Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

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S. V. Serak [a], A. A. Kovalev [a] & G. L. Nekrasov [a]

[a] Institute of Electronics Belarus Academy of Sciences, 220090, Minsk, Logoiski trakt, 22

Version of record first published: 04 Oct 2006

To cite this article: S. V. Serak, A. A. Kovalev & G. L. Nekrasov (1997): Dynamic Holograms Induced by Nanosecond Laser Pulses in Bleachable Dye Activated Liquid Crystals, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 299:1, 111-116

To link to this article: http://dx.doi.org/10.1080/10587259708041982

Mol. Cryst. Liq. Cryst., 1997, Vol. 299, pp. 111–116
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DYNAMIC HOLOGRAMS INDUCED BY NANOSECOND LASER PULSES IN BLEACHABLE DYE ACTIVATED LIQUID CRYSTALS

S.V. SERAK, A.A. KOVALEV, G.L. NEKRASOV
Institute of Electronics, Belarus Academy of Sciences
220090 Minsk, Logoiski trakt, 22

Abstract. Orientation effect induced in bleachable dye doped planar nematic layer has been observed. It is believed that the effect is caused by dye-induced torque and hydrodynamic reorientation of the layer.

INTRODUCTION

Liquid crystals activated with bleachable dyes are of special interest because they can be successfully used in laser systems as both passive filters with variable initial characteristics[1] and WFC-mirrors for intra- and extracavity correction and reconstruction of wave fronts[2]. The authors of ref [2] have reported wave-front conjugation of monopulse ruby laser radiation in four-wave mixing in homogeneously oriented activated nematic layers. They have shown that wave-front conjugation efficiency is considerably dependent on monopulse intensity and layer temperature, and is determined by various mechanisms of activated LC response to incident radiation.

Up to now, a variety of physical processes have been revealed in LC fluids that are induced by high-power laser radiation and coupled to changes in such basic parameters as density, temperature and orientation direction of the layer. Among them are optical reorientation [3] and generation of ultrasonic waves [4] in the field of nanosecond laser pulses. Khoo and others [5], Macdonald and Eichler [6] have observed flow-alignment in a homeotropic 5CB layer under excitation with picosecond pulses. Flow-alignment is caused by formation of ultrasonic waves as a result of thermal expansion or electrostriction and by photoelastic stress[7]. Also possible in absorbing LCs is photo-hydrodynamic reorientation flow arising from pressure difference under thermal volume expansion [8].

In addition to traditional saturation of absorption and heating, dye-activated LC fluids can display specific orientational effects where nonlinearity parameters are higher. For example, light-induced Frederiks transition in activated LC is characterized by threshold two orders of magnitude lower than that observed in a pure LC [9]. Janossy has given [10] molecular interpretation of this phenomenon based on the assumption that the light-induced torque is a superposition of the direct optical torque

and a "dye-induced" torque. The orientational effect of this kind has been observed in a planar dye-doped LC cell under the action of Ar^+ laser radiation [11] Specific behavior of activated LC medium consists in the fact that polar complexes can also be formed from excited dye and solvent molecules. The authors of refs. [12,13] have discovered Ar^+ laser-induced reorientation of layer in static d.c. electric field.

In order to gain a deeper insight into peculiarities of interaction of monopulse radiation with bleachable-dye-activated LC, we have studied dynamic holograms by using a probing c.w. laser.

EXPERIMENT

A ruby laser (λ = 694,3 nm, pulse energy W_p = 0,3 J and duration t_p= 80 ns) was used as a pump source. Pump radiation was split into two beams E_1 and E_2 of equal intensity which converged on a cell at an angle of 2° 20′ into a spot of 2 mm diameter (pin-hole cuts out a spot uniform over area). In this case, the grating period Λ was 33 μm. The thickness of the planar sample was 50 μm. For LC orientation we used the deposition of silicon monoxide layer on glass substrates. To exclude induced phase transition at room temperature, we used as a solvent five-component mixture of cyanobiphenyls (CB-5) with nematic-isotropic transition temperature of 56°C. Two bleachable dyes were used: 4,11-di(phenyl-3'4') bisantene (DPB) and vanadyl phtalocyanine (VOPhc). DPB is characterized by a linear absorption oscillator and,

