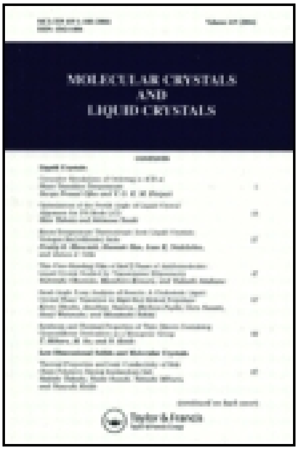

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SURFACE-MEDIATED BEAM COUPLING IN NOMINALLY PURE NEMATIC LIQUID CRYSTAL

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We have carried out experiments that indicate a surface-charge-mediated photorefractive effect in nominally pure liquid crystal homeotropically aligned on ITO-electrodes. We suggest that interfering beams modulate distribution of the charges in a layer of LC molecules adsorbed by the ITO-electrodes surfaces. It results in a tangential component of dc-field applied to the cell which re-arranges liquid crystal molecules in the adsorbed layer. This re-arrangement causes an easy axis modulation and the grating recording. A bulk-mediated mechanism related to an instability induced by the dc-field may give a contribution to the grating formation in specific range of the recording parameters.

Keywords: nematic liquid crystal; photorefraction; photo-charges

1. INTRODUCTION

Photorefraction in liquid crystals has recently attracted significant attention since it is characterized with strong optical nonlinearities at low light intensities and voltage [1–16]. Currently, liquid crystalline materials are considered a leading candidate for those applications that do not require long storage times. It is believed that the photorefractive effect in a LC cell could be initiated by light induced charge generation both in the cell

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volume and in the LC orienting layer. In case of volume-mediated photorefraction, an incident optical intensity grating generates photo-charges in LC bulk, which migrate and diffuse within the LC and set up various *dc* space charge fields [1–9]. These space charge fields create a torque on the liquid crystal director, forcing director reorientation and hence large refractive index changes. In the case of surface-mediated photorefraction, spatially-modulated light field results in a modulation of the surface charges on aligning surface that in turn causes the anchoring modulation and surface angular torque in the LC bulk. Tabiryan *et al*. proposed to use a photorefractive aligning surface with this aim [10]. Singer, Reshetnyak, Reznikov *et al*. found that *surface-mediated* effect could also arise if photo-induced charges in the aligning layers changed the distribution of ions in the LC bulk [11] or due to the photochemical reaction on the aligning surface [12]. It was shown recently that surface mediated photorefractivity can be also initiated due to a light-induced adsorption of fullerene-based dopant [13]. S. Bartkiewicz *et al*. found effective photorefractive effect due to redistribution of the electric field between aligning layer and LC at light-induced changes of photoconductivity of the aligning layer [14].

The distinctive feature of photorefractivity of LCs is that it can be obtained in nominally transparent, undoped LCs. Slussarenko reported on a possibility to get photorefraction gratings without light-induced charge generation [15]. He proposed that initial light field-induced reorientation of a LC in a cell with dc-field applied could result in space-charge profile due to a bulk Helfrich-Carr effect. Pagliusi and Cipparone observed surface-charged induced photorefraction in planarly aligned pure LC [16]. Similar to J. Zhang *et al*. [12], Boichuk *et al*. [11] proposed that *dc*-field accumulated charges near the polymer aligning surface and light field locally modified the surface charge density resulting in the director reorientation in the bulk. The origin of the light-induced modification of the not-controlled surface charges on nominally not-photosensitive aligning layers has not been set yet.

We report here on the first study of photorefractive effect in cells with nominally pure LC, which is homeotropically aligned on certainly not photosensitive surfaces of indium tin oxide (ITO). Our experiments indicate that the observed photorefractive effect is caused by spatial modulation of charges in 5CB layer adsorbed by the ITO surfaces.

2. EXPERIMENTS AND DISCUSSIONS

We studied the formation and characteristics in 20-μm cells of nematic LC pentil-ciano-bephenyl (5CB). The glass substrates were coated with ITO without any aligning layers above. The cells were filled at elevated temperature (~60°C) by capillary forces and slowly cooled down to room temperature. By this means a good homeotropic orientation achieved, which kept its quality during several weeks. After this, areas of inhomogeneous tilted alignment appeared. All experiments were carried out during a week after the cell preparation.

