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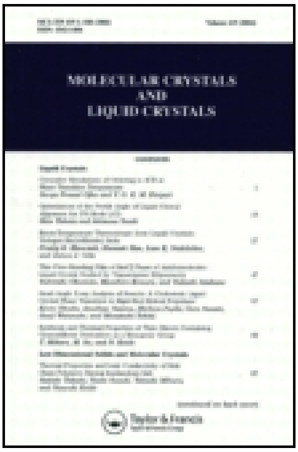

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

Effect of Optical Nonlinearity Dynamical Enhancement in Dye Doped Liquid Crystal Under A Electrical Field

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Version of record first published: 18 Oct 2010

To cite this article: A. V. Agashkov, A. A. Kovalev, S. S. Serak & J. Parka (2002): Effect of Optical Nonlinearity Dynamical Enhancement in Dye Doped Liquid Crystal Under A Electrical Field, Molecular Crystals and Liquid Crystals, 375:1, 269-280

To link to this article: http://dx.doi.org/10.1080/10587250210565

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Mol. Cryst. Liq. Cryst., Vol. 375, pp. 269-280
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Effect of Optical Nonlinearity Dynamical Enhancement in Dye Doped Liquid Crystal Under A Electrical Field

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We report observation of the dynamic enhancement of photorefractive effect in dye-doped nematic liquid crystal in the presence of an applied ac electric field. The multi-wave mixing efficiency in a planar cell with photoconductive orienting layers has been increased by two orders.

Keywords: liquid crystal; dye; laser; enhancement; photorefractivity; multiwave mixing

INTRODUCTION

Photo-refractive materials have numerous potential applications in fields such as high density holographic data storage, optical switching and addressing, wave front conjugation and so on[1,2]. The "orientational photorefractive effect" in nematic liquid crystals (LC) has been intensively studied during last years[3-20]. The main attention of investigations has been focused on the enhancement of the diffraction efficiency of recorded gratings by means of a new design solutions as well as application of new materials. It is known that the efficiency of the wave mixing increases monotonically with increasing a light incidence angle up to 45° [10]. The dependence of diffraction efficiency on external dc electric field has a maximum, while without field photorefractive effect practically absents[3-5,11,12,18]. A considerable enhancement of the wave mixing diffraction efficiency can be achieved by adding to the LC dopants having a high photo-charge production yield[6-8,22]. 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]. This feature is highly important for practical application. The phase-shifted (non-local) response in LC's has been obtained in cells having photoconducting orienting layers[11,12].

Electrohydrodynamic (EHD) instabilities excited in LC's with ac field have been the subject of extensive research for a long time, and various types of domain patterns have been described[23]. But still hitherto this LC feature is not of practical use. It is known, that applying an external ac electric field the nonlinear response of photorefractive crystals to the light illumination is significantly increased[24]. Therefore, investigations of photorefractive effect in dye-doped LC's in the presence of an ac electric field are of an undoubted interest. In our previous work[25] we observed for the first time an ordering of the electrically excited domain pattern under the action of He-Ne laser radiation of low power (1 mW). In this paper we present experimental results on multi-wave mixing in dye-doped nematic LC under applied ac electric field of square-wave form. Several wave processes, such as probe beam diffraction, self-diffraction, degenerate four-wave mixing (DFWM) with optical phase conjugation and degenerate two-wave mixing (DTWM) with image amplification have been studied. #

EXPERIMENT

The sample

The sample was a 9 μm thick cell, its substrates 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 mixture of anthraquinone dyes AD-1 and AD-2 (absorption coefficients at wavelength λ = 633 nm are $\alpha_{\parallel}$ = 3300 cm^{-1}, and $\alpha_{\perp}$ = 800 cm^{-1}). Nematic LC 6CHBT exhibits positive dielectric anisotropy $\Delta\varepsilon$ = 8 (its components are $\varepsilon_{\parallel}$ = 12.0, $\varepsilon_{\perp}$ = 4.0), it has the refraction indices n_e = 1.672 and n_o = 1.520 and Freedericksz threshold U_{FR} = 1.9V obtained in the 40 μm thick cell.