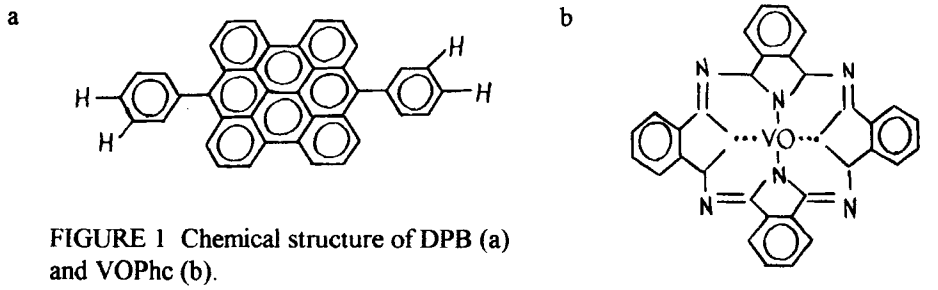

FIGURE 1 Chemical structure of DPB (a) and VOPhc (b).

consequently, high absorption dichroism in LC (d = 0.7), its ordering degree being $S_{DPB}=S_{LC}=0.63$. Maximum absorption coefficient at λ = 694,3 nm is $k_{||}$= 49 cm^{-1}. VOPhc-dye exhibits low dichroism with d = 0.12 ($k_{||}$= 55 cm^{-1}) and has a plane absorption oscillator. Typical concentration of the samples is 10^{-3} M.

Dynamics of LC response was investigated by using He-Ne laser radiation (λ=632.8 nm, 30 mW power). The probe and pump beams had the identical polarizations $E_{probe} \parallel E_{pump}$. First-order diffracted probe beam provided information about characteristic relaxation process in the cell, while control of dynamics in the interaction region as a whole enabled us to estimate medium restoration times. For the latter purpose, a photomultiplier was used with high photocurrent, which recorded an increase in the cell transmission as a pulse of "negative polarity" (below zero-line) and a decrease in transmission as a "positive" signal.

RESULTS AND DISCUSSION

Under monopulse excitation LC-dye solutions get bleached, VOPhc featuring a two-level saturation scheme s_1 - s_2 [14] (Fig..2a) and DPB involving metastable level (2b)[15]. We have found the lifetime in triplet to be $3 \cdot 10^{-6}$ s. Pump intensities at which solutions are only bleached are $< 5 \cdot 10^5$ W/cm^2. As power is increased, there occurs scattering on nonuniformities which is seen at $W_p < 10^7$ W/cm^2 even on the monopulse leading edge (2c). Against the background of the scattering, damped oscillations were observed which left the interaction region in about 500 ns (2c). The latter value agrees with calculations where sound velocity in LC is taken to be 1540 m/s [4] and the distance from the middle of excitation "gaussian" spot on the cell is 1 mm. Initial transmission of the layer was restored in 100 ms.

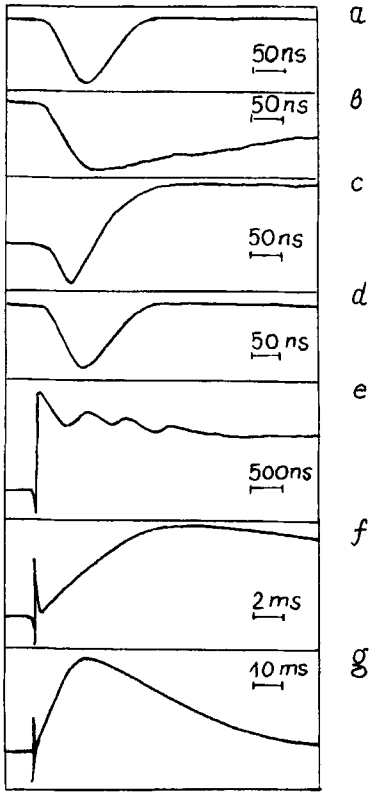

FIGURE 2 Oscillograms showing variation of probe radiation transmission: a,b - $W_p = 5 \cdot 10^5$ W/cm^2; d - excitation pulse; c,e,f,g - 10^7 W/cm^2; (DBP and VOPhc in LC).