The experimental set-up was a standard two-wave arrangement (Fig. 1). Two parallel beams $I_{1,2}$ from continuously operated HeCd-laser ($\lambda = 0.44\,\mu\text{m}$) of equal intensity, $I \approx 100\,\text{mW}\,\text{cm}^{-2}$ passed through the wide-aperture lens and crossed in its focus plane at the angle θ, producing the interference pattern with period

$$\Lambda = \frac{\lambda}{2\sin(\theta/2)}.$$

The sample was set perpendicular to the plane of the beams intersection and could be rotated in the plane of the beams incidence (Fig. 1). The polarization of the beams was either *e*- or *o*-polarized. The period of the interference pattern was controlled in a range (5-100) μm by the shift of the prism, *P*. The irradiation of the sample resulted in the recording of the dynamic diffraction grating. Raman-Nath diffraction was observed in all cases. Four photodiodes *PhD*, detected the intensities of the passed beams (zero diffraction orders), $I_{1,2,(0)}$ and of the intensities of the (-1)-order diffraction beams, $I_{1,2,(-1)}$. Besides, the grating was tested by a weak beam from HeNe–laser ($I \approx 1\ \text{mW}\,\text{cm}^{-2}$). The probe beam did not influence the effect under investigation. *Dc*-field in the range $\pm 10\,\text{V}$ was

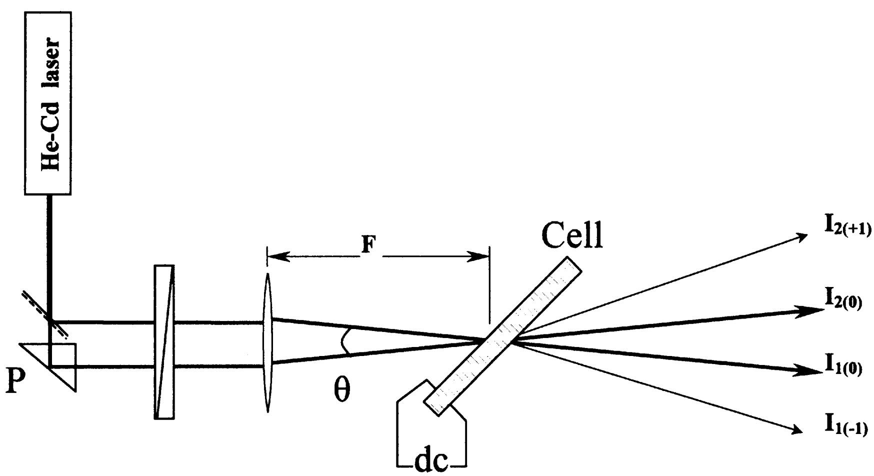

FIGURE 1 Basic scheme of the experiments.

applied to the cells. Several experiments were carried out, as follows (we followed the investigation scheme used in [12]):

- *The grating was written in a cell to which a dc-field was never applied.*The writing beams were turned on with no applied electric field (Fig. 2). A weak not-stationary self-diffraction and diffraction of the test beam (efficiency, $\eta \sim 10^{-3}$) was observed at the 45° angle of incidence. The first-order self-diffraction and test-diffraction disappeared after approximately 1 min exposure. A beam coupling between recording beams was also observed in this case. We could not figure out what factor determines the direction of the energy transfer in these experiments.
- *A (1-9)-V dc-field was applied for ten seconds, and then the circuit was opened.* The writing beams were then turned on. Self- and test diffraction and effective beam coupling were observed at the 45° angle of incidence. The direction of the energy transfer was determined by the sign of the *dc*-field (Fig. 3) and by the position of the beams tilt with respect to the cell normal. When the projection of the *dc*-field on the wave-vectors of the beams was positive, the beam which is closer to the normal to the cell is enhanced (see insertion in the Fig. 3). The measured gain coefficient achieved $\Gamma \approx 10^4\,\text{cm}^{-1}$, was typical for

