Dynamic photorefractivity in nematic liquid crystal panels with photoconducting polymeric layers

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A novel mechanism of photorefractivity in nematic liquid crystal (LC) cells is presented. Dynamic photorefractivity is based on temporal quasiperiodic modules excitation in the liquid crystal layer by applied external alternating electric field and their reorientation due to the charges induced by interference optical field on a surface of photoconducting orienting layer.

Keywords: liquid crystal, dynamic photorefractivity, novel mechanism.

1. Introduction

Photorefractive materials have numerous potential applications in the fields such as optically addressed devices, wave front correctors, and so on [1,2]. In recent years, this range has been widened by discovery of one of the most interesting and technologically promising phenomenon, i.e., a light-induced modulation of the refractive index as well as index grating recording in dye-doped LCs (DDLCs). The grating generation mechanism originating in the space charge field is similar to the processes occurring in photorefractive crystals and the effects have been named "orientational photorefractive effect" [3-23]. The wave mixing efficiency can be enhanced by orders of magnitude with minute amounts of dopants having a high photo-charge production yield [6-9]. The energy transfer and image amplification in a two-beam coupling mode is possible when there is a phase shift between a light interference pattern and a refractive index grating [1,2]. The phase-shifted (non-local) response in LCs has been obtained in the cells with photoconducting orienting layers [11-13]. In Ref. 24 we have demonstrated for the first time the dynamic enhancement of photorefractive effect in dye--doped nematic LC under ac electric field applied to the planar cell with photoconducting orienting layers. The peak of diffraction efficiency in the dynamic mode compared to orientational photorefractivity in a LC with applied dc electric field has exhibited enhancement by two orders of magnitude and fast buildup to a steady-state level [24,25]. In this paper, we investigate experimentally the novel mechanism of photorefractivity (called as dynamic photorefractivity) in the nematic planar cell under applied external ac electric field.

2. Experimental

2.1. The sample

The photorefractive effect has been studied in 9-µm-thick cell. The glass plates of the cell were coated with conductive ITO electrodes and covered with polyimide. Surface polymeric layers were uniaxially rigidly rubbed in order to provide a planar orienting of nematic LC. The cell was filled with nematic 4-trans-4'-n-hexyl-cyclohexyl-isothiocyanatobenzene (6CHBT) activated with anthraquinone dyes mixture AD-1 and AD-2 (absorption coefficients $\alpha_{\parallel} = 3300 \text{ cm}^{-1}$, $\alpha_{\perp} = 800 \text{ cm}^{-1}$) [24].

2.2. Experimental set-up

Schematic view of the wave mixing experiment geometry is shown in Fig. 1. External square-wave AC field with amplitude $U_a = 25$ V and frequency 1 Hz has been applied to the cell. The linearly polarized He-Ne laser ($\lambda = 632.8$ nm) with 25-mW power in a fundamental mode was used. A laser beam was split into two p-polarized mutually coherent input beams (pump and signal beams). P-polarization was found to be a necessary condition for observing a maximum photorefractive response. The crossing angle θ between two writing beams and the sample tilting angle α were chosen to satisfy "angular resonance" [24]. He-Ne laser beam of 0.5 mW in a fundamental mode, p- or s-polarized, has been used as a probe one. To monitor light scattering in the direction of a diffracted beam the converging lens was inserted. It has collected light scattered in a cone angle 2° on a photodiode, the diffraction order was blocked. Optical diffraction in our case corresponded to the Raman-Nath regime.

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Fig. 1. Wave mixing geometry. LC cell is tilted at an angle $\alpha = 25^{\circ}$ to the bisector of the two writing beams, crossing angle $\theta = 5.8^{\circ}$.

