Orienting Effect of Light on Dye-doped Liquid-Crystal Polymer


Abstract—Light-induced reorientation of the dye-doped liquid-crystal polymer director is found and studied. The director rotates from the light field direction, which corresponds to the negative optical nonlinearity. The nonlinearity coefficient is several times larger than that of a low-molecular liquid crystal doped with the same dye.

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Introduction. For nematic liquid crystals (NLCs), the effects of light-induced director rotation are well known [1]. In transparent NLC, rotation results from the effect of the light electric field on induced dipoles. In absorbing NLCs (nematic hosts doped with dye), the orientational effect of light is associated with a change in the interaction of excited dye molecules whose distribution is determined by the light field direction and is asymmetric with respect to the NLC director with the nematic host [2–4].

To our knowledge, previous studies of the orientational interaction of light with NLCs were restricted to the case of low molecular mass nematic hosts. In polymeric nematic liquid crystals, the director reorientation was observed only upon exposure to low-frequency electric and magnetic fields [5–7]. In the present paper, we report about the first observation of the light-induced director rotation in a polymer nematic liquid crystal.

Experimental samples and experimental technique. The object of the study was comb-shaped polymer PAA (polyacrylate) with side mesogenic fragments

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\text{O}_2\text{N-}
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\text{N=N-}
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\text{(C}_8\text{H}_{17})_2
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(PAA)

Liquid-crystal polymer (LCP) has a nematic phase at temperatures below 123 °C; at 26 °C, the glass transition occurs. For the planar LCP orientation, a glass cell of thickness \(L = 50 \mu m\), whose substrates were coated with an orientant (polyimide) layer, was used. The cell was filled under capillary forces with a liquid-crystal mixture at a temperature above the clearing point and then cooled from 125 °C to room temperature with a rate of 0.1 °C per minute. The absorbances of extraordinary and ordinary waves,
measured using an MS-122 spectrophotometer at room temperature at the wavelength $\lambda = 473$ nm, were $\alpha_0 = 199$ cm$^{-1}$ and $\alpha_o = 34$ cm$^{-1}$.

The LCP orientational nonlinearity was studied using the aberrational self-action of a light beam [8]. The schematic of the experimental setup is shown in Fig. 1. The linearly polarized light beam from solid-state laser 1 (LCS-DTL-364, Laser Export) with wavelength $\lambda = 473$ nm passed through double Fresnel rhomb 2, was focused ($w_0 = 43$ $\mu$m) using lens 3 on sample 4 placed between heating plates 5. The temperature was measured using a thermocouple. The case of oblique light incidence ($\alpha = 40^\circ$) was studied. The polarization vector and unperturbed director were in the incidence plane (horizontal plane); in this case, the extraordinary wave was excited in LCP. Polarization of light passed through the sample was determined using analyzer 6. The aberration pattern shaped as a system of concentric rings was observed on screen 7 placed behind the sample. The number of rings $N$ is controlled by the nonlinear phase shift on the beam axis [9] and is related to the light-induced refractive index by the expression

$$|\Delta n| = \frac{N\lambda \cos \beta}{L},$$

where $\beta$ is the light refraction angle in LCP.

**Experimental results.** Measurements were performed in the temperature range of 90–110 °C. When the light beam of power $P \sim 0.5$ mW passed through the sample, a system of aberrational rings appeared on the screen; light polarization remained unchanged. At an addition of a probing ordinary wave, i.e. a small polarization plane rotation from the horizontal, and a vertical analyzer position, no aberration pattern was observed. This suggests that the refractive index of the ordinary wave was unchanged and excludes the thermal nonlinearity mechanism. Hence, the observed LCP nonlinearity is caused by director reorientation in the light beam incidence plane.

The aberration pattern transformation due to a crystal displacement with respect to the light beam allows the determination of the optical nonlinearity sign [8]. In the case at hand, an upward LCP displacement caused disappearance of the upper part of the aberration pattern, which indicates negative nonlinearity, i.e., a decrease in the refractive index of the extraordinary wave due to director rotation from the light field.

Figure 2 shows the time dependences of the light-induced refractive index, measured on the sample exposed to an extraordinarily polarized light beam, for various sample temperatures. We can see that the time of approaching the steady state significantly decreases with increasing temperature. This effect is associated with a decrease in the polymer viscosity.

At constant temperature $T = 110$ °C, stationary light-induced LCP refractive index increases with light beam power and the reorientation time decreases (Fig. 3).

The LCP director reorientation is reversible. As irradiation is terminated, the exponential relaxation $|\Delta n(t)| = |\Delta n(0)| \exp(-t/t_0)$ is observed. Its characteristic time at 110 °C is $t_0 = 14$ min (Fig. 4). The existence of relaxation suggests that the observed LCP director reorientation is a bulk process and is unrelated to the effect of photoalignment on cell substrates [10]. The relaxation time $t_0$ is comparable with the relaxation time after LCP director reorientation by a low-frequency electric field.

The above experimental results allow the conclusion that light-induced director reorientation caused by a change in intermolecular forces during selective excitation of dye molecules takes place in the