

Study on laser-induced ripple structures in dye-doped liquid crystal films in high-intensity regime

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The photoalignment effect induced by an Ar⁺ linearly polarized laser beam on a dye-doped nematic liquid crystal in a pump-probe experiment is investigated. A symmetric capillary cell with two polymer coated glasses was used. An easy axis was induced by rubbing both surfaces in the same direction. The exciting Ar⁺ laser beam has the polarization direction perpendicular to the easy axis. The time dependent transmitted intensity of the probe He-Ne laser beam (polarized parallel to the easy axis) has been measured for various power of Ar⁺ laser in high-intensity regime and the beginning time of the ripple structure formation has been estimated. The spacing and the depth of the ripple structure were measured by using atomic force microscopy.

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1. Introduction

The alignment surface of the liquid crystal (LC) molecules is very important in liquid crystal displays. Conventionally, the alignment of LC's is performed by rubbing a polyimide/polymer film coated on glass plates. This contact alignment method causes some dust particles and electrostatic charges on the rubbed surface. Techniques of surface-assisted photoalignment may solve these problems. The photoalignment can be achieved by doping azo dyes into LCs. Lucchetti *et al.* [1] and Fuh and co-workers [2] observed the dye-induced LC reorientation on the surface with and without polymer coated, respectively. Methyl-red- (MR-) doped LC's represent a very interesting system for studying photoalignment. Under proper irradiation with the blue-green light, the MR molecules tend to be adsorbed onto the surface of the substrate, reorienting the LC molecules into a new permanent LC configuration [3]. The surface morphology of the adsorbed substrate has been studied by Lee *et al.* [4] and three different morphologies have been observed – a homogeneous and fine layer of adsorbed dyes, a layer with microgrooves, and an inhomogeneous ribbon like and rough adsorbed layer. The first and second types of layers dominate, in the early and late stages, respectively, in the weak-intensity regime, tending to cause the LC's to reorient perpendicular and parallel to the polarization direction of the pump beam. The last type of layer dominates in the strong intensity regime, possibly severely disturbing the orientation of the LC's.

The aim of our study is to investigate the photoalignment effect induced by an Ar⁺ linearly polarized laser beam on a sandwich glass cell filled with 4'-n-pentyl-4-cyanobiphenyl (5CB) doped with MR, by a pump-probe experiment. The irradiation of the sample has

been performed for a very short time (9 minutes) but at a high-intensity regime of the pump beam. The polarization direction of the He-Ne probe beam was parallel with the easy axis induced by unidirectional rubbing of the two polymeric film coated on the glass plates. The photoalignment effect of the pump Ar⁺ laser beam was studied for various powers of the pump laser and for polarization direction of pump beam perpendicular to the easy axis. The evolution of the transmitted intensity of the probe beam during irradiation has been explained taking into account the photoalignment effect. The images of the sample in the pumped areas obtained by using a polarizing optical microscope (POM) confirm the results of the pump-probe experiment. We studied the irradiated zones by using an atomic force microscope (AFM) and we observed that the surface morphology depends on power of Ar⁺ laser beam. The spacing and the depth of the laser-induced ripple structure were also measured and the azimuthal anchoring energy has been evaluated.

2. Experiment

The experiments were realized at a temperature of 25 °C by using a standard sandwich glass cell filled with a nematic liquid crystal 4'-n-pentyl-4-cyanobiphenyl (5CB) doped with MR (1.5% by weight). Their structural formulas are presented in Fig. 1.

Azo dyes are usually in the stable *trans* state in the dark. The absorption band of MR dyes is in the blue-green region [4]. When excited by blue-green light, MR is transformed from the *trans* to *cis* form, causing the absorption band to shift to a region with a longer wavelength (red or infrared). This means that the *cis*- form MR can absorb red light.

The thickness of the cell was 23 μm (obtained by Mylar spacers). The inner surfaces of the glass plates were previously covered with a thin polyvinyl alcohol film by spin coating method. The monomer, polyvinyl alcohol (PVA) was dissolved in H_2O at a concentration of 1.5% wt.

