Coexistence of light-induced thermocapillary and orientational effects in thin nematic films with a free surface

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Abstract. The effect of nonlinear light action on a thin ($\sim$10 $\mu$m) films of the nematic liquid crystal deposited onto the absorbing substrate is experimentally investigated. The dynamics of the orientational and thermocapillary effects is directly studied. The two types of orientational processes were found out. The first one appears for several hundreds of milliseconds when the light beam irradiation is turned on or off. The second one develops much slowly and does not relax during the light beam irradiation.

1. Introduction
The light beam absorption in isotropic liquid causes the nonlinear response due to variation of its properties [1–3]. For instance, the light beam heating can change the surface profile forming a dimple, which works as a lens for transmitted light [4, 5]. This effect is mainly caused by the decrease in surface tension and the appearance of thermocapillary forces inducing the hydrodynamic flows [6–8].

Thermo-optical effects are much less investigated for the anisotropic liquids, i.e. for liquid crystals (LCs). In contrast to the ordinary liquids, LCs have partial orientational and, in some cases, translational ordering. On the macroscopic level, the average direction of the long molecular axes is defined by a unit vector, the LC director. The director field is very sensitive to various external stimuli, like electric, light and thermal fields. Thus, one can expect the interplay between the heat flux and the LC orientation when the light beam is absorbed in the medium. The most known example of thermal LC orientation is the Lehmann effect [9, 10], the director rotation in chiral nematic LCs, which, in particular, can be induced by light [11]. The light beam absorption can cause the hydrodynamic flows resulting in the director reorientation in hybrid and twist aligned nematic films [12–15]. It is interesting to note that the director deformation can be a consequence of the temperature gradient only [16, 17]. In this case, the temperature gradient field results in LC director deformation similarly to electric and magnetic fields.
In the previous study, we described the orientational effect induced by light beam heating of nematic LCs with free surfaces [18]. The nematic film was deposited onto the indium-tin-oxide (ITO) glass substrate which provides the homeotropic anchoring and particular light absorption. An air-LC interface restricts the heat outflow and the axially symmetric heat flux reorients the LC director forming an umbilical defect. As for the isotropic liquids, the dimple formation is accompanied by the LC director deformation or appears without it.

In this study, we directly observe both the light-induced surface distortion and LC director reorientation using optical microscopy methods. In particular, we focus on the moments just after turning on and off the light beam focused onto the thin nematic films with a free surface.

2. Experimental
The E7 nematic liquid crystal (Synthon Chemicals) was deposited onto the ITO-coated glass substrate. The LC droplet of the size of about 2 mm and variable thickness was held on the substrate by capillary forces. Because of the wettability of ITO-coated surface by the LC, the edge of the LC droplet can be considered as a wedge film (Fig. 1a) whose surface can be modified by a focused light beam (Fig. 1b). Both LC surfaces (LC-ITO and LC-air) provide homeotropic boundary conditions and, hence, specify the uniform orientational distribution.

![Figure 1](image-url)

**Figure 1.** Schematic representation of the LC sample before (a) and during light beam irradiation (b); velocities $V_1$ and $V_2$ show the mass transfer from the heating area. The blue dashes show the local director orientation. The scheme of experimental setup (c): S is the shutter, L is the lens, GP1 and GP2 are glass plates, P and A polarizer and analyzer, LED-1 and LED-2 are light emitting diodes.

The optical setup (Fig. 1c) consisted of solid-state continuous-wave laser (SSP-LN-532-FN-300-0.5-LED, CNI, China) with the wavelength of $\lambda = 532$ nm, home-made mechanical shutter with switching on and off times $\sim 1$ ms, focusing lens, the sample with visualization scheme, and semitransparent screen. The radius of laser beam $w_0 = 26$ $\mu$m (at the intensity level of $e^{-2}$) was measured by the “knife-edge” method [19]. The sample was placed vertically in the beam waist.
The angle of incidence of the p-polarized light beam was $45^\circ$. The diffracted light beam was observed on the semitransparent screen placed behind the sample. The image of the illuminated area was observed with the help of a high-speed camera equipped with the 16X optical objective with a large working distance. For background illumination, two light emitting diodes (LEDs) with luminescence at $\lambda_{\text{LED}} \sim 620$ nm were used separately or simultaneously. The first LED source allows one to visualize the orientational deformation when the sample is placed between crossed polarizers (polarizer P and analyzer A). The light from the second LED was partially reflected by the glass plate (GP2) and then gave the reflection image. The opened and closed shutter positions and camera recording were triggered by microcontroller. The moments of the light beam switching were controlled using a photodiode (PD) by the registration of light reflected by the glass plate (GP1) placed after the shutter. The experiments were carried out at room temperature.