Part of our work has been presented on the conference ELC2001 (Zakopane, Poland) and will be published in SPIE proceedings

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Experimental set-up

The geometry of the wave mixing experiment in LC cell is shown in Figure 1.

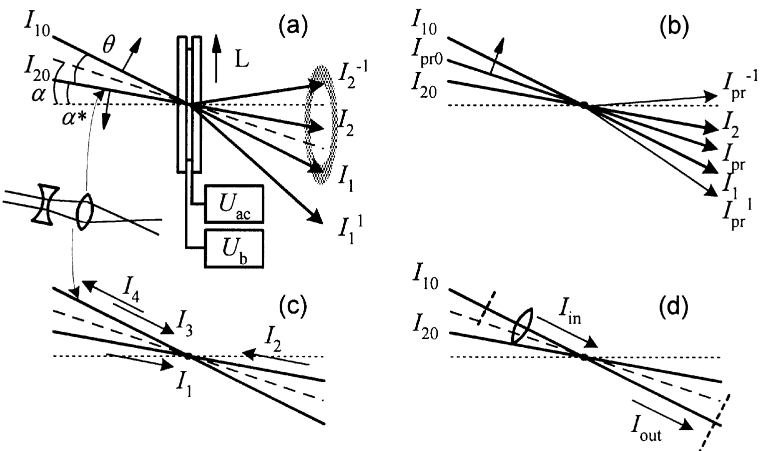

FIGURE 1 Wave mixing geometry: self-diffraction and energy transfer (a), probe beam diffraction (b), wave front conjugation in DFWM (c), image amplification (d)

The laser used is a linearly polarized He-Ne laser (λ = 632.8 nm) with power of 25 mW in a fundamental mode. Laser beam was split into two p-polarized mutually coherent input beams (pump and signal beams). The attenuation of input beams power has been made by means of calibrated neutral light filters. The experimental geometry of self-diffraction and energy transfer between beams is shown in Figure 1a. To investigate a possibility of distortion correction in DTWM experiment the same setup has been used. Probe beam diffraction configuration is shown in Figure 1b. P-polarized He-Ne laser beam of 0.5 mW in a fundamental mode has been used as a probe one. The geometry of wave front conjugation in DFWM process is shown schematically in Figure 1c. The distortions of a signal beam in DFWM and more powerful pump one in DTWM have been made by means of the optical system. It consisted of positive and negative lenses to provide the divergence of the beam and the unchanged spot size in the LC cell. The image amplification geometry is shown in Figure 1d. The transparent object (scale with division of 0.1 mm) and image forming lens were positioned in front of the cell. The image plane was behind the cell at the distance of 30 cm (60 cm for interference pattern recording). The director of the LC layer was in the plane of incidence for all configurations shown above.

We have defined the efficiency of self-diffraction of the beams I_{i0} (i = 1,2) as $\eta_i = I_i^1/I_{i0}$, the efficiency of the probe beam diffraction as $\eta_{pr} = I_{pr}^1/I_{pr0}$, the phase-conjugate reflectivity in DFWM as $R = I_4/I_3$, the signal beam I_{10} (image I_{in}) gain in DTWM as $g = I_1/I_{10} = I_{out}/I_{in}$, and the pump / signal intensity ratio as $m = I_{20}/I_{10}$ (for DFWM as $m = I_{10}/I_{30}$).

The combined electric field has been applied to the LC cell. It is schematically shown in Figure 2. The square-wave unipolar field with controllable amplitude U_{ac} in a range of 0 - 50 V and repetition rate f in a range of 0.5 - 10 Hz have been applied to only one conducting layer of the cell. The controllable dc bias voltage U_b has been applied to another one. A combination of the dc and ac voltage has been used to achieve maximum efficiency of wave mixing. To study temporal behavior of transmitted and diffracted beams a light chopper and photodiode detectors, connected to the oscilloscope, were used. All the dependencies shown in this work were obtained at $f = 1$ Hz.