2.3. Results and discussion

As it can be seen in Fig. 2, the scattered light polarization possessed radial symmetry when p-polarized single beam passed through the cell. It took place even in the case of white incoherent light. The circular ring structure was reported in Ref. 27 too, but polarization research was not carried out. The authors of this paper have associated the circular ring pattern with the quasiperiodic herringbone pattern observed under crossed polarizers. As we have shown earlier [26], in the presence of ac electric field, under long-continued irradiation of LC cell, the scattering ring undergoes photo-ordering. The measurements of a polarization degree under single p-polarized beam irradiation were carried out for ordered scattering pattern. From spot to spot it magnitudes varied slightly and were ~90%, as shown in Fig. 3. Under nonpolarized beam irradiation, corresponding variations were from ~50% to ~90%.

Figure 4 shows dynamics of diffraction efficiency, light scattering, and amplification of the signal beam. To monitor light scattering in the direction of a diffracted beam, the collecting lens was inserted and the diffraction order was blocked (see Fig. 1). It can be seen that origin of an interference optical field causes diminution of the light scattering [Fig. 4(a), lower trace], increasing of the p-polarized and diminishing of the s-polarized probe beam losses [Fig. 4(b)]. The ratio of the writing beams intensities and total intensity in Fig. 4(c) were chosen far from optimal ones used in Ref. 24. It permitted to illustrate with the same trace the ac electric field action both up to, and after the pump beam switching-on.



Fig. 3. The normalized polarization curves of ordered scattered patterns under single p-polarized beam irradiation. The field circles denote the experimental results, the solid curves are approximations.

The results obtained with LC director orientation in the incidence plane or perpendicular it were qualitatively similar at given ac voltage. In all cases shown in Fig. 4, the beam switching-off transformed the temporal behaviour to reversed one with corresponding time constants.

Light interference pattern created by two mutually coherent p-polarized beams with the pump/signal intensity ratio $m \sim 1$ at the temperature less then 18°C modifies the scattering pattern. The circular ring structure seems to be broken, it assumes the form of discrete spots in both directions perpendicular to diffraction orders, as shown in Fig. 5. At $m \gg 1$, the interference field affects scattering pattern considerably less. To increase signal/noise ratio it is necessary to elevate an applied voltage as shown in Fig. 6.

Single beam passing through the cell is scattered. The cone angle of scattered light increases while as applied ac voltage raises. The corresponding optical spacing *S* changing is shown in Fig. 7. To calculate it we used the formula [26]

$$S = m\lambda/\cos[\beta + (\omega_m - \omega_{-m})/2]\sin[(\omega_m + \omega_{-m})/2],$$

where λ is the wavelength in the air, β is the beam incidence angle ($\beta = 35^{\circ}$ in our experiment), $\omega_{\rm m}$ and $\omega_{\rm -m}$ are



Fig. 2. Scattered radiation passed through polarizer. White arrow denotes the polarizer orientation. To magnify contrast of a scattered radiation it is partially concentrated by a lens. The transmitted beam is blocked.

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the angles of diffraction (scattering) in the plane of incidence, m = 1, 2, ... are diffraction orders. One needs to restore angular resonance on varying the applied ac voltage, it can be realized by cell rotation. We did it to obtain intensity distributions shown in Fig. 6.



Fig. 4. Photorefractivity dynamics in LC 6CHBT: (a) pump beam first-order diffraction (upper trace), light scattering in the direction of a diffracted beam (lower trace), (b) P-polarized (upper trace) and s-polarized (lower trace) probe beam transmission, (c) amplification of the signal beam transmitted through the cell, stationary sections of plot correspond to 50% transmission, lower trace - 0% transmission. Total input intensity is 280 mW/cm² (a, b), and 35 mW/cm² (c); pump/signal ratio is 1.7 (a, b), and 80 (c); sample temperature is 24°C (a, b), and 15°C (c). Intensity scale

is arbitrary units and time scale is 1 s/div for all cases.



Fig. 5. Interference field disturbance on a scattering pattern, photo from a screen.



Fig. 6. Intensity distribution in the direction of the first diffraction order at near- (a, b, c) and far-field (d, e, f) at $m \approx 10$. Applied ac voltage: 10 V (a, d), 15 V (b, e) and 25 V (c, f).



