 75 mm from the sample.

number of fringes. The value on n_2 for the 5CB, E7 and ZLI2061 cells are estimated to be about −0.028, −0.014 and −0.007, respectively. In case of a beam polarized perpendicular to the NLC director, as shown in Figures 5(d) – 5(f), the diameter of the transmitted laser beam increased with decreasing T_{NI}. The beam diameter depends on the focal distance of the positive lens originating in the positive refractive index change in the NLCs. The positive lens originating in self-phase modulation in the NLCs with low T_{NI} should show a shorter focal distance than that in the NLCs with high T_{NI}, inasmuch as the NLCs with low T_{NI} show large nonlinear effects.

4. CONCLUSIONS

In conclusion, we observed diffraction rings from the NLCs on the dye-doped polymer surfaces for the first time to our knowledge. The far-field patterns of a laser beam passed through the NLCs with different nematic–isotropic transition temperatures (T_{NI}) were shown in this study and we discussed that spatial self-phase modulation induced by a photothermal refractive index change could explain the results. The far-field patterns of the parallel and perpendicularly polarized beam with respect to the NLC director could be explained by the sign of the refractive index change. The nonlinear coefficient increased with decreasing T_{NI} of the NLCs. We attributed this large coefficient to the large gradient $\partial n/\partial T$ in the NLCs with low T_{NI}. In case of the negative refractive index change, the value on n_2 for the 5CB, E7 and ZLI2061 cells are estimated to be about −0.028, −0.014 and −0.007, respectively.

Acknowledgements

We wish to thank Professor K. Sakuda of Nagaoka University of Technology for useful suggestions.

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