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Dynamics of Light-Induced Reorientation in Planar Cell Filled with the Azo-Dye Doped Nematic Liquid Crystal

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There have been intense research interests in the field of nonlinear optical materials capable of recording permanent holographic gratings. The goals to be achieved include low recording intensity, high storage density, and fast recording/erasing times. One of the promising directions in this field is the use of cells filled with azo dye-doped nematic liquid crystals as a recording medium. The present work is devoted to the study of recording mechanisms in such objects. The processes of formation of a director of permanent orientation in a nematic liquid crystal doped with an azo dye in a cell with two different limiting surfaces under light irradiation are studied. It is shown that the fixation of the director on the surface with a weak adhesion energy, caused by the competition of the adsorption and desorption processes of the dye, depends on the polarization of the irradiating light.

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1. Introduction

The light-induced director reorientation process in planar combined cells filled with the azo-dye doped nematic LC has attracted much interest. Many articles have been published on this subject recently [1–10] but the final microscopic model of the process still is a subject of open discussion. The LC director reorientation is due to light-induced changes in anchoring conditions on the aligning surface. In the combined cells one surface has strong planar anchoring and the second provides weak isotropic anchoring. Irradiation with linearly polarized laser light from the adsorption band of Methyl Red (MR) leads to the twist structure formation in the irradiated area [11]. A new light-induced easy orientation axis, parallel to the surface due to anisotropic adsorption of MR molecules by the aligning surface. The light-induced easy orientation axis manifests itself through the LC director reorientation on the surface. At the same time, the anchoring conditions at the reference surface are retained and the twist structure is formed in the irradiated area. The light-induced anchoring is retained in the cell for a very long period (five and more years).

The described effect may be of interest both in fundamental and applied physics. The possibility to record binary images and holograms was shown in [4]. Sensitivity of the effect to the excitation light polarization enables one to record polarization gratings on perpendicular polarization of the two writing beams [5], using LC doped with the azo-dye as a high-resolution recording medium. The method of changing the configuration of the LC director due to the azo component currently remains one of the most popular and allows for the creation of unique photonic systems - complex gratings [12-16] and controlled q-plates [8, 16].

But the effect is not as simple as it seems. Recent investigations have shown that the MR molecules adsorbed on the aligning

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surface without light irradiation also played an important role [17]. The light-induced desorption of MR molecules from the "dark-adsorbed"layer results in the easy orientation axis perpendicular to the light polarization direction. Moreover, the anomalously strong orientation nonlinearity [4] and bulk director reorientation must be taken into account during investigation of the influence exerted by polarized light on the LC doped with MR. Summarizing, the dynamics of the director reorientation under the action of polarized light is complicated, being driven by the bulk director reorientation and the surface-mediated director reorientation. The bulk director reorientation is due to trans-cis conformations of MR molecules[17]. The surface-mediated reorientation is the light-induced effect due to processes of light-induced adsorption/desorption of MR molecules on the aligning surface. The final director orientation is frozen on the aligning surface and distinguished by competition of all these mechanisms. This work presents investigation into the dynamics of the director reorientation under irradiation with linearly-polarized laser light. In the process of investigation, the permanent (frozen at the surface or surface-mediated) and reversible components of the reorientation have been distinguished. The obtained results demonstrate evolution of the surface-mediated anchoring in terms of the light-induced adsorption/desorption processes of dye molecules on the substrate.

2. Experiment

The experiments were carried out using sandwich glass cells (thickness of 25-28 λm) filled with mixtures of the 5CB nematic liquid crystal and the Methyl Red azo-dye in different weight concentrations. The inner surface of the first glass (control surface) was coated by an isotropic layer of polyvinyl-cinnamate-fluoride (PVCNF), whereas the inner surface of the second glass (reference surface) was coated with polyimide layer and rubbed to provide strong

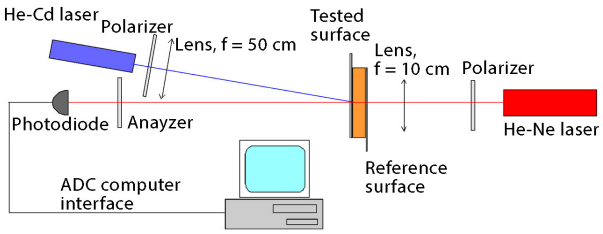

FIG. 1: Experimental setup.

planar anchoring. This surface determined the initial planar alignment in the cell.