We have found out an enhancement of wave mixing efficiency at the presence of an ac electric field. We have also found out, that the maximum effect is achieved, when the crossing angle θ between the two incident beams is close to the angle of the first-order diffraction on the periodic structure excited in the cell by the external ac field. To reach this effect, the tuning of the beams crossing angle θ has been made. At first one beam was passed through a cell only. Its first-order diffraction pattern looked like diffusive ellipse pulsing in time with electric pulse edges. The ellipse turned into the ring for the case of normal incident beam. The diffraction pattern at $\alpha = 0°$ corresponded to the domain structure optical period $\Lambda_d = 6.5$ µm ($\Lambda_d = \lambda/\sin(\theta_d)$, $\theta_d = 5.6°$ - diffraction angle). It means,[23] that vortex size $d \approx \Lambda_d/2 \sim 3$ µm.

The second beam intersected with the first one in the LC layer in such a way that it was directed on a diffraction ellipse after passing the cell. In the experiment the incidence angle α was taken equal to 25°, which corresponds to high enough efficiency of the wave mixing under applied dc electric field. Corresponding to this the experimentally found crossing angle of the beams $\theta = 5.8°$. Using the appropriate equations[25] (with some corrections), it is possible to calculate an optimal crossing angle θ

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$$\theta = \arcsin[(\lambda/\mathrm{m}\Lambda_\mathrm{d})(f\sin\alpha^* - 1)] - \alpha^*, \tag{1}$$

$$\theta_{(-)} = \alpha^* - \arcsin[(\lambda/\mathrm{m}\Lambda_\mathrm{d})(f\sin\alpha^* - 1)], \tag{2}$$

$$f = \left[(\Lambda_\mathrm{d}/\mathrm{m}\lambda)^2 - (\mathrm{n}^2 - \sin^2\alpha^*)^{-1}\right]^{1/2} \tag{3}$$

$$\alpha = \alpha^* + \theta/2 \tag{4}$$

where m =1 (the first-order diffraction) for a given case. The angle $\theta_{(-)}$ is formed, when the second beam is directed closer to normal (coincides with direction of a beam I_2^{-1} on Figure 1). The equations for θ and $\theta_{(-)}$ at m = 1 can be simplified

$$\theta \approx \arcsin(\lambda/\Lambda_\mathrm{d} + \sin\alpha^*) - \alpha^* \tag{5}$$

$$\theta_{(-)} \approx \alpha^* - \arcsin(-\lambda/\Lambda_\mathrm{d} + \sin\alpha^*) \tag{6}$$

By changing angle α about initially selected value one can move away from the optimum position. It can be expected to reach even greater efficiency at α = 45°. However it will be difficult to establish presence of an "angular resonance" in this case. In accordance with the work[10] the wave mixing efficiency depends on an incidence angle as sin(2α) reaching maximum at α = 45°, too.

Results

To analyze the grating conditions a dimensionless parameter Q is used[2].

$$Q = 2\pi\lambda l / \mathrm{n}\Lambda^2,$$

where l is the thickness of the LC layer, n is a refractive index of the medium, and Λ is a grating period. One defines the regime Q > 1 as the Bragg regime of optical diffraction (thick grating). In this regime only one order of diffraction is produced. Another regime Q < 1 is the Raman-Nath regime (thin grating), and many orders of diffraction can be observed. In our case Q < 1, which corresponds to the Raman-Nath regime.

