# Polarization controllable laser-induced ripple structure in dye-doped liquid crystal films

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## ABSTRACT

The laser-induced ripple structure of dye-doped liquid crystals has been investigated with the various polarization of pump beam. The process of laser-induced ripple structure involves two stages, the dye adsorption on substrate and the ripple structure formation. According to the dynamic measurement, the maximum intensity of transmission was due to the twisted nematic configuration induced by homogeneous dye adsorption on the command surface and the decay of transmission was due to the gradual formation of ripple structure. From the observations of atomic force microscopy (AFM), the directions of ripple structure were formed parallel to the polarization of linear polarized pump beam or the elliptical long axis of elliptically polarized pump beam.

***Keywords***: ripple structure, dye-doped liquid crystal, polarization controllable

## 1 INTRODUCTION

Azo-dye-doped liquid crystals have drawn considerable attentions for a long time because of their potential applications for optic devices. It is well known that the photoisomerization of dye molecules can be induced from trans- to cis-configurations by the visible light . Gibbons et al. [1] and Chen et al. [2] observed that a negative torque can be generated to align liquid crystal molecules perpendicular to the optical field by the photo-excited azo dye on DDLC films. The additional photo-induced alignment effect caused by the unidirectional microgroove has been reported. The microgroove can be produced by reactive-ion etching [3], photolithography [4], laser ablation [5] and alignment of the LCs along the groove direction. These phenomena were described by Berreman [6].

Another photoalignment approach, which involves the laser-induced ripple structure on the adsorbed dye molecules in DDLCs has also been reported [7~10]. Under proper irradiation on DDLC film by blue-green light, the dye molecules tend to be adsorbed onto the surface of the substrate and reorient the LC molecules into a new aligned configuration. As the light successively irradiates for a long period, the ripple structure were grown gradually and the liquid crystals were reoriented parallel to the ripple structure due to the anchoring force of the microgroove.

In this study, we report the formation of ripple structure under various polarization state of pump beam, including linear and elliptic polarization which possess different azimuthal angle with respect to the rubbing direction. The direction of ripple structure depends on the polarization state of pump beam. According to the results measured by atomic force microscope (AFM), the directions of ripple structures could be formed parallel to the direction of linear polarized light or the major axis of elliptically polarized light.

## 2 EXPERIMENT

Figure 1 shows the experimental setup for the generation and the dynamic measurement of the formation of ripple structure. The experiment was performed with sandwiched glass cell which was filled with a mixture of the liquid crystals (E7) and azo dye of methyl red (MR). The weight concentration of dopant is about 1.5%. MR is a well known photosensitive material which optical absorption lies in the wavelength range between blue and green. The dichroic ratio of MR is approximately six in the wavelength range between blue and green. The sandwiched glass cell was separated by two 12-μm-thick plastic spacers. The reference surface ($S_R$) of glass substrate was covered with the rubbed alignment film of polyimide and the command surface (Sc) of glass substrate was coated by the polymer film of NOA81 without rubbing treatment. After the sample was pumped for the specific period, the substrate of the cell was removed carefully. Then the substrate of Sc was submerged into solvent of hexane for a few second to dissolve the LCs. The laser-induced ripple structure on Sc was carefully elucidating using AFM.

The transmission axis of the polarizer (P), which was placed in front of the probe beam He-Ne laser ($\lambda_R$ = 632.8 nm), is in the y direction and the transmission axis of the analyzer is in the z direction. The initial dark state was obtained under the condition of the crossed polarizers. The energy and the polarization of the pump beam (DPSS laser, $\lambda_G$ = 532 nm) were tuned by both half wave plates and the linear polarizer. The pump beam was impinged normally onto Sc and the diameter of spot was about 5 mm.

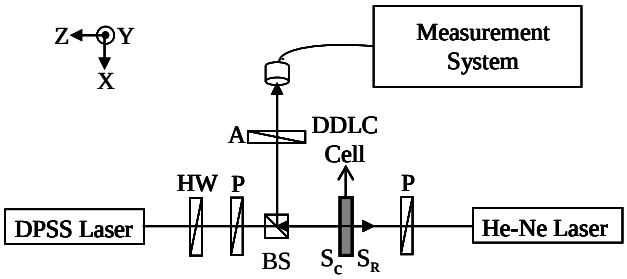


Figure 1 : Experimental setup for generating ripple structures in DDLCs and the measurement of dye adsorbed process. (A: analyzer, P: polarizer, BS: beam splitter, D: detector, HW: half wave plate, $S_c$: command surface, $S_R$: reference surface)

The probe beam was impinged normally onto $S_R$ and the spot was covered by that of the pump beam. The power of the probe beam was adjusted less then 1mW to avoid the absorption of the dye molecules which was in the cis state. After the probe beam passed through the DDLC sample, the beam was divided by beam splitter and the signal was incident into the detector.

This study utilizes one polarized pump beam to excite the DDLC sample. When the pump beam was impinged onto the sample, the azobenzene molecules absorbed the energy of pump beam and were excited to the unstable cis state. The dipole moment of methyl red in the cis state is stronger than that in the stable trans state. Consequently, the dye molecules in the cis state will induce a dipole moment of the adjacent polymer molecules. By the hydrogen bound intermolecular interaction between dye and polymer molecules, the methyl red molecules were adsorbed onto the coated surface [11]. When DDLC sample is continuously excited by the pump beam, the azobenzene molecules will go through many trans-cis-trans isomerization cycles and finally align perpendicular to the polarization of pump beam. Morever, the adsorbed azobenzene molecules will reorient the adjacent LC molecules into a direction perpendicular to the polarization of pump beam.