A simple pump-probe experimental setup was used to control the LC director reorientation in the cell (see Fig. 1). The pump beam focused to the diameter about 0.25 mm was impinging the cell from the side of the control surface. The polarization direction of the pump beam formed an angle of 45° with the initial director orientation fixed by the rubbed substrate. At the same time, the test beam focused to 0.07 mm was incident at the center of the irradiated area from the side of the rubbed substrate. Polarization of the test beam was fixed parallel to the initial LC director orientation on the rubbed surface. The test beam was darkened by the analyzer that was crossed with initial test beam polarization. The LC director orientation was changing under irradiation, causing variations in the test beam polarization on propagation though the cell. The test beam intensity measured behind the analyzer was zero before irradiation and nonzero when the LC director orientation was changed. In the case under study the Maugine regime was valid and changes of the test beam polarization governed the LC director reorientation. Detailed description of this method is given in [4]. The LC director reorientation dynamics was measured for three weight concentrations (0.1%, 0.5% and 1.5%) of the Methyl Red (MR) dye in the nematic matrix of 5CB (Merk).

Let us designate the LC director reorientation outward of the pump light polarization by a negative value of the director reorientation angle, whereas positive values mean turning of the director towards the light

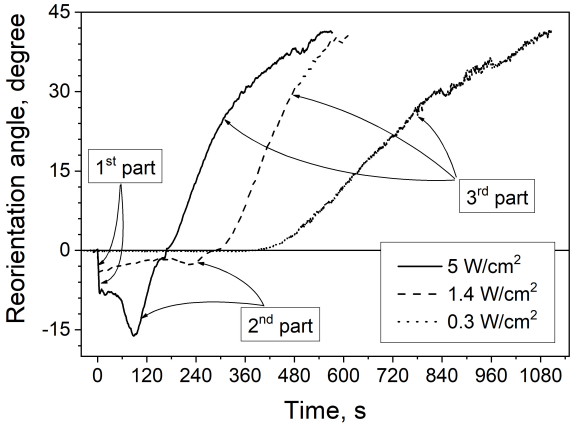

FIG. 2: The LC director reorientation dynamics for different pump intensities in the cell with MR at C=0.1%.

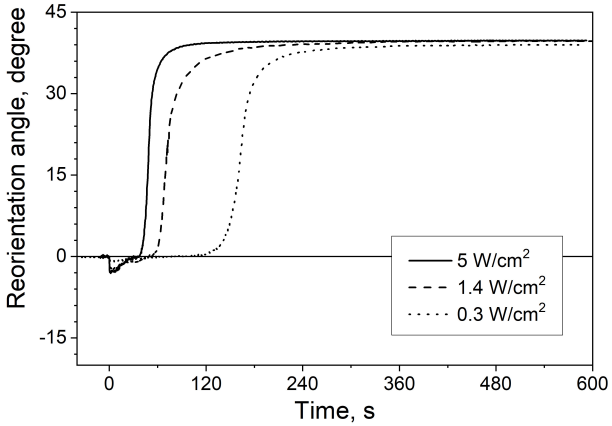

FIG. 3: The LC director reorientation dynamics for different pump intensities in the cell with MR at C=0.5%.

polarization. The initial director reorientation angle is equal to zero. If the light polarization vector makes an angle of 45° with the initial LC director orientation, then maximal possible director reorientation angles may be: 45° (when the director is parallel to polarization) and -45° (director is perpendicular to polarization).

The regular reorientation dynamics is shown in Fig. 2. It is seen that the LC director turns outward the pump light polarization right after the beginning of irradiation. A negative director reorientation is changing to the positive one after some period of time depending on the pump intensity and the dye concentration in the mixture (see Figs. 3 and 4).

The dynamics of the light-induced director reorientation for different intensities of excitation light at the dye concentration C = 0.1% is shown in Fig. 2. It is seen that the reorientation is monotonic at low pump intensity I = 0.3 W/cm^2. The LC director attains its stable position at about 40 degrees after 30 minutes of irradiation. There is no reversible reorientation in this case. Increasing of the pump intensity (see the curves for 0.4 - 5 W/cm^2) leads to the appearance of a reversible part of reorientation. The fast director turning outward the pump polarization, known as "sliding", becomes visible. Besides, the second part of the outward director reorientation is observed.

