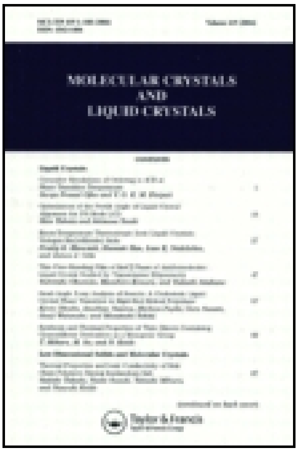

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Transient Laser Induced Orthogonal Director-Axis Reorientation in Dye-Doped Liquid Crystals (DDLC)

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TRANSIENT LASER INDUCED ORTHOGONAL DIRECTOR-AXIS REORIENTATION IN DYE-DOPED LIQUID CRYSTALS (DDLC)

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Abstract We report the results of dynamic grating experiment with a planar dye-doped liquid crystal. The dye used is D2 (from EM chemicals). It was found that for this particular DDLC system, the excited state dye molecules caused interesting transient reorientations of the liquid crystal director axis in directions orthogonal to the initial molecular alignment.

Introduction

The large molecular reorientational nonlinearities of nematic liquid crystals are known to have many applications in display, image processing and optical switching[1]. In recent years, it is found that dye-doped liquid crystal has some new and potentially useful features in light induced reorientational nonlinearities[2-6]. These new features include enhanced positive reorientational nonlinearity[2,5], different types of negative reorientational nonlinearities under different time regions (e.g., transient and steady-state)[3,4,6], formation of permanent gratings under relatively low laser input and erasability of the gratings. All these features are potentially applicable in optical storage and real time holography.

The basic principle of optical reorientational nonlinearities is the optical dielectric anisotropy $\Delta\varepsilon$. For undoped nematic liquid crystals, because $\Delta\varepsilon$ is positive, the liquid crystal molecules tend to align themselves in the direction of the applied optical electric field. This is true for both planar and homeotropic samples. However, when doped with absorbing dye molecules, the orientational nonlinearity takes on different features. In particular, Janossy *et al.* have observed that some classes of anthraquinone dye molecules will further enhance the positive reorientational nonlinearity when these dye molecules, together with the liquid crystal host molecules, are photoexcited[2,5]. While Gibbons *et al.*[3] and Chen and Brady[4] have observed that, the molecules of DDLC will align themselves in a direction orthogonal to the optical electric field and the propagation wave vector under prolonged exposure (i.e., a permanent orientational state has been reached) to the optical field, which is associated to the negative nonlinearity. In a recent paper by Khoo *et al.*[6], the observations of new extraordinarily large negative reorientation effect in the transient regime have been reported. In reference[6], the experimental results with two different types of dye molecules were presented. One is a dichroic dye D16 (from EM chemical), which is one of the several dye materials examined by Janossy and Kosa[2,5], the other is the diazodamin dye, used by Gibbons *et al* [3]. Khoo *et al.* has established that in the transient regime, i.e., under relatively short laser pulse illumination compared to the speed of the material response to the nonlinear process, the negative nonlinearity is associated with the liquid crystal director axis realigning toward the direction of the wave vector of the propagation field, which is different from the negative nonlinearity of the same DDLC material under prolonged exposure to optical field as reported in reference[2]. In addition, an effective optical nonlinearity in association with the measured diffraction efficiencies as large as 10^{-2} cm^2/watt has been observed[6].

In this paper, we report the preliminary observation of the negative reorientational effect using the dichroic dye material D2 (from EM chemicals) doped nematic liquid crystal 5CB (from EM chemical). Our studies show that in such system, the direction of the transient reorientations of the director axis appear to have both components orthogonal to the direction of the initial molecular alignment, i.e., for an initial director axis alignment in the y-direction, the reorientations possess both x- and z-components.

