

Change of Birefringence in Dye-Doped Nematic Liquid Crystals Under Laser Illumination

S. Eren SAN, Oğuz KÖYSAL, Serhat ÖZDER, F. Necati ECEVİT

Department of Physics, Gebze Institute of Technology,

P. O. Box 141, Gebze 41400, Kocaeli, TURKEY

e-mail: erens@gyte.edu.tr

Received 09.06.2003

Abstract

Birefringence property of E7 nematic liquid crystal is investigated via voltage dependent transmittance spectrums. Measurements are performed at wavelength 632.8 nm, which is the absorbance peak of our sample including anthraquinone derivative Disperse Blue 14. Results of dye-doped samples are compared with those undoped for dark and laser illuminated cases. It was observed that birefringence is dependent on laser illumination for dye-doped samples, up to a threshold voltage after which it is constant.

Key Words: Dye-doped liquid crystals; Transmittance; Birefringence; Molecular reorientation.

1. Introduction

Liquid crystals (LC) are highly nonlinear optical materials and are very sensitive to their optical environments. Several nonlinear mechanisms have revealed the promising characters of these materials. The difference in refractive indices, for fields polarized along, and perpendicular to, the director axis brings about a large birefringence property, $\Delta n = n_{\parallel} - n_{\perp}$, observable from visible to infrared. This property is an opportunity for various potential applications [1]. Director axis reorientation-based effects causing a change of refractive index and observations of several interesting dynamic and storage wave-mixing effects, have also been extensively studied so far [1-3]. It is experimentally proved that doping with a small amount of dye decreases the required threshold of molecular reorientation in dye-doped nematic LC. This phenomenon has potential application such as holographic data storage. Birefringence property and its dependency on molecular reorientation play an important role in understanding the molecular reorientation mechanisms. Moreover, birefringence enhancement is of primary importance for the innovation of different electro-optic applications [4, 5]. In this study, evaluation of birefringence dependency upon absorbance characteristics of sample is examined.

2. Method and Experiment

Nematic LC are rod-like molecules and they exhibit orientated order (on average) along a common direction known as director \hat{n} . If the polarization axis of the laser is tilted 45° with respect to the director axis of the LC molecules, polarized laser light propagating along the LC experiences a phase difference δ between the ordinary and extraordinary components of the propagating light. The fundamental principle of birefringence measurement is to measure this phase difference δ and an accurate way of determining δ is to measure the $I_{\perp} / I_{\parallel}$ ratio, where I_{\perp} and I_{\parallel} are the filtered intensity values of perpendicular and parallel

polarized laser beams, respectively. Once this ratio is determined, the following equations are employed for the evaluation of δ [6]:

$$|\delta| = N\pi + 2 \tan^{-1} \sqrt{\frac{I_{\perp}}{I_{\parallel}}}, \quad N = 0, 2, 4, \dots \quad (1a)$$

$$|\delta| = (N + 1)\pi - 2 \tan^{-1} \sqrt{\frac{I_{\perp}}{I_{\parallel}}}, \quad N = 1, 3, 5, \dots \quad (1b)$$

N is determined from the relationship between the number of peaks in I_{\perp} , I_{\parallel} and applied voltage, V and birefringence Δn is determined according to the following equation by using the δ values that are obtained from Eq. (1a), (1b):

$$\delta = \frac{2\pi d \Delta n}{\lambda}. \quad (2)$$

Here, d is the thickness of the cell and λ is the wavelength of the probe laser. The sample is rotated 45° in plane. An analyzer is placed in front of the detector so that vertical and parallel transmissions, which are used in the calculation of birefringence, are filtered. Details of the optical configuration is depicted in Figure 1. An auxiliary He-Ne laser is used as a pumping source to observe the absorbance based birefringence modulation. Polarizations of the lasers are perpendicular to the plane of incidence and power of the pumping laser is around 40 mW, while the probe power is ~ 1 mW. The angle between the probe laser and pumping laser is arranged to be $\sim 1^\circ$.