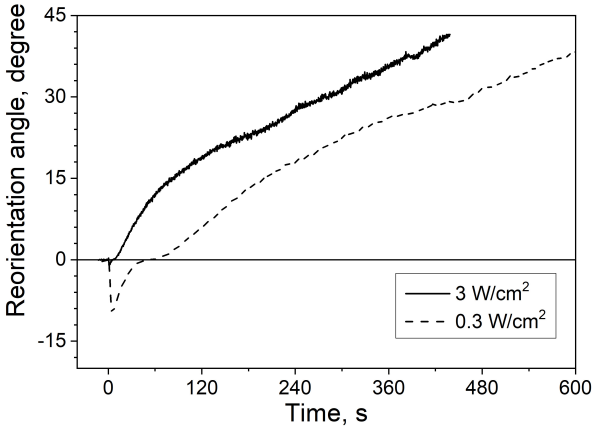

FIG. 4: he LC director reorientation dynamics for different pump intensities in the cell with MR at C=1.5%.

Then the director tends to turn towards the pump polarization. The time needed for zero-line crossing is reduced as the pump intensity increases.

As demonstrated by similar experiments at higher concentrations of the dye, with an increase in the dye concentration (see Figs. 3 and 4), the parts associated with sliding and outward reorientation are suppressed and disappear for the dye concentration C = 1.5%. The director relaxation dynamics was also measured. The

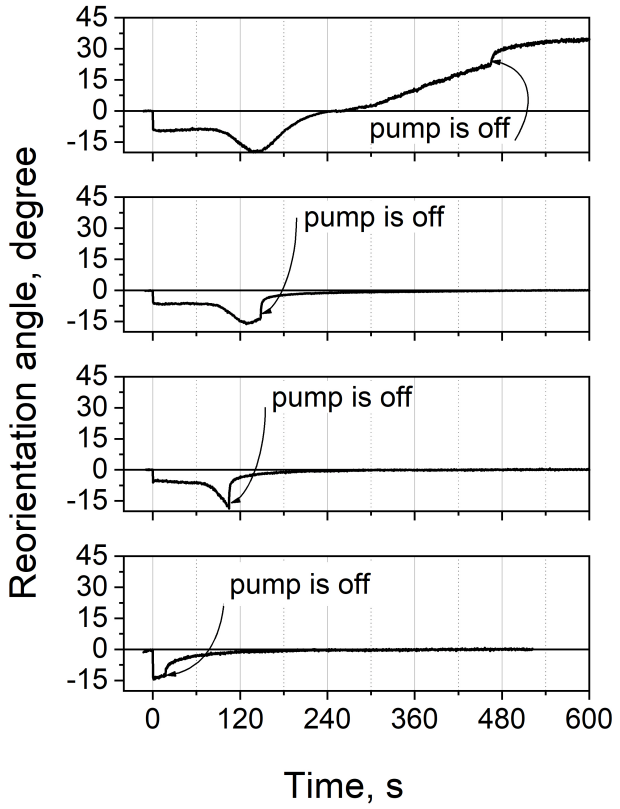

FIG. 5: The director relaxation dynamics. C=0.1%; P=1.4 W/cm^2.

result is presented in Fig. 5. The moments of switching-off the pump are indicated by arrows.

3. Discussion

The processes of sliding and stable director reorientation (parts 1 and 3 in Fig. 2) have been described in detail earlier [4]. It should be noted that three processes are observed in the cell on irradiation. Each of the processes affects the LC director. Just competition of these three processes defines the director orientation due to irradiation.

The light-induced adsorption of MR molecules seems to be responsible for the appearance of the easy orientation axis parallel to polarization[11]. This assumption has been confirmed in [4].

The authors of [18] have presented the evidence that the light-induced desorption was responsible for the appearance of the easy orientation axis perpendicular to light polarization. The experiments in the isotropic phase have demonstrated the director reorientation outward of polarization in the irradiated area after slow cooling of the sample down to the nematic phase. Also, the authors have suggested "dark"(not light-induced) adsorption of MR molecules on the surface, with the formation of a dark-adsorbed layer of the dye molecules at the surface.