Experiments and Observations

Figure 1 is a schematic diagram of the experimental set-up. The DDLC sample is planar aligned in the y-direction by pretreating two glass slides with PVA, then sandwiching the liquid crystal between the two glass slides. The DDLC is prepared by doping the nematic liquid crystal (5CB) with dichroic dye molecules (D2, chemical structure shown in figure 2). The thickness of the sample is 50μm. A linearly polarized cw Ar$^+$ laser beam (λ=0.5145μm) is electronically shuttered to create square pulses, it is split into two coherent beams and combined at an angle of 1° on the DDLC sample. The probe beam is a linearly polarized CW He-Ne laser beam. The polarizations of the pump and probe beams are oriented in different combinations to study the polarization dependence of the laser induced index change (figure 3) as well as the direction of the molecular reorientation. The temporal behaviors of the first order diffraction of the He-Ne beam are recorded.

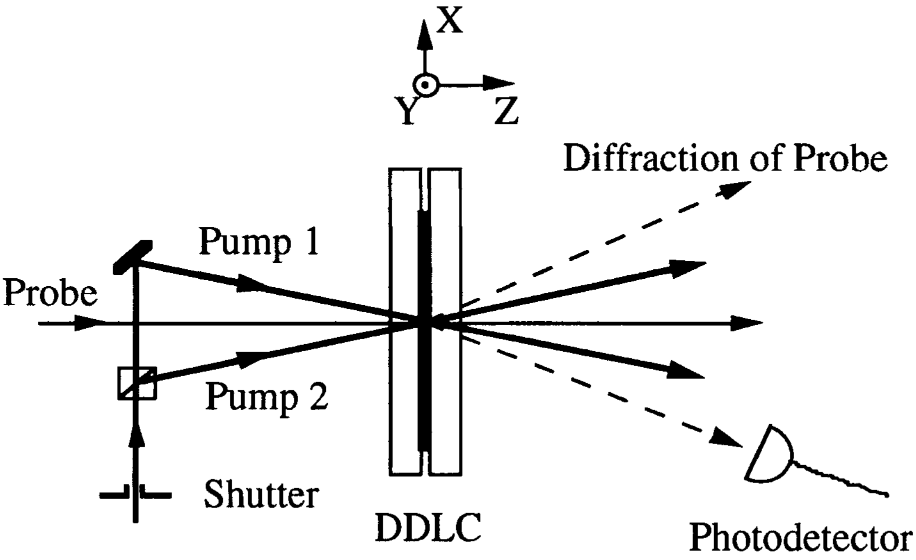

Figure 1. Schematic diagram of the experimental setup for dynamic grating diffraction study.

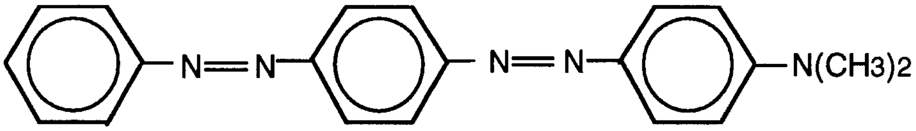

Figure 2. Chemical structure of the dichroic dye D2.

Figures 4a and 4b show the temporal behaviors of the He-Ne diffraction from the index gratings generated by the Ar^+ pump beams in the DDLC sample. In figure 4(a), the polarizations of the pump beams are parallel to the initial direction of the molecular axis of the DDLC sample, whereas in figure 4(b), the pump beams are polarized orthogonal to that of the molecular axis. For both figures 4(a) and 4(b), the upper traces are the diffraction behaviors of the probe beams when the probe polarization is parallel to the direction of initial molecular alignment, the lower traces are the diffraction behaviors when the probe polarization is orthogonal to the direction of initial molecular alignment. So figures 4a and 4b include all the different polarization configurations of the pump and probe beams shown in figure 3.