The dependence of wave mixing efficiency on the incidence angle α, the recording beam intensities ratio m, total intensity I_{sum} and bias voltage U_b have been investigated. Figure 2 shows the position of

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diffracted light pulses with respect to the applied electric pulses. The potential of the conducting layer with applied dc voltage bias is set to be zero. It was necessary to use asymmetrical ac voltage because the distortions of the beams visibly increased in case of usual bipolar voltage with amplitude $U_{ac} < 20$ V (in our notation $U_b = U_{ac}/2$). Sets of oscillograms for sequence of bias voltages are shown in Figure 3: single beam transmitted through the cell (a), first-order diffraction I_{pr0} on the

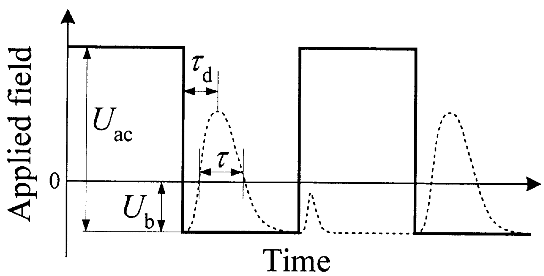

FIGURE 2 Schematic depiction of applied electric field. The solid curve denotes electric field, and dotted one denotes diffraction pulses. U_{ac} is the amplitude of ac field, and U_b is the bias voltage; τ_d is the time delay, and τ is the width of diffraction pulse.

dynamic holographic grating recorded by I_{10} and I_{20} beams (b) and reflection from phase-conjugated mirror in DFWM (c). As the experiment has been shown the single beam power diffracted on domain structure is going up with increasing α. But contribution of diffraction losses in total optical losses is less then 10% even at $\alpha = 45°$ (bipolar voltage, $U_{ac} = 20$ V)[25]. Therefore the main contribution into the pulse transmission reduction is given by the change of dye molecules absorption due to their reorientation. The amplitude and the shape of the single beam transmission pulse were not changed by changing the incident light intensity in a range of 0.001-1 W/cm^2 (neutral light filters were replaced from the position behind the cell to the position in front of it). The pulse width decreased with 30 up to 5 ms when bias voltage U_b was increased with 5 to 25 V ($U_{ac} = 50$ V). If signal I_{10} and pump I_{20} beams were switched on the losses of I_2 and I_{pr} noticeably increased due to the diffraction on the grating. It is clear seen, that at given U_{ac} and U_b the width of diffracted optical pulses (Figure 3b,c) close to the width of the single beam transmission pulses (Figure 3a). The wave mixing efficiency is time-dependent in our method. Therefore to plot η_i, η_{pr}, R and g dependencies, their peak values calculated from oscillograms have been used. It should be noticed that after writing beams were turned on, these pike values reached steady-state meanings after two-three oscillations of electric field.

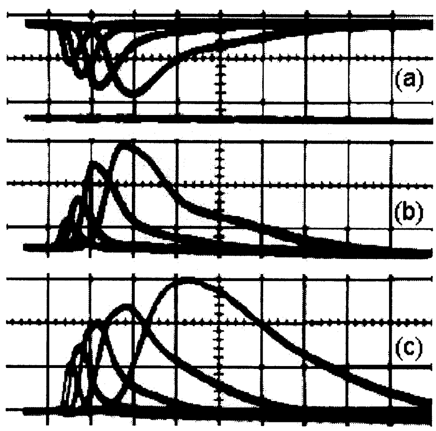

FIGURE 3 Oscillograms of the probe beam transmission without recording of the grating (a), first-order diffraction on the dynamic grating (b), and wave front conjugation in DFWM scheme (c). U_b = 10, 15, 20, 25 V (a, b), and U_b = 5, 10, 15, 20, 25 V (c); m = 0.58 (b), and 230 (c). Here α = 30°, U_{ac} = 50V, triggering by negative front of electrical impulse, scale factor is 10 ms/div

Figure 4 shows the dependences of η_{pr}, R and g on incidence angle α. All curves have a maximum nearby α = 25°. The comparative investigations showed that at the presence of only dc electric field η_{pr} and R values monotonically grew with increase of incidence angle up to α = 45°. But even in a case, when the optimum bias voltage was applied, any of these magnitudes have not exceeded 0.5%. Thus the probe beam diffraction efficiency η_{pr} and the phase-conjugate reflectivity R has been increased as much as two orders nearby the angle resonance in the presence of ac electric field.