Based on the results of these works and taking into account the bulk director reorientation, we can describe the dynamics of the light-induced director reorientation in the nematic phase and try to make clear the situation with competition of the processes leading to the director reorientation. Right after filling the cell, the molecules of MR are adsorbed on the tested isotropic surface without light irradiation ("dark"adsorption). Just finite number of MR molecules may be adsorbed on the substrate due to dark-adsorption. The number of MR molecules in a dark-adsorbed layer is determined by the properties of the surface.

The MR molecules, located near the tested surface absorb light and are adsorbed on the surface. The MR molecules located in the dark-adsorbed layer are desorbing from the surface when they absorb light. The number of the MR molecules located in the vicinity of the surface depends on the dye concentration in the mixture. Diffusion of the MR molecules is strong enough to keep the number of bulk molecules constant in the irradiated area.

Let's consider step by step the competition between the mechanisms producing the light-induced director reorientation. The bulk director reorientation is taking place when the anchoring energy at the boundary surface is low. The bulk director reorientation is revealed as sliding (first part of the curve in Fig. 2). The director reorientation above the tested surface

without anchoring corresponds to fast trans-cis conformations of the light-absorbing MR molecules. To reduce free energy of the system, the MR molecules reorient in the direction perpendicular to light polarization. Collective rotation of MR molecules turns the matrix due to the host-guest effect [19, 20]. That is why the LC director quickly turns the outward polarization, while anchoring provided by the substrate is weak. The LC director sliding is completely reversible (see Fig. 5). The second and the third parts of the director reorientation curve correspond to permanent anchoring on the previously isotropic anchoring-free surface.

Under irradiation, both the light-induced adsorption and desorption of MR molecules produce two easy orientation axes on the surface. The MR molecules being in the dark-adsorbed layer may leave the layer when they absorb light. Since light absorption is more probable for the MR molecules parallel to polarization, the molecules oriented along the polarization direction leave the dark-adsorbed layer. As a result, the dark-adsorbed layer becomes anisotropic. This anisotropy has the form of a distribution function with a maximum perpendicular to the polarization. Therefore, the easy axis perpendicular to light polarization is growing. Since the number of molecules in the dark-adsorbed layer is finite and running out due to desorption, the anchoring energy corresponding to the desorption-mediated easy orientation grows and decreases.

At the same time, the light-induced adsorption of MR molecules on the surface produces the easy orientation axis parallel to polarization. The MR molecules located within the cell, close to the surface may absorb light and get adsorbed on the surface. The light-induced adsorption leads to strong interaction between the MR molecules and the surface. In this case the molecules subjected to light-induced adsorption have no possibility to leave the surface due to light-induced desorption. Light absorption is more preferable for MR molecules which are parallel to the light polarization direction. The light-induced adsorption results in the easy orientation that is parallel to the light polarization. Since the cell's bulk is almost an infinite reservoir of MR molecules and diffusion is quick enough, the anchoring energy of the adsorption-mediated easy orientation axis is increased. It is interesting to consider the cases with different dye concentrations and different light intensities. When a concentration of MR in the bulk is low (0.1%, see Fig. 2), the light-induced adsorption produces the easy orientation axis slowly and one can observe all the regions on the reorientation dynamics curve. If the MR concentration is higher, the adsorption-mediated easy axis grows faster, reducing the time for observation of the outward director reorientation. When C = 1.5%, the adsorption-mediated easy axis grows faster than the desorption-mediated. There are no manifestations of the outward director reorientation and only sliding can be observed.

In conclusion, the processes of bulk reorientation and light-induced adsorption/desorption of MR molecules govern the light-induced director reorientation in MR-doped nematic LC. The bulk reorientation produces the reversible director sliding above the tested surface. When the tested surface receives anchoring due to the adsorption/desorption processes, the vector sum of the corresponding easy axes determines the director orientation. Desorption is a finite process, whereas adsorption is almost infinite. Desorption depends on the surface properties and adsorption – on the dye concentration in the mixture. Both processes depend on the pump light intensity. It should be noted that diffusion processes in the studied cells are also affected by the presence of a photovoltaic field and may influence the dynamics of the processes [20–24]. We believe that the LC director reorientation can be described by dynamic equations.

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