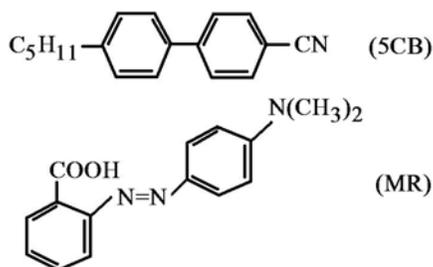


Fig. 1. The structures of 5CB and MR in trans form.

The PVA solution was spin coated on the glass plates, then baked at 120°C for 60 minutes and then cool at room temperature. An easy axis was induced by rubbing both surfaces in the same direction that impose a planar alignment through the whole cell. The optical arrangement used in the pump-probe experiment is shown in Fig. 2. Our experimental set-up is an improvement version of that described in Ref. 5.

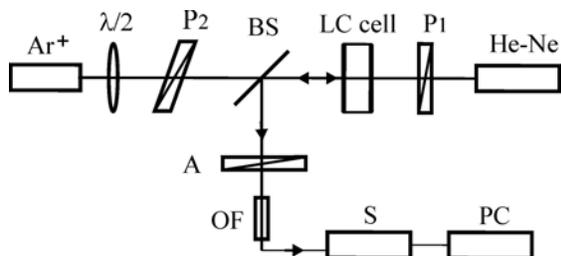


Fig. 2. The experimental set-up.

The cell was placed normally to the exciting beam of Ar^+ laser ($\lambda=476.5$ nm; power P_{exc} was varied between 5 mW and 30 mW; the irradiation time being fixed at a value of 9 minutes). This is a very short time as compared with 3 hours, the irradiation time in the study performed by C. R. Lee *et al.* [4].

In general, with a laser pump beam traveling across a liquid crystal cell the induced thermal gradient can create a fluid flow that in turn will reorient the molecules through a flow-alignment mechanism [6]. To prevent this effect from occurring we used a large pump beam diameter. It is worth noting that thermal effects can be excluded in our geometry they cannot contribute to the observed signal.

The diameter of the Ar^+ laser spot on the cell was ~ 3 mm and the mean intensity of the pump beam in the irradiates areas has values between 160 mW/cm^2 and 960 mW/cm^2 (corresponding to 5 mW and 30 mW, respectively), that are characteristic for a high-intensity regime. The polarization direction of the excited beam was horizontal, being established by using a half wave plate

($\lambda/2$) and a polarizer P_2 . The spot of the probe He-Ne laser beam ($\lambda=633$ nm; power $P_{probe} \sim 1$ mW) was covered by that of the pump light. The polarization direction of the He-Ne laser is vertical and is parallel with the rubbing direction. After having passed through the cell, the probe beam was deviated by a beam-splitter (BS) and went through the analyzer (A) of which transmission direction was horizontal. In these conditions the appearance of probe He-Ne light behind the analyzer indicated a change of the liquid crystals orientation.

The probe beam intensity measurement was performed by an optical fiber (OF) connected with an "Ocean Optics" spectrometer S2000 (S) and a computer (PC). The intensity of the transmitted probe beam has been recorded during irradiation of the cell, in different zones, at various pump power (5, 12, 25, and 30 mW). After irradiation, the induced permanent structure in the sample has been studied by using a POM. In order to study the laser-induced surface morphology, the sample has been submerged into hexane, for a few minutes, until 5CB is dissolved. The glass plate of the cell that has placed to the He-Ne laser during irradiation is transparent and this fact demonstrates that the MR molecules have not been adsorbed on this surface. On the other glass plate appear red zones which coincide with the pumped regions. The presence of these red zones demonstrates that some MR molecules are adsorbed on PVA within the irradiation of the sample. The morphology of the pumped regions has been analyzed by using AFM.

3. Results and discussion

The intensity of the transmitted probe beam as a function of the irradiation time for different values of the exciting laser power is presented in Fig. 3.

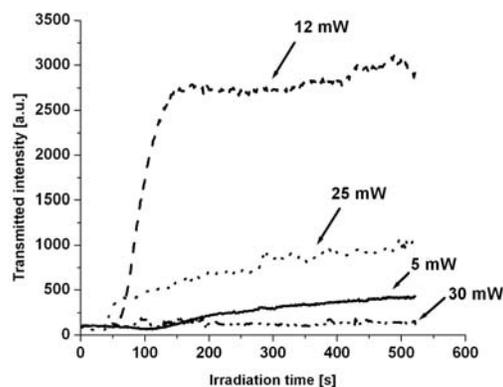


Fig. 3. Transmitted intensity versus irradiation time.