Fig. 3 shows typical oscillograms of intensity variation of the first-order-diffracted probe beam on different time scales in DBP (a,b) and VOPhc (c,d) for the following interaction geometries: the layer director **L** is parallel to pump waves polarization vectors $\mathbf{E}_1$ and $\mathbf{E}_2$ ($\mathbf{L} \parallel \mathbf{E}_1 \parallel \mathbf{E}_2$); **L** is perpendicular to them ($\mathbf{L} \perp (\mathbf{E}_1 \parallel \mathbf{E}_2)$), and polarizations are crossed $\mathbf{E}_1 \perp \mathbf{E}_2$. Variations of the refractive index causing probe beam diffraction due to both thermal and orientational nonlinearities have additive nature: for $\mathbf{L} \parallel \mathbf{E}_1 \parallel \mathbf{E}_2$ n_e decreases and for $\mathbf{L} \perp (\mathbf{E}_1 \parallel \mathbf{E}_2)$ n_o increases. Two processes with relaxation times of about 200 μs and 50 ms, respectively, can be discerned. The first one is associated with blurring of the grating due to thermal diffusion. Its characteristic time defined by the known relationship $\tau_T = \rho \cdot C_p \Lambda^2 / \lambda_T 4\pi^2$, where ρ is the density, C_p is the heat capacity, λ_T is the thermal conductivity coefficient, will be 250 μs for typical LC's ($\rho = 10^3$ kg/m^3, $C_p = 1500$ J/ K· kg, $\lambda_T = 0.16$ J/ K·s·m). The duration of the second slow process corresponds to orientation

relaxation and agrees well with calculated value $\tau_\Theta = \gamma \Lambda^2 / 4\pi^2 K$ =30 ms, where γ is the viscosity-related constant (0.1 Poise), K is the elastic constant (10^{-6} dyne).

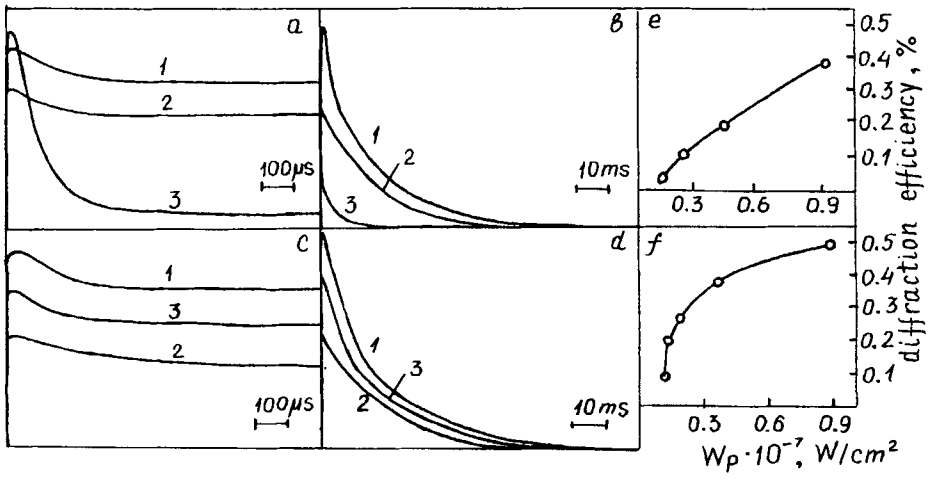

FIGURE 3 Oscillograms showing changes in first order diffraction intensity for grating recorded in DPB (a,b,e) and VOPhc (c,d,f): 1 - $\mathbf{L} \parallel (\mathbf{E}_1 \parallel \mathbf{E}_2)$; 2- $\mathbf{L} \parallel \mathbf{E}_1 \perp \mathbf{E}_2$, $\mathbf{E}_{probe} \parallel \mathbf{E}_1$; 3 - $\mathbf{L} \perp (\mathbf{E}_1 \parallel \mathbf{E}_2)$; $W_p = 10^7$ W/cm^2. Diffraction efficienty dependence of the pump beam power for reorientation component ($\mathbf{L} \parallel \mathbf{E}_1 \parallel \mathbf{E}_2$).