Fig. 7. Optical spacing dependence on applied ac voltage.

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Fig. 8. Schematics of dynamic photorefractivity in nematic LC 6CHBT cell with polyimide orienting layers under external ac electric field.

To explain the obtained results we offered the mechanism of dynamic photorefractivity schematically presented in Fig. 8. When an ac electric field is applied to the cell, system of the orientational quasiperiodic "subgratings" is excited in the LC layer [Fig. 8(a)]. We suppose, that the polarization of extraordinary wave is directed along the stripes, and ordinary one is perpendicular by them. Subgratings amplitudes (or director deviations) depend on the applied electric field and LC parameters, such as anisotropy, elastic constants and so on. The interference optical field induces charge distribution on photoconducting orienting layers [Fig. 8(b)] and thereby modulates the surface anchoring [22]. Thus, a surface-charge grating acts on dynamic "subgratings" and reorients amount of them, probably, stabilizing their phase and spacing [Fig. 8(c)]. Obviously, the reorientation rate should be increased at higher voltage that Fig. 6 illustrates. It is evident that maximal efficiency should be observed when a spacing of "subgratings" is close to surface-charge period. The complete wave front conjugation of complex optical field is inaccessible due to the "subgrating" spacing equal to ~6 µm. These features were experimentally described in our previous paper [24]. In the case of a single beam or white one passing through the cell a nonperiodical charge is being induced on a surface of the orienting layer. With time the charge field can order the structure of "subgratings" in illuminated space [26]. Our preliminary experiment has shown that dynamic scattering intensity increases with temperature, but ordering reduces in investigated span 10-30°C.

The maximum wave mixing efficiency was observed in a temperature span 15-18 °C, and at the temperatures higher than 20 °C it decreased fast.

It should be noted that instabilities responsible for light scattering and enhancement of wave mixing efficiency are not the Williams domains:

• the Williams domains are simply observed with polarizing microscope [28], but in our case it is not so [26]. Moreover, in our experiment we have observed that even at angular resonance and signal/pump ratio $m \sim 1$ while as maximal self-diffraction occurs, the probe beam diffraction efficiency approaches to zero at its normal incidence. It means, that the angle of the director deviation is not continuous function of coordinates, for example, it is a zigzag,

- the Williams domains spacing does not reveal continuous changing with the applied voltage [28], in our experiment it does (see Fig. 7),
- the instabilities are exited effectively at the frequencies
 10 Hz whereas the Williams domains are observed even at the frequencies > 100 Hz [28].

3. Conclusions

Classical orientational photorefractive effect in nematic LCs is connected with local reorientation of the director in the combined external dc electric and space-charge fields. Photoexcited charge is spatially modulated according to the interference optical field, i.e., has the same periodicity. In nematics, the index grating amplitude is usually a small part of their birefringence and due to the balance between anchoring forces and spatially modulated electric field. We have obtained the dynamic enhancement of photorefractive effect in dye-doped nematic liquid crystal in the presence of an applied ac electric field [24]. In the present work we have shown, that it is not one-way process. Only effective action of an interference field on instabilities exited by ac field results in multi-wave efficiency enhancement. We have shown that novel mechanism is based on index grating assembling of the modules exited by an applied external ac electric field. The amplitudes of instabilities are determined by applied field magnitude and cell parameters. The interference optical field induces charge distribution on a photoconducting orienting layers and thereby modulates the surface anchoring. As a result, the surfacecharge grating reorients instabilities azimuthally, also stabilizing their spacings. Therefore wave mixing efficiency in a dynamic mode may exceed considerably its magnitude in a classical orientational photorefractive effect. We obtained phase-conjugated reflectivity in degenerate four-wave mixing of as much as 80% in Ref. 24.

Certainly, it is very simplified flattened simulation. Some experimental results, not included in this paper, revealed that it is necessary to take into account a three-dimensional model. It is obvious that further investigations are needed to clarify proposed mechanism of dynamic photorefractivity.

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