We remark that the transmitted intensity begins to increase after an irradiation time τ_b . We found that τ_b decreases if the power increases, the values of τ_b being 102, 51, and 39 s for $P_{exc}=5, 12$ and 25 mW, respectively. The transmitted intensity can increase during irradiation if an easy axis is induced on PVA of the glass plate placed to

the Ar^+ laser and the angle between this gliding induced easy axis and the vertical direction increase too.

The orientation of the ripple structure must be parallel with the polarization direction of the pump beam [4] and, consequently, a twisted nematic structure is generated in the pumped region. The time τ_b corresponds to the beginning time of the ripple structures formation.

Such ripple structures are believed to be produced by interference between the incident polarized Ar^+ light and waves scattered from the dye-absorbed surface with microscopic roughness. This yields a spatially periodic intensity distribution, causing a corresponding variation in the absorption of MR on the inner left surface of the cell.

For $P_{exc}=12\text{mW}$ the transmitted intensity is greater than that for 5mW due to the fact that the ripple structure growth more rapidly at a high intensity of pump laser beam. At a power of 25mW the transmitted intensity is less than that for 12mW due to the fact that at a higher pump power the concentrations of the MR molecules in the cis-isomer form increases and the absorbance of MR in the red light region significantly increases [7]. At $P_{exc}=30\text{mW}$ the cis-isomer concentration grow quickly and the probability that the dye aggregates significantly increases. These aggregates are adsorbed on microgrooves to form ribbon-like adsorbents and have a poor capacity to align LC molecules in the direction parallel to the polarization direction of pump beam.

The permanent nematic structure obtained in the irradiated zones has been analyzed by using a polarizing optical microscope with crossed polarizers. Figure 4 show the images of these regions. The irradiated regions are bright while the unpumped zones are dark. Because the Ar^+ laser beam is gaussian and its intensity decreases from the center to the periphery of the laser spot, the effect of irradiation is more pronounced in the center of the pumped zone.

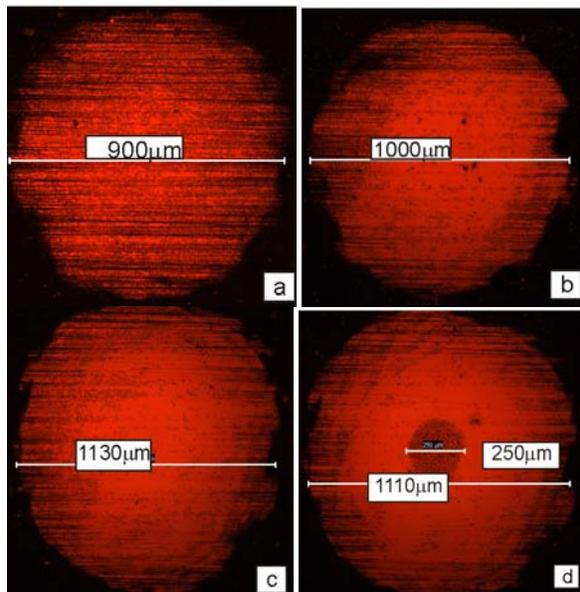


Fig. 4. Images of the irradiated zones obtained by using a POM with crossed polarizers: (a) $P_{exc}=5\text{ mW}$; (b) $P_{exc}=12\text{ mW}$; (c) $P_{exc}=25\text{ mW}$; (d) $P_{exc}=30\text{ mW}$.

The bright region demonstrates that a twisted nematic structure has been induced during irradiation. The twisted angle has been measured in the center of the zones corresponding to the pump power of 5, 12, and 25 mW and the obtained values are 65° , 75° and 83° , respectively.

For the power of 30mW a twisted angle can not be measured in the center of the irradiated zone because appear dye aggregates (Fig. 4 (d)).