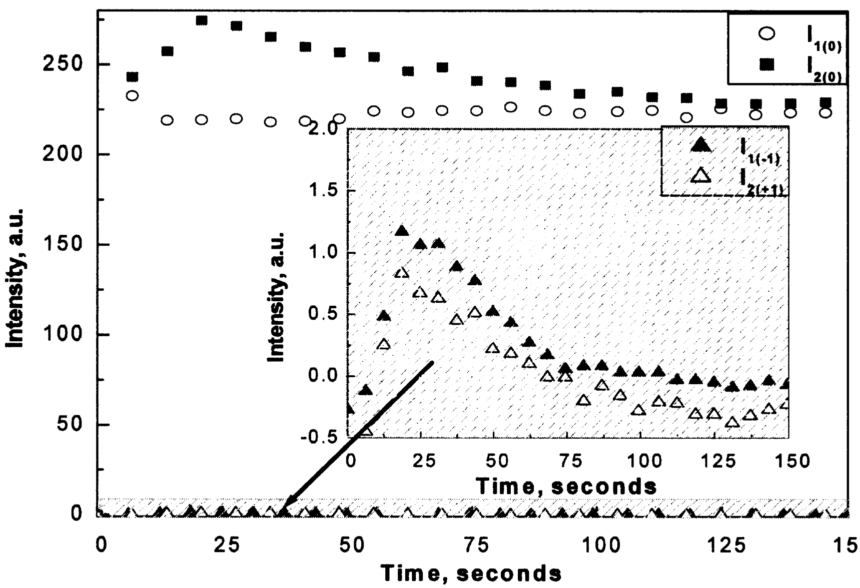

FIGURE 2 Grating recording with no dc-field applied. The enlarged plot of the dependence of the intensities of (±1) not-Bragg diffraction beams is in the insertion.

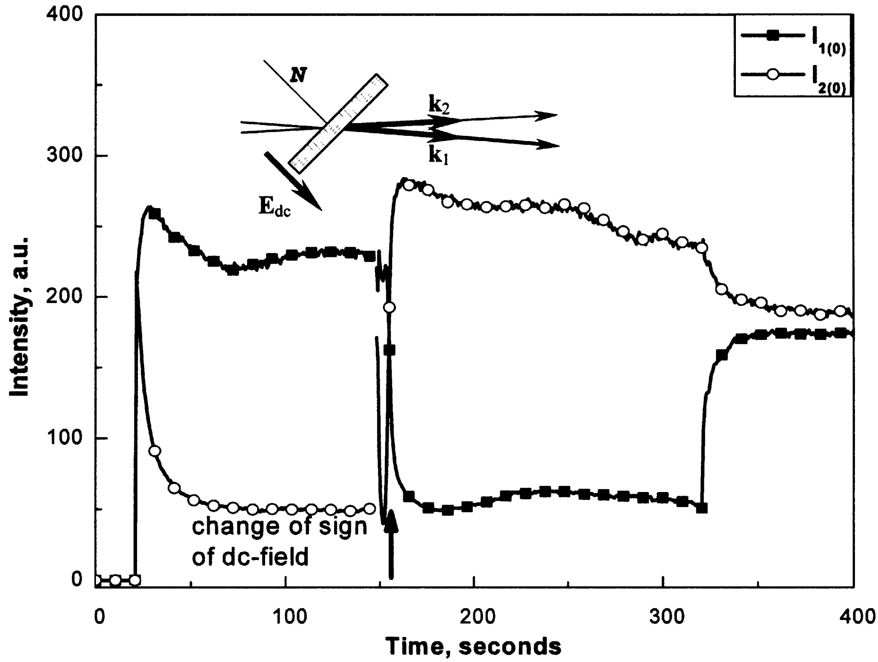

FIGURE 3 Beam coupling between the recording beams. The arrow points the change of the sign of dc-field.

photorefraction in LC systems [7,12], although we note that in the Raman-Nath regime such characteristic would not be descriptive as a figure of merit for applications. We did not find a self-diffraction but just the test diffraction at the normal incidence to the cell.

