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“Domain”-Like Reorientation in Dye Doped Nematics Induced by High-Power Radiation

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High power laser ($P \sim 10^6 \text{ W/cm}^2$) induced “domain”-like reorientation in homogeneously oriented by silicon monoxide dye doped liquid crystals (DDLC) has been observed. Reorientation can be accompanied with writing of static gratings that are induced by a radiation on monoxide layer surface.

Keywords: reorientation; dye; liquid crystals; laser; grating

INTRODUCTION

A static grating formation induced with monopulse radiation from ruby laser (at a power of $5 \cdot 10^6 \text{ W/cm}^2$) has been reported previously^[1-2]. This phenomenon was observed in the dye doped nematic layers oriented by silicon monoxide. The storage time of those gratings was a few weeks. We found that at the forming of static gratings the strong orientational effect is simultaneously observed. In order to study the physical mechanism of reorientation we carried the electron and polarizing microscope investigations. The dynamics of formation of gratings, induced in DDLC by power laser radiation also was studied.

1. Experiment

Experimental arrangement is shown in Fig. 1a. The two coherent beams from ruby laser monopulse energy ($W = 0,5 \text{ J/cm}^2$, duration $\Delta t = 80 \text{ ns}$) interfered in

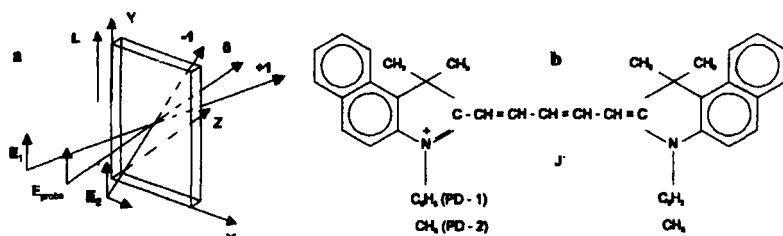


FIGURE 1 Geometry of the interaction (a) and chemical structures of dyes PD-1 and PD-2 (b).

LC-cell in area diameter of about 2 mm. The cell thickness was 50 μm . The gratings with different period were written. The He-Ne laser probe beam diffracted on a grating and first-order diffraction dynamics was registered with an oscilloscope. Both homeotropic and homogeneous LC-layers obtained by the different orientational coatings (silicon monoxide, rubbed films of polyvinyl alcohol and polypyromellitimide) were studied. The monoxide coating was obtained by vacuum thermal deposition on a glass substrate at the tilt angle 92° . The mixtures of azoxybenzenes with 4-cyanophenyl-4'-hexylbenzoate (LC-1, $T_c=65^\circ\text{C}$) and cyanobiphenyls (LC-2, $T_c=56^\circ\text{C}$) were used as LC-media in our experiments. Different dyes were tested as dopants, as a result we found that the bleachable polymethine dyes were most effective dopants. The structure formulae two of used polymethine dyes PD-1 and PD-2 are pictured in Fig. 1b. Typical absorption coefficient values at $\lambda = 694,3\text{nm}$ were in range $30\text{-}50\text{cm}^{-1}$.

2. Results

2.1 Static gratings in dye doped LCs

It was observed that static gratings were formed only in oriented by silicon monoxide homogeneous layers of DDLCs, at pump intensity of about $5 \cdot 10^6 \text{ W/cm}^2$. The view of grating is shown in Fig. 2. The investigation of specimens by electron microscope (JEOL JSM-35c) showed that writing of a static gratings at those pump intensities is not coupled with failure of orientation coating. At the increased pump intensity (about $5 \cdot 10^7 \text{ W/cm}^2$) a grating was written on the surface of the orientation coating. The view of an interference field with period of 9 μm is illustrated in Fig. 3a. The silicon monoxide coating

texture that has been formed as a result of radiation action is shown in Fig.3b. It must be noted that boundary of laser action area is clearly determined (on left side is an area that had not been irradiated). The film had a coarse-grained column-like texture that loosened itself and looked like more coarse grains the size of about 500 nm after irradiation.

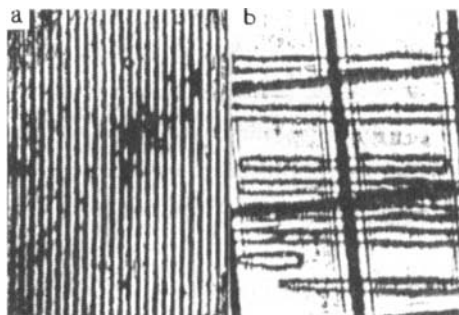


FIGURE 2 The view of grating obtained polarizing microscope with the different resolution: 500 lines/mm (a) and 200 lines/mm (b)

Let us consider now a static grating the formation of them is not caused with destroying of orientational layer. A polarizing microscope investigations showed that the gratings are formed on orientational layer surface just on the glass facing to radiation beam. The gratings are the disclination lines that are fixed at some microscopical defects caused with laser irradiation on the orientation layer surface. The disclination lines are immovable and their appearance depends on the shape of the defect (Fig.2b)^[3]. A slight pressing on glass is able to induce a wall in the layer. That wall collapses inside the layer without disclination grating destroying. A grating can be broken by the layer heating above phase transition or by applying the electric field ($>30V$). After this the layer returns in initial homogeneously