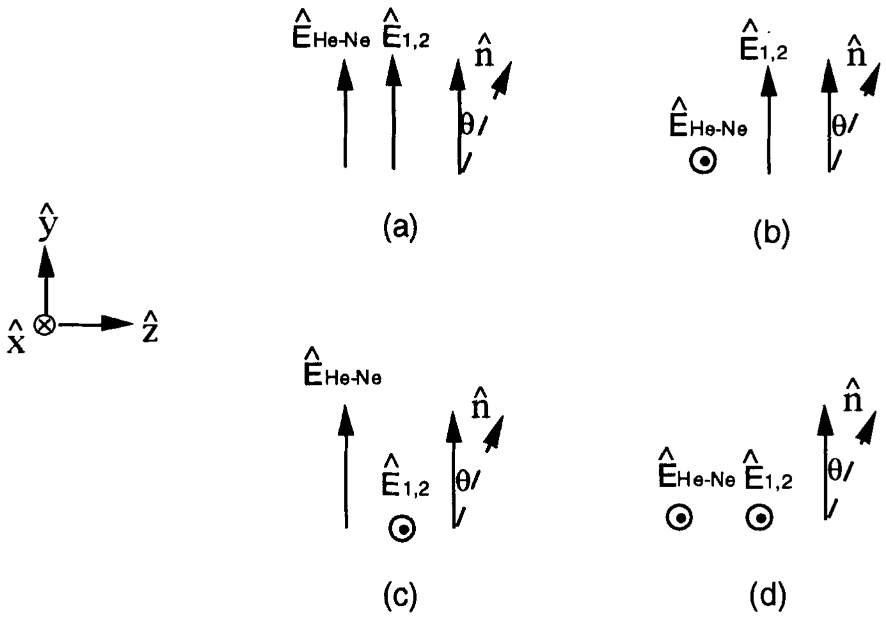

Figure 3. Polarization configurations of the pump and probe beams for studying the polarization dependence of the laser induced index change.

If the molecules realign themselves in the direction of the wave vector of the propagation field, i.e., the z-direction in figure 3, the grating formed by the two Ar^+ pump beams can be detected only when the polarization of the probe beam is parallel to the initial direction of the molecular axis of the DDLC sample, i.e., only the e-probe can detect the grating(figure 3(a) and (c)). On the other hand, if the molecular realignment has an x-component which is orthogonal to both the initial alignment (y-direction) and the direction of the wave vector (z-direction), then the grating can be detected when the polarization of the probe beam is either parallel (figure 3 (a) and (c)) or orthogonal (figure 3(b) and (d)) to the initial direction of the molecular axis of the DDLC sample, i.e., both e- and o-probes can detect the grating. Our experimental results (from figure 4) show that indeed the molecular realignment has an x-component, since both e- and o-probes produce diffractions.

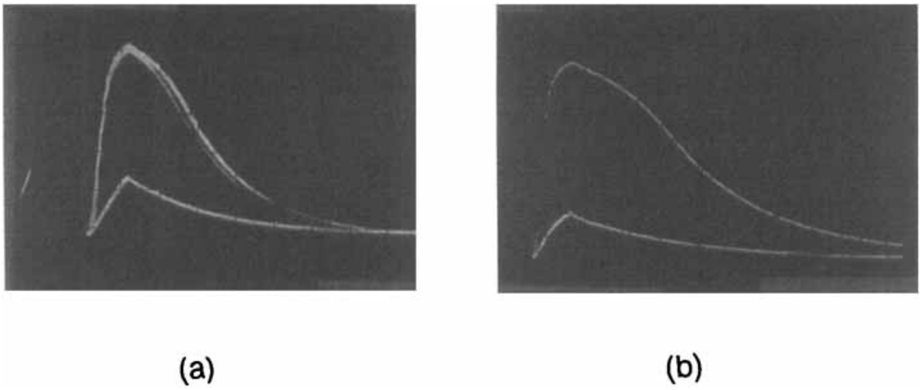
(a) (b)

Figure 4. Oscilloscope traces of the He-Ne diffraction from the index grating on the DDLC sample generated by the Ar+ pump beams. Horizontal scale: 0.1 second/division.
(a) Upper trace: pump and probe polarization configuration as shown in figure 3(a); Lower trace: pump and probe polarization configuration as shown in figure 3(b). (b) Upper trace: pump and probe polarization configuration as shown in figure 3(d); Lower trace: pump and probe polarization configuration as shown in figure 3(c).