- *After relaxation of the grating recorded at the dc-field applied, the switching on of the dc-field within several hours (up to days)* resulted in not-stationary self- and test diffraction, that is a long-living "hidden grating" exists after visible disappearance of the self- and test-diffraction. It should be noted that a hidden grating could be the cause of confusion if to record a grating in the area where the hidden grating already existed.
- *The writing beams (no dc-field applied) were turned on till the not-stationary grating disappeared and the dc-field* ($U \sim 2V$) *was then turned on.* Not-stationary test diffraction on a "hidden grating" was observed. The diffraction disappeared in tenth of seconds.

These results point out that surface-mediated photorefractive effects play a significant role in the mechanism of the hologram recording. They are similar to the obtained in [12] but the authors of that paper did not observed a grating without application of a *dc*-field. To figure out if a surface mediated effects is dominant and to try to lift up a curtain above the mechanism of photorefraction in the studied system, the following experiments were then performed.

- We studied the dependence of the grating efficiency on the applied *dc*-voltage. The efficiency of the grating increased with the increase of the voltage achieved maximum in the range (2–4) V and slowly decreased after. It is particularly remarkable that this range corresponds to the voltage of LC instability, which was also observed in the cell. This instability manifested as irregular stripes with period 20–35 μm, visible just through a polarizer at tilted incidence that pointed out at director reorientation but the hydrodynamic effect.
- We measured the dependence of the diffraction efficiency, η, on the grating period, Λ, (Fig. 4). It was not-monotonic and differed from published before [2,9,15] where dome-shape dependences were reported. We found that generally the diffraction efficiency increased with the period of the grating but a slightly expressed pick in the range 20–35 μm existed on the base line of the dependence $\eta(\Lambda)$. This pick was obtained in the measurements of all of eleven studied cells and cannot be caused by an experimental error.

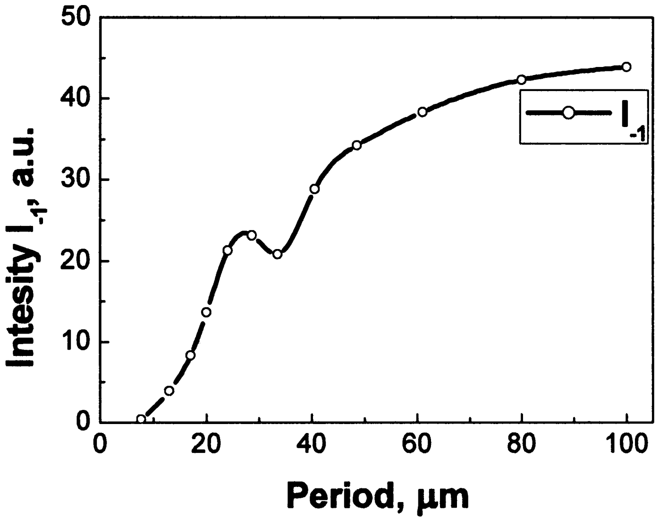

FIGURE 4 The dependence of the intensity of (+1) non-Bragg diffraction beam on the period of the interference pattern.

Last two observations pointed out at a correlation between the mechanism of the grating recording and LC instability. Slussarenko suggested that instability of Helfrich-Carr type could induce the spatially-distributed torque. According to [15], this torque appears due to the initial light-induced director reorientation, which results in a profile of charges in LC bulk. The charge profile leads to larger director reorientation and finally efficient photorefractive grating appears. Observation of the optimal diffraction in the region of the parameters Λ and U, where the instability exists suggests that this kind of effect may play a role in the grating recording. Although this role could not be a dominant one, since the diffraction was observed in the whole range of the values $\Lambda \subset (10–70)\,\mu m$ – not only at the periods characteristic to the instability $(20–30)\,\mu m$. Moreover, additional experiments showed that effective diffraction existed at very large periods, $\Lambda \approx 200\,\mu m$. The grating was also observed at the voltage $U < 1V$, what is much less then needed for instability.