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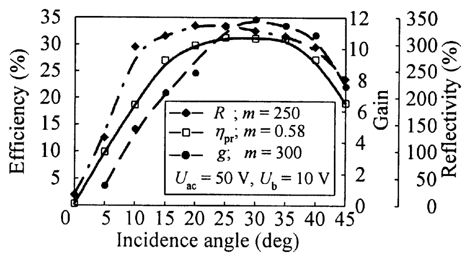

FIGURE 4 The dependence of the wave mixing efficiency on incidence angle α: ◆ the phase-conjugate reflectivity R of DFWM mirror; □ the probe beam diffraction efficiency η_{pr}; ● signal beam gain g in DTWM scheme

The efficiency of the wave mixing process depends on the intensity ratio of the pump and signal beams m and the total laser beam intensity $I_{sum} = I_{10} + I_{20}$, Figure 5 and Figure 6 show some of them. As can be seen from Figure 5 the efficiency of self-diffraction at fixed I_{20} is going down with increasing of I_{10}. The dependence of the probe beam diffraction efficiency η_{pr} on total intensity I_{sum} is shown in Figure 6a, all curves reaches a stationary level of $\eta_{pr} \approx 30\%$. The dependences of the phase-conjugate reflectivity R of DFWM mirror on the I_{sum} are shown in Figure 6b. Maximum phase-conjugate reflectivity R of as much as 800% was obtained. The achieved value is not a limit, because the saturation of R at fixed m is not observed.

The investigation of the energy transfer between beams is of special interest. The dependence of the signal beam gain g on the intensity ratio of recording beams m is shown in Figure 7a. The maximum reached gain g = 20. Figure 7b shows the signal beam pulsed gain dependences: amplitude g, width τ and time delay τ_d on bias voltage U_b. It should be noticed that at m >>1 the dependences of the optical pulse width τ on U_{ac} and U_b voltage for self-diffraction, probe beam diffraction, DFWM and signal beam (image) gain are practically identical. It is obvious that for the given electric field it depends mainly on the physical properties of LC such as viscosity, elasticity, dielectric anisotropy and the surface anchoring energy. At m ~1 the energy transfer between the beams undergoes oscillations. Thus at U_{ac} = 50 V and U_b = 10 V there were observed up to five oscillations in gain and attenuation of beams during more then 100 ms.

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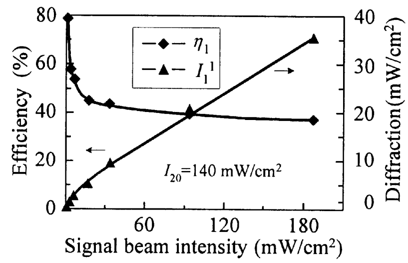

FIGURE 5 Self-diffraction efficiency η_1 and first-order diffraction intensity $I_1^{\ 1}$ versus signal beam intensity I_{10}

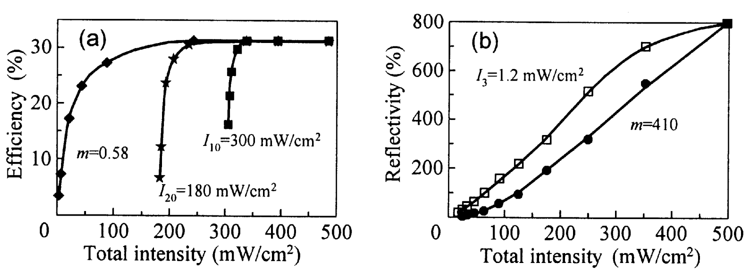

FIGURE 6 Probe beam diffraction efficiency η_{pr} (a), and the phase-conjugate reflectivity R (b) versus the total input intensity I_{sum}