There is a difference in the diffraction efficiency between e- and o-probes as shown in figure 4. This can be explained by the fact that the refractive index changes of e- and o-component are different for the same reorientation angle. If the probe is an e-wave, then the refractive index change "seen" by the probe is given by,

$$\Delta n_e = \frac{n_o n_e}{\sqrt{n_e^2 \sin^2\theta + n_o^2 \cos^2\theta}} - n_e \qquad (1)$$

where θ is the reorientation angle. If the probe is o-polarized, the refractive index change is,

$$\Delta n_o = \frac{n_o n_e}{\sqrt{n_e^2 \cos^2\theta + n_o^2 \sin^2\theta}} - n_o \tag{2}$$

When θ is very small, we have the following approximations after simple algebra,

$$\Delta n_e \approx -\frac{n_e^2 - n_o^2}{2n_o^2} \cdot n_e \cdot \theta^2 \tag{3}$$

and

$$\Delta n_o \approx \frac{n_e^2 - n_o^2}{2n_e^2} \cdot n_o \cdot \theta^2 \tag{4}$$

Therefore, the ratio of the ordinary and extraordinary refractive index changes is given by, [e.g., for 5CB $n_e = 1.72$, $n_o = 1.52$]:

$$\frac{\Delta n_e}{\Delta n_o} \approx \frac{n_e^3}{n_o^3} \approx 1.5 \tag{5}$$

For thin grating, the diffraction efficiency is related to the refractive index change by[1],

$$\eta \sim \left(\frac{\pi d}{\lambda} \Delta n\right)^2 \tag{6}$$

So the diffraction efficiency related to the reorientational nonlinearity probed by an e-ray should be more than twice of that probed by an o-ray. This is in good agreement to our experimental observations.

The decay dynamics shown in figure 4 are clearly associated with the orientational grating decay. The orientation grating relaxation time of the liquid crystal can be estimated using the following equation:

$$\tau_r = \frac{\gamma}{Kq^2} \qquad (7)$$

where the viscosity coefficient $\gamma \approx 0.1$ poise, the elastic constant $K=4\times10^{-7}$ dynes[6], For a crossing angle of 1 degree, the grating constant $\Lambda=50\mu m$, we estimated that the orientational relaxation time is thus about 1 second. This value is close to the experimentally observed grating decay time(From figure 4, we can see that the decay time is about 0.6 seconds). On the other hand, the thermal grating decay time

$$\tau_{th} = \frac{1}{Dq^2} \qquad (8)$$

is much shorter. For $D_\perp$ and $D_{//}$ in the order of 10^{-3} cm^2 /s, and $q=2\pi/(50\mu m)^2$, τ_{th} is on the order of 6 ms, which is 2 orders of magnitude faster than those depicted in figure 4. We conclude therefore that the observed results are due to excited-dye-molecule assisted liquid crystal reorientations.

Conclusions

Laser induced negative director-axis reorientations of dye-doped liquid crystals have been studied with a dynamic grating technique. The polarization dependence study confirms that the excited-dye-molecule assisted reorientations could occur in both directions orthogonal to the initial director-axis alignment. Further studies, including systematic investigations of laser induced nonlinearities of DDLC under different time scales, dependence of these nonlinearities on dye concentrations and molecular structures of different dye materials, and a quantitative analysis to explain the polarization and energy dependence observed in all the experiments mentioned above, are clearly in order, and are currently in progress.

Acknowledgment:.

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