For DPB, efficiency of orientational grating depends considerably on the interaction geometry and excitation intensity. For the cases $\mathbf{L} \parallel \mathbf{E}_1 \parallel \mathbf{E}_2$ (3-1) and $\mathbf{E}_1 \perp \mathbf{E}_2$ (3-2) shows reorientational component clearly. The diffraction efficiency was measured to be 0,4% and 0,2% for a pump beam intensity of 10^7 W/ cm^2 , respectively. This fact is natural for crossed polarizations where thermal and acoustic gratings are largely suppressed and can only emerge due to depolarizing factors. It should be noted that dye plays a leading role in reorientation in view of the fact that in the geometry $\mathbf{E}_1 \perp \mathbf{E}_2$ reorientation was not observed in pure LC at intensities $<5 \cdot 10^7$ W/cm^2. As the cell is rotated by 90° ($\mathbf{L} \perp (\mathbf{E}_1 \parallel \mathbf{E}_2)$, (3-3)) efficiency of the orientational components falls abruptly, and the process is mainly controlled by thermal damping. The diffraction efficiency for the thermal component is 0,8%. A decrease in the monopulse intensity leads to decreased intensity of the orientational component (3e,f). For VOPhc molecule with weak dichroism another behavior is observed. In case of geometries $\mathbf{L} \parallel \mathbf{E}_1 \parallel \mathbf{E}_2$ and $\mathbf{L} \perp (\mathbf{E}_1 \parallel \mathbf{E}_2)$, the diffraction efficiency of orientation process differs slightly (0,5% and 0,4% respectively). Efficiency of a polarization grating was slightly less (0,3%). This may be connected with the experiment error. (Twist-cell rotated the plane of polarization).

The supporting evidence in favour of reorientation is provided by radiation-induced static holograms which are formed when power or absorption coefficient are increased (power $>5 \cdot 10^7$ W/cm^2). We have previously observed such holograms [16].

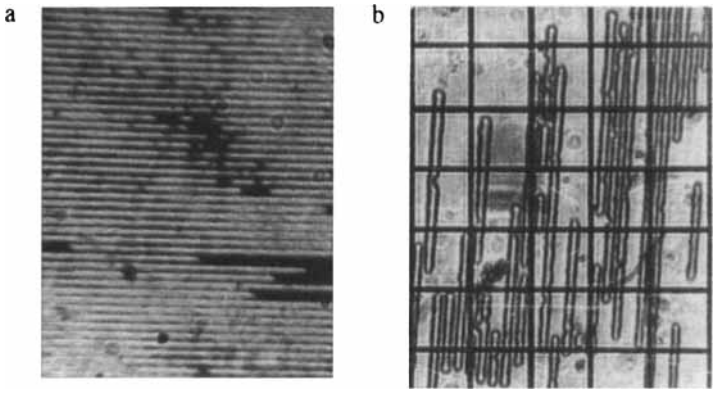

FIGURE 4 .

Fig.4 shows microphotographs of holograms recorded at 500 line/mm. We have managed to record interference field at the spatial frequency of 800 line/mm. The holograms was stored a few months. Orientation can be easily restored by applying < 30 V electric field. Static holograms represent equidistant regions surrounded by walls between which LC molecules have a stable orientation which differs from the main orientation by a small pre-tilt (5-10°) to glass surface (4b). The investigation under microscope is showed that the walls are bounded on sample surface by linear disclinations [17]. With our experimental geometry, formation of walls in pure LC at 10^8 W/cm^2 was observed.

We assume that orientational phenomena in dye activated LC's may have different mechanisms. When dye molecules switch to their ground state as a result of radiationless deactivation, there occur temperature rise in microvolume (micro-boiling is even observed in isotropic volatile solvents [18]), microvolume expansion, and generation of density waves and hydrodynamic flow, the latter perturb layer orientation, especially, in the center of the cell where cohesion with substrate is weak. The time required for the establishment of hydrodynamic velocity $\tau_F = 2\rho \cdot \Lambda^2 / \eta \, 4\pi^2$, [5], where η is the flow viscosity ($7 \cdot 10^{-2}$ kg/m·s), is 350 ns. The fact that wall formation threshold is an explicit function of the absorbed energy (in a pure LC, absorption can be affected by various impurities) and that walls are observed in the geometry $\mathbf{L} \,||\, \mathbf{E}_1 \,||\, \mathbf{E}_2$, suggests that there occurs flow-alignment in the layer.

On the other hand , for the excited molecules of DBP in LC , wich have a long - lived excitation level and very anisotropic shape and are bleaching by the monopulse leading edge, we assume, that one may be observed the induced rotation. The rotation may be caused both the field of LC surrounding and the field of light wave. When the molecules go to excited state, energy is concentrated in that part of molecule which determines the corresponding absorption band. Changing of the electron density distribution manifests itself in changing of polarizability of molecule and main axis of induced polarizability ellipsoid, in general case does not coincide with the light wave electric vector. However, detailed examination of this mecanism required investigatios of other dyes and exitation times.

We have reported our experimental observations of the orientational effects in bleachable-dye-activated LC's. A more detailed analysis of hologram efficiency and study into the mechanisms causing formation of wall-surrounded stable regions with tilted director orientation would lie beyond the scope of this paper.

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