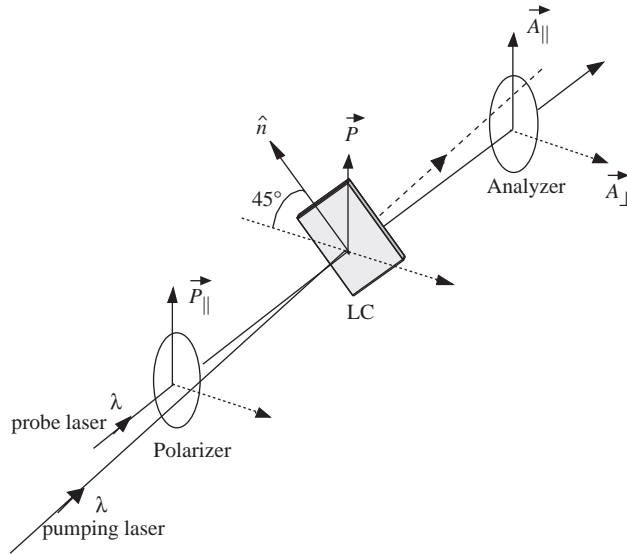


Figure 1. Optical configuration of birefringence measurement. P: Polarizer; A: Analyzer; \hat{n} : Director; λ : wavelength of the incident laser beams.

The nematic host E7 was supplied from Merck and the doping dye, Disperse Blue 14, was provided by Radiant Color. Dye was added to E7 in 1% w/w ratio for the preparation of the sample. Chemical formula of the dye and spectroscopic information of the dye-doped sample is given in Figure 2. It can be seen that the absorbance takes place at the wavelength of a He-Ne laser at around 632.8 nm. The measurement cell was made up of two glass slides separated by Mylar sheets of appropriate thickness. Before constructing the cell, glass substrates were spin coated with Polyvinyl alcohol (PVA) at 2000 rpm and were cured at 50°C

for ~ 2 hours. The coating was deposited to a thickness of ~ 100 nm, then subjected to a surface treatment of unidirectional rubbing with velvet in order to develop conditions for molecular orientation. Spacing of the cells is measured by a discrete Fourier transform technique described in [7] prior to the filling process. Three cells are produced, two of which are $5 \mu\text{m}$ thick and are used to characterise the birefringence dependence on voltage, respectively for doped and undoped cases. The other cell is $100 \mu\text{m}$ in thickness and it is used for the measurement of birefringence dispersion in the visible region. Both cells are planar with 2 degree rubbing tilt on their PVA coating and are filled by capillary action with the sample mixed in water bath at 80°C .

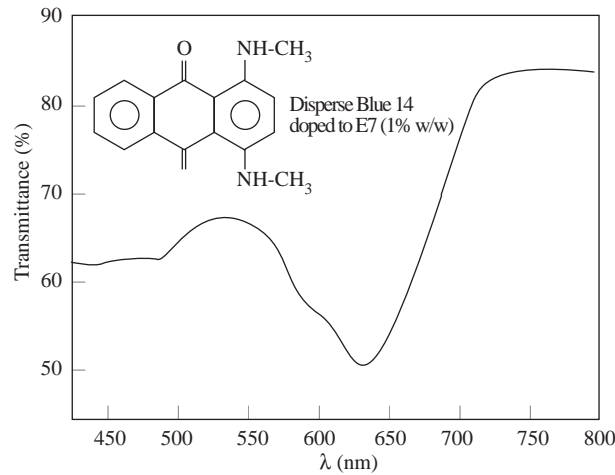


Figure 2. Transmittance spectrum of dye-doped liquid crystal sample in visible spectral range, and the chemical formula of the doping dye (shown in the inset).

3. Results and Discussion

Birefringence dispersion of the undoped sample was performed by Fringe counting technique [8] from the transmittance spectrum given in Figure 3a and it is fitted to Cauchy's formula of the form