The dynamic characteristics of the grating also point out a surface and not a bulk origin of the gratings. The dynamics of the grating recording and relaxation is depicted in Figures 5–6.

The typical plot of the grating recording is shown on Figure 5. At the simultaneous switching on of the *dc*-field and writing beams the self- and test-diffraction grow up during several minutes and finally achieve a stationary value. This value depended on the *dc*-voltage magnitude and on the period of the grating and attained up to 23%. Prior “training” of

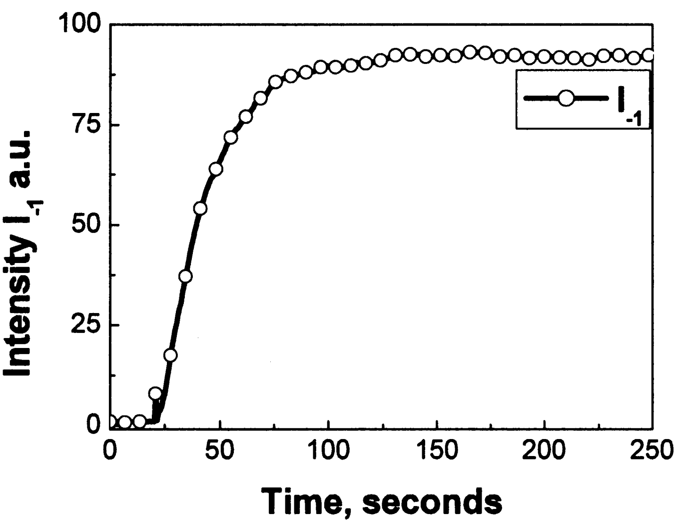

FIGURE 5 The dependence of the intensity of (+1) non-Bragg testing diffraction beam on exposure. U = 2.5 V.

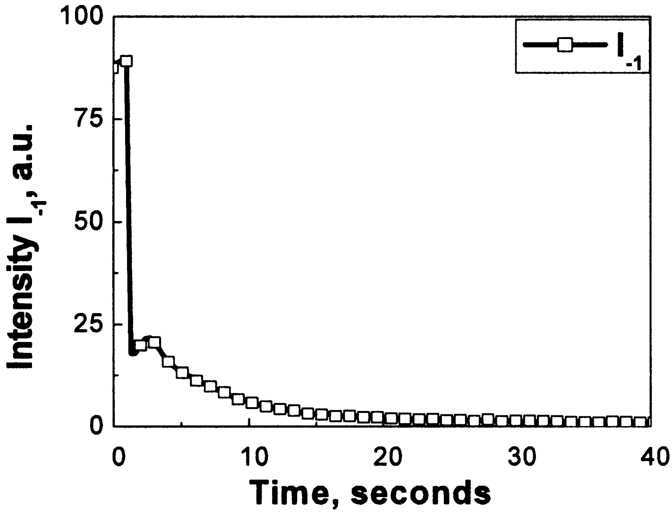

FIGURE 6 Relaxation of diffraction of (+1) non-Bragg testing beam: electric field and the writing beams are switched off simultaneously.

the cell by keeping it in the dc-field (2 V, 20 s) results in the decrease of the recording time to some tens of seconds. The relaxation behaviour strongly depends on the preceding exposure and *dc*-field parameters. At the simultaneous switching off of the electric field and writing beams, the relaxation of the grating contains both fast and slow components (Fig. 6). At t_{exp} $\leq$ 25 s the test-diffraction quickly decreases with the characteristic time, t_{decay}, 1 $\approx$ 0,5 s followed by the residual signal slowly disappearing during tens of seconds. At $t_{exp} \geq$ 35 s the relaxation becomes not-monotonic; after the quick decrease the diffraction slowly increases and followed to zero during tens of seconds. If to keep the *dc*-field on, the quick decay component disappears and the grating relaxes much slower (Fig. 7). If to keep the writing beam on and to switch the dc-field off, the sharp "splash" of diffraction is observed as was found before in [12].