3. Results and Discussion

The interference of light reflected by the LC-substrate and LC-air interfaces results in the formation of bright and dark lines (Fig. 2a-2d). Note that these interference patterns are similar to those observed for isotropic liquids [4, 5]. The LC thickness varies between bright and dark lines by the value of $\delta h = \lambda_{\text{LED}}/4n_o$, where $n_o = 1.52$ is the refractive index of an ordinary light wave. Thus, the surface profile can be reconstructed by the positions of brightness extrema along the $x$ axis (Fig. 2e) with an accuracy of $\delta h \sim 0.1 \mu$m. Typical LC film profiles before and during the laser irradiation are shown in Fig. 2f. The variation of LC film thickness $|\delta h|$ with time (Fig. 2g) is measured by alternating brightness in the area of the light beam axis. The obtained data clearly show that the light beam irradiation results in the gradual variation of surface profile. The initial region in temporal dependence can be approximated by the exponential curve $|\delta h| = |\delta h_{\text{max}}|(1 - \exp(-t/\tau))$ with the characteristic time $\tau \sim 3$ ms, where $|\delta h_{\text{max}}|$ is the maximum thickness change. Thus, despite of the relatively slow process of thermocapillary effect [18], the initial development is at least one order of magnitude faster.

At the LC thickness of about $h \sim 10$ $\mu$m and light beam power $P = 80$ mW, the light-induced director deformation is visualized by the bright areas in crossed polarizers (Fig. 3). Initially, only small bright regions due to the surface inhomogeneity are visible (Fig. 3a). During the light beam exposure, the bright cross appears, then becomes more intense and finally vanishes (Fig. 3b-3f). This bright cross corresponds to the axially symmetric orientational deformation (Fig. 1b). Surprisingly, the bright cross is temporary observed after turning off the light beam (Fig. 3g-3l). The described effect is non-local; the size of the director deformation is several times larger than the diameter of the light beam. The appearance of the bright cross in the visualization scheme is accompanied by broadening the light beam passed through the sample.

A more complex process occurs at higher light beam power (Fig. 4). The large bright cross develops and starts to relax during several tens of milliseconds after turning on the light beam (Figs. 4a-4e). Then the smaller bright cross manifests itself (Figs. 4f). It becomes more intense for several seconds (Figs. 4f-4h) and does not change during the further light irradiation. The size of this cross pattern, which corresponds to the axially symmetric director deformation, is comparable to the beam diameter $2w_0$. After turning the light beam off, this deformation relaxes and, as in the previous case, the large bright cross pattern is temporary formed (Figs. 4i-4n).

Note that the described effects do not depend on the light beam polarization and only slightly depend on the angle of incidence. The orientational deformation is symmetrical with respect to the axis which is normal to the plane of substrate in spite of the oblique incidence of the light beam.

Thus, we have two types of thermo-optical orientation in the LC film. The first one is non-steady-state and depends on the rate of the film thickness variation (Fig. 5). The increase in the light beam power leads to the more rapid variation of the film thickness as well as the
Figure 2. The images of the LC film edge (in reflected light) at different times of light beam exposure (a-d). The light beam power is $P = 120$ mW. The image brightness distribution $I(x)$ along the $x$ axis at $t = 0$ ms (e). The reconstructed surface profiles along the $x$ axis (the dashed line corresponds to the position of light beam axis). The time dependence of relative thickness variation $|\delta h|$ at light beam axis (g).

The second type requires sufficient light beam intensities, develops much slowly and remains...
Figure 4. Polarizing microscopy images of the irradiated region (formed by the transmitted light) at different moments during and after the light beam exposure. The light beam power is $P = 120 \text{ mW}$, the exposure time is $t_{\text{exp}} = 4000 \text{ ms}$, the LC film thickness on the light beam axis is $h = 9.1 \mu\text{m}$.