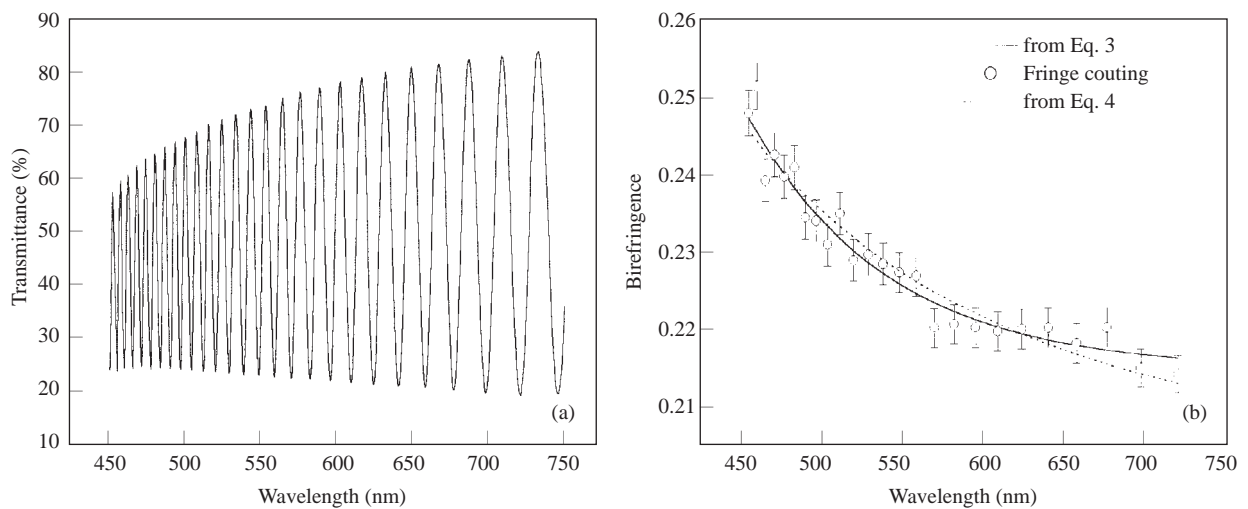


Figure 3. (a) Transmittance spectra of nematic LC E7 in visible region for $d = 100 \mu\text{m}$ cell. (b) Wavelength dependency of birefringence at $T = 25^\circ\text{C}$ by Fringe counting technique (open circle), and fitted data using Eq. (3) (solid line) and Eq. (4) (dotted line).

$$\Delta n(\lambda) = A + (B/\lambda^2) + (C/\lambda^4). \quad (3)$$

Here A , B , and C are constants that are characteristics of the film [9] and are found to have the following values: $A = 0.21437$, $B = -6.6932 \times 10^3 \text{ nm}^2$ and $C = 2.5906 \times 10^9 \text{ nm}^4$. Equation (3) is plotted in Figure 3b and is shown as the solid line. Δn can also be described by an equation derived by Wu [4] as

$$\Delta n = G \frac{\lambda^2 (\lambda^*)^2}{\lambda^2 - (\lambda^*)^2} \quad (4)$$

and is also shown on Figure 3b for comparison. Here, λ^* is the mean resonance wavelength and G is a constant and have been found with the values $G = 4.63 \times 10^{-6} \text{ nm}^2$, $\lambda^* = 205.4 \text{ nm}$ for E7 LC. The values are consistent with those given in [4] ($G = 3.06 \times 10^{-6} \text{ nm}^2$, $\lambda^* = 250 \text{ nm}$). In Figure 3b, Δn corresponds to about 0.219, which is literature value associated with 632.8 nm He-Ne lasers used to investigated LC material [6]. Birefringence as a function of voltage is explored for undoped and dye-doped samples under dark and illuminated conditions. Figure 4 shows this dependency in such a way that Δn value has not been changed by laser pumping in the undoped sample (see Figure 4a), whereas there exist $\sim 4\%$ gain in birefringence with laser illumination in the vicinity of threshold voltage for the dye-doped sample (see Figure 4b). Above the threshold, the tendency is the same for all cases, as it is expected by the strength of voltage aided reorientation order. Overall expanded uncertainty is 0.5% in birefringence measurements and it is calculated according to GUM [10].