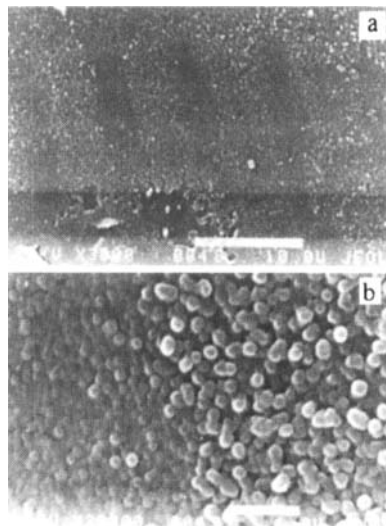


FIGURE 3 Microtexture of silicon monoxide coating induced by high-radiation: a-marker 10 μ m, b- 1 μ m

oriented state and the gratings can be rewritten again.

One of the causes of defects arising is an impurities formation due to dye photodestruction. The polymethyne dyes are the least photostable among the investigated bleachable dyes. On other hand this dyes are the ionic compounds and they can form the complexes with LCs molecules under irradiation. The defects precipitate on orientation layer surface and can exist for a long time.

2.2. Reorientation in homogeneously oriented dye-doped LC-layers

A static gratings formation is merely the consequence of more complex physical mechanism, that develops during LC-layer excitation. We studied the dynamics of this process with respect to the relaxation times. In Fig.4 the typical oscilloscope traces of diffracted in the first order probe beam at the pump intensities about 10^6W/cm^2 are shown. When a power laser radiation interacts with a bleachable dye-doped LCs one can observe both the traditional phenomena as well-known absorption coefficient saturation, density change, thermal diffusion and the orientational phenomena, that are typical for LCs [4-6] and for dye doped LCs [7-9].

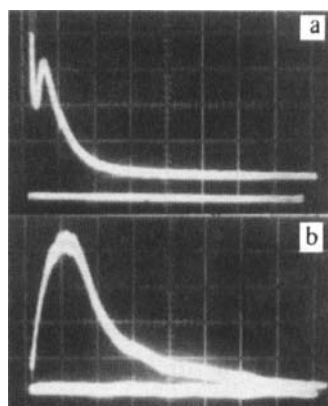


FIGURE 4 Oscilloscope trace of density-temperature (a) (1V/div) and reorientation effects (b) (0.2V/div) for PD-2 in LC-1. $\mathbf{E}_1 \parallel \mathbf{E}_2 \parallel \mathbf{E}_{\text{probe}}$

Their characteristic times are related to the dye molecules excited state lifetime and also to the time constants of changes in the density $\tau_\rho = \rho_0 \Lambda^2 / \pi^2 \eta$ (the time of the establishment of hydrodynamic velocity), in the temperature $\tau_T = \rho_0 C_p^2 \Lambda^2 / 4\pi^2 \lambda_T$ and in the reorientation $\tau_\theta = \gamma \Lambda^2 / 4\pi^2 K$. Here the LCs typical values are: $\rho_0 = 10^3 \text{kg} \cdot \text{m}^{-3}$, $\eta = 7 \cdot 10^{-2} \text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$, $C_p = 1500 \text{J/K} \cdot \text{kg}$, $\lambda_T = 0,16 \text{J/K} \cdot \text{s} \cdot \text{m}$, $\gamma = 0,1$ Poise, $K = 10^{-6}$ dyne. If the period of grating is $70 \mu\text{m}$, these time constants values are $\tau_\rho \sim 3,5 \mu\text{s}$, $\tau_T \sim 1 \text{ms}$ and $\tau_\theta \sim 100 \text{ms}$.

Relaxation processes that have the time constants τ_T and τ_θ are well distinguished in Fig.4, where at the time-base $100 \mu\text{s/div}$ (a) a dynamics of

change in the refraction index due to density oscillation in the form of spike on the thermal component of decay is shown. The time of thermal diffusion is about 500 μ s. At the long time-base (10ms/div (b)) the pronounced orientational effects is observed, that develops for 15ms and disappears in about 100ms. Its diffraction efficiency I_1/I_0 reaches three percent.