Figure 5. The time dependences of thickness variation $|\delta h|$ during and after the light beam irradiation. The light beam power is $P = 80$ (1) and $120 \text{ mW}$ (2), the exposure time is $t_{\text{exp}} = 4000 \text{ ms}$. The inset illustrates the initial stage of thickness variation.

the same (Fig.4h) when the LC film thickness variation reaches the plateau (see curve 2 in Fig.5). This can be caused either by the temperature gradient [16, 17] or by the circular flow convection observed in LCs and polymers [22, 23].

Both types of director deformation require further theoretical study as well as computer simulations to determine the physical origin of the orientational processes more precisely. The situation can be more complex because of the interference of a number of effects, which cause the director reorientation [10].

Conclusion

To summarize, the thermo-optical orientational processes are studied experimentally for thin nematic films with free surfaces. The light absorption by the ITO layer causes both thermocapillary and orientational effects. The non-steady-state orientational deformation is observed during several hundreds of milliseconds after turning on or off the light beam. The rapid variation of the film thickness in the irradiated area indicates the development of hydrodynamic flows near the LC surface which can be responsible for this deformation. At sufficient light beam intensities, the stable LC orientation is formed. The latter one can be caused by both hydrodynamic flows and thermo-orientational effect. In each case the orientation is axially
symmetric with respect to the axis oriented normally to the substrate surface.

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**References**

[1] Costa G D and Calatroni J 1978 *Applied Optics* **17** 2381–2385
[2] Da Costa G 1986 *Optical Engineering* **25** 1058–1063
[3] Da Costa G and Escalona R 1990 *Applied Optics* **29** 1023–1033
[4] Duarte-Quiroga R A and Calixto S 2000 *Appl. Opt.* **39** 3948–3954
[5] Bezuglyi B A and Fedorets A A 2001 *Technical Physics Letters* **27** 359–361
[6] Bayazitoglu Y and Lam T T 1987 *Journal of Heat Transfer* **109** 717–721
[7] Longtin J P, Jikata K H and Ogawa K 1999 *International Journal of Heat and Mass Transfer* **42** 85–93
[8] Zykov A Y and Ivanova N A 2017 *Applied Physics B* **123** 235
[9] Oswald P, Dequidt A and Poy G 2019 *Liquid Crystals Reviews* **7** 142–166
[10] Oswald P, Dequidt A and Poy G 2021 *Liquid Crystals* (Wiley) pp 117–191
[11] Bono S, Sato S and Tabe Y 2017 *Soft Matter* **13** 6569–6575
[12] Akopyan R S, Alaverdyan R B, Santrosian E A and Chilingarian Y S 2001 *Journal of Applied Physics* **90** 3371–3376
[13] Hakobyan R S, Alaverdyan R B, Gevorgyan G S and Kirakosyan A A 2010 *JETP Letters* **92** 457–459
[14] Aleksanyan A, Gevorgyan G, Hakobyan R and Alaverdyan R 2012 *International Journal of Modern Physics: Conference Series* **15** 120–128
[15] Hakobyan M R, Alaverdyan R B and Hakobyan R S 2014 *Molecular Crystals and Liquid Crystals* **596** 152–162
[16] Demenev E I, Pozdynakov G A and Trashkeev S I 2009 *Technical Physics Letters* **35** 674–677
[17] Trashkeev S I and Britvin A V 2011 *Technical Physics* **56** 747–753
[18] Shvetsov S A, Zolotko A S, Voronin G A, Emelyanenko A V, Avdeev M M, Bugakov M A, Statsenko P A and Trashkeev S I 2021 *Optical Materials Express* **11** 1705
[19] Khosrofian J M and Garetz B A 1983 *Applied Optics* **22** 3406
[20] Belyaev V V 2001 *Physics-Uspekhi* **44** 255–284
[21] Pan R P, Hsiung H and Shen Y R 1987 *Phys. Rev. A* **36** 5505–5508
[22] Dradrach K, Bartkiewicz M and Miniewicz A 2016 *Physical Chemistry Chemical Physics* **18** 3832–3837
[23] Choi H and Takezoe H 2016 *Soft Matter* **12** 481–485