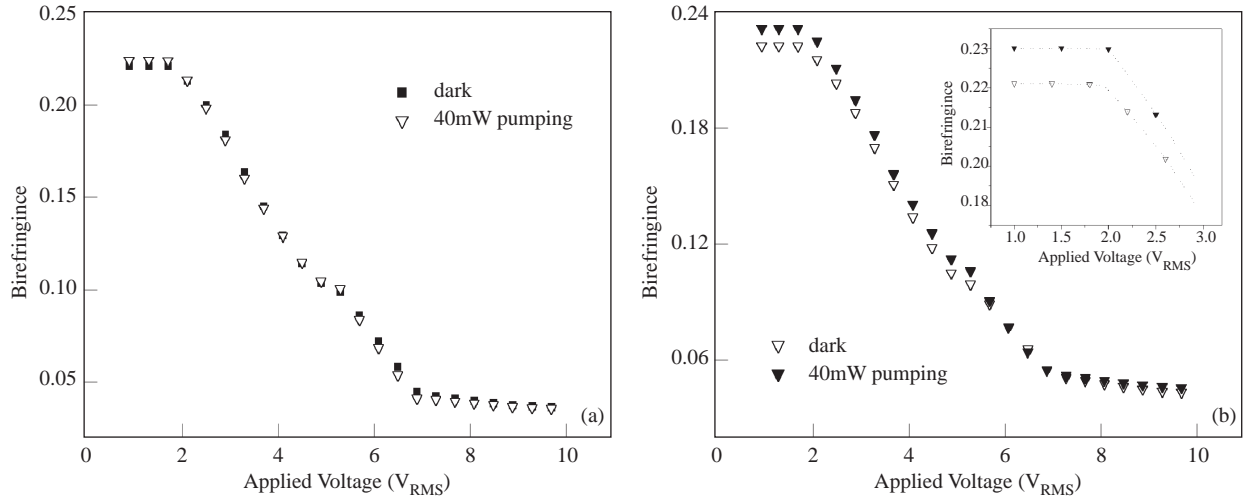


Figure 4. Voltage dependency of birefringence; for $d = 5 \mu\text{m}$ cells, with $f = 1 \text{ kHz}$ sine waves, $\lambda = 632.8 \text{ nm}$ and $T = 25^\circ \text{C}$ (a) for undoped sample, (b) for dye-doped sample.

Reorientation mechanism, including Anthraquinone derivative dyes, is well explained by the Jánossy effect [11, 12]. In the explanation of dye-enhanced reorientation mechanism, the starting point is the interaction between liquid crystal molecules, and it is assumed that the mean fields acting on the dye molecules are different when the dye molecules are in their ground state and when they are in their excited state. This assumption brings about a modified version of optical torque that is expressed as

$$\vec{\Gamma}_{\text{DYE}} = \eta \vec{\Gamma}_{\text{OPT}}, \quad (5)$$

where $\vec{\Gamma}_{\text{OPT}}$ is optical torque, $\vec{\Gamma}_{\text{DYE}}$ is the dye torque and η is the characteristic parameter of the dye that is only dependent of the structure of the dye, so one can write the $\vec{\Gamma}_{\text{DYE}}$ as

$$\vec{\Gamma}_{\text{DYE}} = (\xi/8\pi)(\hat{n} \cdot \vec{E})(\hat{n} \times \vec{E}). \quad (6)$$

Here, $\xi = \eta\Delta\varepsilon$, where $\Delta\varepsilon$ is the dielectric anisotropy; \hat{n} is the unit vector along the director axis and \vec{E} is the electric field. If the dye molecules are in their excited state, this means that angular momentum of dye molecules are increased by pumping laser. That is why host molecules behave in such a way that they minimize their angular momentum so that total angular momentum should be conserved, and this could be realized by reorientation of the LC molecules. The interaction of light with dye excites the dye molecules and this energy is conducted to nematic hosts via the angular momentum conservation, whereby reorientation of liquid crystal molecules takes place in the frame of Eq. (6) [11, 12]. What we have found out is the enhancement of birefringence under laser pumping, supporting the postulates of the Jánossy effect. It is our view, that absorbance-based dye torque is responsible for birefringence modulation and this modulation causes molecular reorientation in dye-doped case. In the scope of this study, a quantitative analysis of optical anisotropy was performed, demonstrating the absorptive effect of dye, the results are encouraging for further exploration into the origin of reorientation mechanisms in dye-doped applications.

Acknowledgement

This work was supported by the Scientific Research Projects grant (02-B-01-02-08) of Gebze Institute of Technology.

References

- [1] I.C. Khoo, M. Shih, M.V. Wood, B.D. Guenther, P.H. Chen, F. Simoni, S.S. Slussarenko, O. Francescangeli and L. Lucchetti, *Proceedings of the IEEE*, **87**, (1999),1897.
- [2] S. Bartkiewicz, A. Januszko, A. Miniewicz and J. Parka, *Pure Appl. Opt.*, **5**, (1996), 799.
- [3] S.E. San, O. Köysal and F.N. Ecevit, *Opt. Commun.*, **212**, (2002), 405.
- [4] S.T. Wu, *Phys. Rev. A.*, **33**, (1986), 1270.
- [5] S.T. Wu, C.S. Hsu, and K.F. Shyu, *Appl. Phys. Lett.*, **74**, (1998), 344.
- [6] S.T. Wu, U. Efron, and L.D. Hess, *Appl. Opt.*, **23**, (1984), 3911.
- [7] O. Köysal, D. Önal, S. Özder, and F.N. Ecevit, *Opt. Commun.*, **205**, (2002), 1.
- [8] R. Chang, *Mat. Res. Bull.*, **7**, (1972), 267.
- [9] H. Mada, and S. Kobayashi, *Mol. Cryst. Liq. Cryst.*, **33**, (1976), 47.
- [10] Guide to the expression of uncertainty in measurement, (1993), 1st ed. ISO.
- [11] I. Jánossy, *Physical Review E*, **49**, (1994), 2957.
- [12] I. Jánossy, L. Csillag, and A.D. Lloyd, *Physical Review A*, **44**, (1991), 8410.