The AFM images corresponding to the central and non-central region of the irradiated zones are presented in Fig. 5. A ripple structure is observed for pump power of 5, 12, and 25mW . The orientation of the microgrooves in the ripple structure is parallel with the direction of polarization of Ar^+ laser beam. The mean depth of the microgrooves, A , obtained for 5, 12, and 25mW are ~ 16 , 19 and 45nm , respectively. We observe that the depth of the ripple structure increases if the pump power is increased. The spacing between microgrooves, Λ , is $\sim 300\text{ nm}$ (see Fig. 5 (b)). According to the laser-induced periodic surface structure theory [8], the spacing of the resulted ripple structure in a dye-doped liquid crystal film is given by $\Lambda = \lambda/n$, where λ is the wavelength of the pump beam in vacuum and n is the refractive index of the material adsorbed on the surface of the dye-doped liquid crystal cell substrate. For $\lambda=476.5\text{ nm}$ and $n \cong 1.6$ (for MR) [3] we calculate a spacing of $\sim 298\text{nm}$, which is consistent with the experimental one.

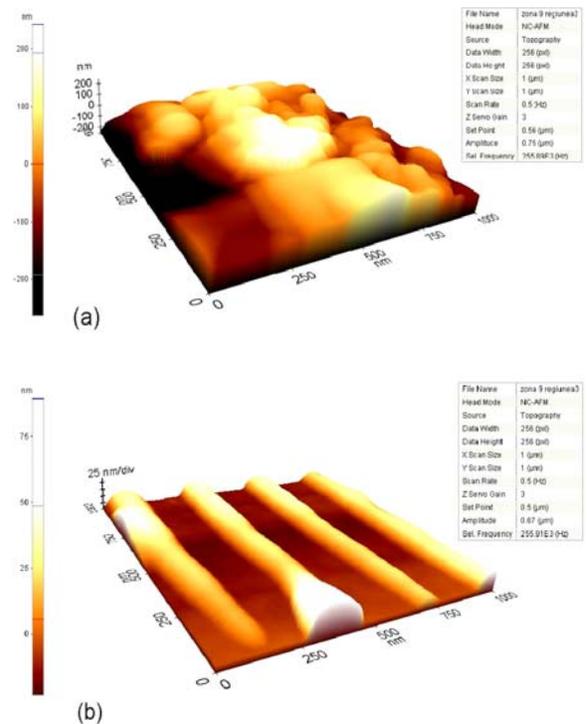


Fig. 5. Three-dimensional AFM images obtained for: (a) central zone irradiated with 30 mW , and (b) a non-central zone irradiated with 25 mW . The microgrooves in (b) are parallel to the pump-beam polarization. The mean depth of the microgrooves is $A \sim 45\text{ nm}$ and the spacing $\Lambda \sim 300\text{ nm}$.

For the pump power of 30mW, in the central zone, is obtained a rough and inhomogeneous surfaces by rapid and random aggregation and adsorption of cis MR molecules Fig. 5(a)).

Important information about the roughness of the scanned area is given by the maximum “peak-to-valley” value, R_{pv} [9]. For 30 mW we obtained the value $R_{pv}=807.54$ nm.

4. Conclusions

In this study, we investigated the photoalignment effect induced by an Ar^+ linearly polarized laser beam on a dye-doped liquid crystal cell in a pump-probe experiment. We recorded the transmitted intensity of the probe beam during irradiation and we observe that it depends on the power of the pump beam. We explained evolution of the transmitted intensity and we evaluated the start time of the ripple structure formation. The induced permanent twist nematic structure in the irradiated zones has been investigated by POM and we conclude that bulk reorientation and surface adsorption are involved in the photoalignment process.

The laser-induced surface morphology in the irradiated regions was studied by AFM. For pump power of 30 mW a rough surface with inhomogeneous ribbon-like has been obtained. A ripple structure parallel with the polarization direction of the pump beam was generated for pump power less than 30 mW. The spacing between microgrooves is not dependent on the power of Ar^+ laser beam but the depth of the ripple structure increases if the power pump is increased. We find that this structure can induce a surface-assisted photoalignment effect.

Our study demonstrates the possibility to control the depth of the ripple structure and, consequently, the induced anchoring energy by a proper choice of the power of the pump beam. Further studies are needed in order to deduce a relationship between pump power, irradiation time and induced anchoring energy

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