The following rules for the orientational effect have been established. The effect is observed at an any angle of director rotation in azimuth plane xy (Fig. 1) when the electric field vectors of pumps E_1 , E_2 and E_{probe} are parallel (Fig. 5a,b). It also is observed at mutually perpendicular orientation of E_1 and

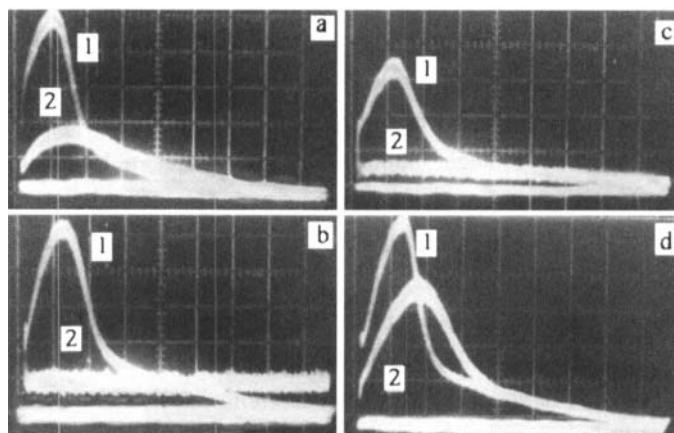


FIGURE 5 Dynamics of reorientation gratings for PD-1 in LC-2 at different geometry: $E_1 \parallel E_2 \parallel E_{probe} \parallel L$ (a,c,d-1), $\perp L$ (a-2), $45^\circ L$ (b); $E_1 \perp E_2$ (d-2); (b,c-2)- thermal component. Oscilloscope amplifier scale 0.2V/div.

E_2 , it is illustrated in Fig. 5c. A presence of the polarization gratings points to existing of the layer director rotation in the plane xy. Also, the orientational effect is observed in the layers at the low dye concentration (on the order of 10^{-4} mol/l) when the thermal component is small. In Fig. 5b,c are shown two components: orientational one (1) (at a time-base of 10ms/div) and thermal one (2) (at a time-base of 100 μ s/div). An amplification is the same in a both cases.

The data obtained as a result of investigation of the orientational effect dynamics in connection with polarization-microscope studies allow to represent the mechanism of gratings formation in a following fashion. At the first place

the medium density changes induced by high-power radiation are observed. The time period that is needed in order to a stationary distribution of the density in the area of interference band had established will be $\tau = \lambda/2 \cdot v$. Since the sound velocity in LC $v = 1540 \text{ m} \cdot \text{s}^{-1}$ we have $\tau = 20 \text{ ns}$, it is less than an exciting pulse duration. Consequently we can observe a displacement of the molecules and disturbance of continuity of director in a layer as a result of striction. If a solution is absorptonable there is also an additional contribution that is due to laser heating.

As a result of an abrupt action of the interference field in the layer areas having different orientation are induced. At the boundary between the irradiated area and non-irradiated one an uniform orientation of the director is disturbed and as a result an advent of wall takes a place. This wall has the appearance of a loop and it is well seen around the area that arised under action of uniform spot of a single pump beam. After this a loop shrinks and disappears. It can be said, that the observed pattern is a grating of "domains" i.e. of the areas that have the other orientation in comparison with parent one and are surrounded with a walls.

The existing two of orientations in a layer is energy advantageous and metastable uniform orientation inside the loop will be established in the first place (its duration is about 15ms). Then the domains shrink and almost soon after (in 100ms or so) of their disappearance the stable homogeneous orientation restores itself. Also, if radiation power is sufficient to form the defects (at a power of $5 \cdot 10^6 \text{ W/cm}^2$) then the shrinking wall breaks on surface and thus forms the disclination^[11]. This is a way of static gratings forming. It should be noted that gratings are written more effectively in the newly made samples. It is known that thermodynamic stable state of molecules orientation in a layer can be reached only after multiple reheating of cell to isotropic state. An energy of interaction of LC molecules with out of square deposited film surface is such that immediately after the cell filling two orientations are often observed, one of them is stable homogeneous and the other one is metastable. They are separated from each other with the walls. This texture was observed in Ref.^[10]. Metastable orientation is viewed with a polarizing microscope like the homogeneous one. Its intensity differs merely slightly in comparison with

intensity of the stable orientation. It is known that at a glancing angle of incidence ($>80^\circ$) one can observe an appearing of the structures that have a slight tilt of director in plane yz. This is observed in our case too.

Unlike other dye solutions in LCs such strong orientational effect in the solutions of polymethyne dyes is apparently due to their ionic structure that favours the formation of metastable domains. The polymethyne dyes in solutions of polar solvents dissociate into organic cationes and aniones J. As a result of motion of J light-weight at inducing of metastable orientation the field of spatial charge can appear, that favours reorientation of LC molecules. The observation of photoinduced conductivity in solutions of laser dyes in 5CB was reported in ^[12,13]. The existence of conductivity was detected in the presence of weak electric field. In favour of such orientational mechanism one can adduce the fact, that molecular reorientation is observed at different interaction geometries and at weak absorptions smaller than 10^{-4} mol/l. At such dye concentrations authors ^[12] observed photorefractive orientation effect.

CONCLUSION

We investigated the orientational phenomena in planar dye doped LC-layers oriented by film of silicon monoxide. The reorientation in both the azimuthal plane and plane perpendicular to substrates was revealed. In the gratings with period of 70μ during ~ 15 ms the metastable orientation was established itself and relaxed with time of 100ms. We suppose that in solutions of ionic dyes in the polar LC the orientational effect is enhanced with the spatial change field, that appears under action of powerful laser radiation.

The revealed mechanism of orientational nonlinearity requires the more detailed consideration of interaction of orienting surfaces, walls in the bulk of LC and molecular reorientation of layer.

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