For a comparison, at $m >> 1$ the single pulse of ~20 ms duration was observed. At $\alpha > 5°$ and $m >> 1$ energy transfer is always observed from intense to low-intensity beam, independently from the direction of applied ac electric field for a given moment. In this aspect the considered photorefractive regime substantially differs from the one described, where the sign of energy transfer in the two-beam coupling process

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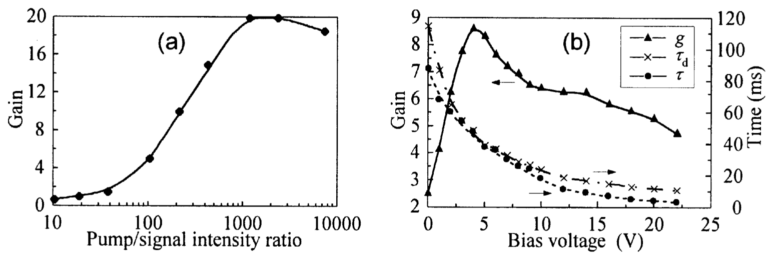

FIGURE 7 Signal beam gain g versus pump/signal intensity ratio m (a). Bias voltage behavior of the gain pulse (b): ▲ amplitude g; ● time duration τ; × time delay τ_d

depends on the direction of dc field[5,12,17,18,21]. The amplified object image at $m = 340$ is shown in Figure 8a. The interference pattern, which arises when the wedge-like plate is inserted in one of the beams, is shown in Figure 8b. If the ac electric field is acting the interference pattern remains during 5 minutes.

The experiment (see Figure 1a,c) demonstrated good correction of spherical distortions in DFWM and DTWM. The far-field patterns of the initial (no distorted) beam and corrected one practically coincided if distortion focal distance exceeded 30 cm. We have not succeeded in complete correction of complicated distortions because the diffraction was in the Raman-Nath regime with large enough grating period of ~6 μm.

It should be noticed, that the dynamic enhancement of the optical nonlinearity is observed too, when the single beam is reading the low-amplitude grating earlier recorded on the interface of the orienting layer. Required condition is the closeness of the grating spacing to the excited domain period. We observed the diffraction orders under applied ac electric field and no diffraction have been observed under applied dc electric field.

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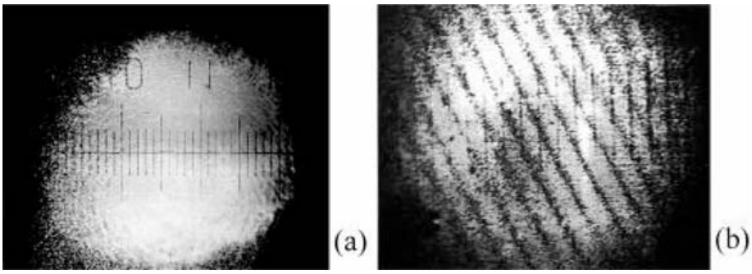

FIGURE 8 Amplified image in DTWM scheme (a) and its interferogram (b); scaleplate with scale factor of 0.1 mm

CONCLUSION

In this work we 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. We believe that the photoconductivity of orienting layers and photorefractive properties of dye-doped LC play a role in the non-local index gratings formation. The possible contributions of EHD instabilities at "angular resonance" are:

(i) vortexes produce a dynamic reorienting action that coincides with operation of the light-induced space charge field;

(ii) the space charge field of EHD instabilities interacts with the light-induced one. When their periods are close, and the phases differ less than on π, one can expect resonance effect;

(iii) at a vortex size $l \approx d/3$ the three-layered structure of the nematic director distribution is possible. The diffraction on it can give substantial enhancement of the intensity maxima relative to a one-layer grating.

Further investigations are needed to clarify a mechanism of the revealed photorefractive effect enhancement.

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