The experimental data obtained allows us to suggest that the charges always presented in LC play a governing role in the mechanism of the grating recording. Our vision of the origin of the observed effect is based on modification of the model earlier proposed by Zhang *et al*. [12]. It is known that LC molecules create an adsorbed layer at ITO surfaces [17]. We suggest that application of the *dc*-field results in adsorption of charged impurities in the adsorbed LC layer on the ITO surfaces. We also suppose that light irradiation shifts a delicate equilibrium between the processes of adsorption and desorption of the charges and free charges nearby the surface. Therefore, irradiation of the cell with interference pattern causes

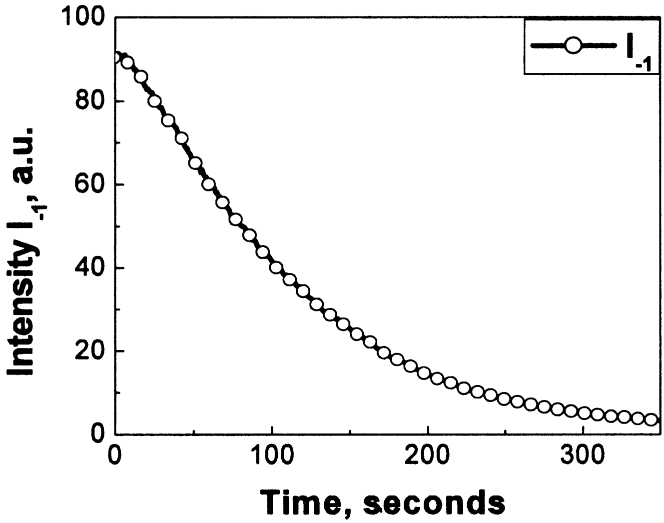

FIGURE 7 Relaxation of the diffraction of (+1) non-Bragg testing beam: *dc*-field is applied and writing beam is switched off.

in-phase charge modulation at the ITO surface:

$$\rho^{\pm}(x, z = 0) \sim (1 + \beta \cos qx), \quad (1)$$

where x is in the substrate plane and z is the normal to it. The grating (1) leads to the appearance of a modulated electric field [12]:

$$E_x(x, z = 0) = E_\rho \sin qx, \quad (2)$$

$$E_z(x, z = 0) = E_{\rho z} + E_\rho \cos qx. \quad (3)$$

Locally-strong E_x component of the surface charge electric field may re-arrange the adsorbed 5CB molecules, that results in a modulation of the easy orientation axis. The modulation of the easy axis in turn, results in a surface torque, which causes a director deviation grating in a cell bulk at any incident angle of the writing beams. The deviation of the director is in-phase with the modulation of the easy axis and phase shifted from the interference pattern (compare (1) and (2)) as verified by the observation of two-beam coupling. When the *dc*-field is switched off the modulation of the surface charges is compensated by bulk charges that hide the grating. When the *dc*-field is re-applied, the hidden grating is revealed again.

The proposed model differed essentially from the model of the Reference [12] by introducing in consideration a layer of LC molecules adsorbed on the ITO surface. This layer plays role of the agent through which the E_x-component of electric field controls the easy axis direction. In addition, the adsorbed LC layer prevents charges from the ITO electrode to compensate the surface charge modulation. It should be noted that despite the proposed model qualitatively describes the experiments, the microscopic origin of the control of the surface charges layer is unclear and requires the following studies.

3. CONCLUSIONS

In summary, we have carried out experiments that indicate a surface-charge-mediated photorefractive effect in pure LC homeotropically aligned on conductive ITO electrodes. We suggest that interfering beams modulate the distribution of the charges in the layer of 5CB molecules adsorbed by ITO surfaces. The spatial modulation of charges results in a tangential component of the *dc*-field applied to the cell, which, in turn, re-arranges LC molecules ordering in the adsorbed layer. Re-arrangement of the ordering results in modulation of the easy axis and surface angular torque in the LC bulk, which finally results in the grating recording. In addition to surface-mediated mechanism, a bulk-mediated mechanism related to an instability induced by the *dc*-field may give a contribution to the grating formation in the specific range of the recording parameters.

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