## 3 RESULTS AND DISCUSSIONS

### 3.1 Linear Polarization

Figure 2 presents the dynamic measurement results of the dye adsorbed process, which were excited by one linear-polarized (parallel to rubbing direction) pump beam with various intensities. After the polarized pump beam was turned on (t=0), the MR molecules were excited and gradually adsorbed onto substrate Sc. The rise of transmitted intensity is due to the anisotropic adsorption of MR molecules on Sc which cause the adjacent LCs to be reoriented form rubbing direction toward the direction

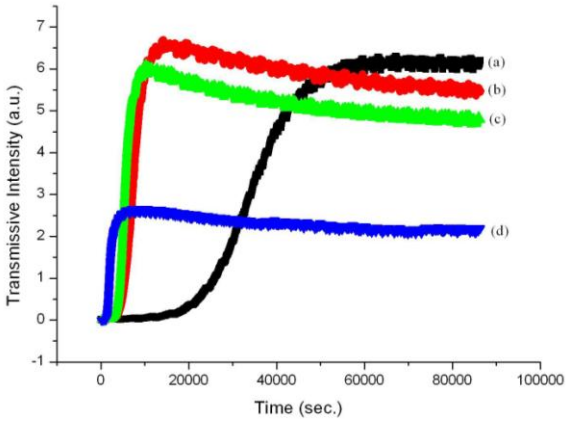


Figure 2 : The dynamic measurement of dye adsorbed process onto Sc.The polarization of pump beam was parallel to the rubbing direction with pump intensity (a) 1 (b) 3 (c) 5 (d) 7 mW/cm$^2$.

perpendicular to the polarization of pump beam. With the increase of irradiation time, the amount of adsorbed dye molecules were increase and the TN structure was obtained which leads to a maximum of transmitted intensity.

Figure 3 shows the polarized optical microscopy (POM) images of DDLC sample irratiated by pump beam with intensity 1 mW/cm$^2$ for pumping duration 24 hours. The polarizer is set parallel to the rubbing direction and the light source was impinged onto the sample from Sc. The pump region shows bright and dark under a set of cross and parallel polarizers, respectively. The images suggest that the TN structure was obtained within the pumped region. The azimuth surface anchoring energy from the adsorbed dyes on Sc can be written as [12] $W_1=2K_{22}\varphi/d \sin\varphi$, where $\varphi$ is the angle of twist, d is the cell gap, and $K_{22}$ is the twist elastic constant. The twisted angle of DDLC sample, which was irradiated by pump beam with intensity 1 mW/cm$^2$ for 24 hrs, was measured about 86°. According to figure 2, the maximum transmitted intensity was decreased with stronger pump beam as well as the azimuthal angle of LCs. Under irradiation by strong pump beam [curve (c) and (d)], the amounts of excited dye molecules were increased to excess in the DDLC sample. It is known that the dipole moment of

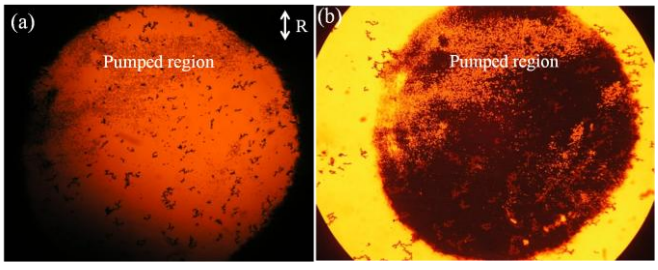


Figure 3 : The POM images of the pumped region under (a) cross and (b) parallel polarizers, respectively. The polarizer was set parallel to the rubbing direction R.

MR at cis-isomer is much stronger than that of trans-isomer [11], so that the dye molecules were rapidly aggregated in bulk and were then adsorbed onto Sc irregularly.

Figure 4 shows the irregularly adsorbent structure on Sc which generated by strong pump beam. Such structure causes the LCs to become reoriented randomly and this phenomenon leads to the scattering of the probe beam. Under this situation, the rapidly aggregation of adsorbent layer causes the probe intensity decrease before the LCs formed 90° TN structure. Curve (a) and (b) showed the adsorbed process under weak excitation. For curve (a), the exciting intensity is too weak to generate ripple structure within the pumping period. For curve (b), the near 90° TN structure were gradually formed due to the anisotropic dye adsorption onto Sc before t = 1.5hr. With the increase of irradiation time, the ripple structure was gradually formed and the depth of the structure was also increased. This structure was induce an anchoring energy which causes the LCs to reoriented parallel to the rubbing direction from TN alignment and cause the decay of probe intensity. According to Berreman's groove theory [14], the anchoring energy of the structure was proportional to the third power of depth of the microgroove so that the probe signal was gradually decay with the increase of irradiation time.

The formation of ripple structure can be observed by the atomic force microscopy. Figure 5(a) showed the resultant ripple structure formed by polarization of pump beam parallel to the rubbing direction. The orientation of the resultant ripple structure is parallel to the polarization of pump beam and the spacing of the structure are measured to be about 0.33μm. According to the theory of laser-induced periodic surface structure [15], the spacing of the formed ripple structure is dependent on the wavelength of pump beam, incident angle and the material. The theoretical value of the spacing is given by

$$\Lambda = \frac{\lambda}{n\cos\theta} \qquad (1)$$

where the λ, θ and n is the wavelength, incident angle and the refractive index of the material which adsorbed on the Sc. Substituting λ = 532 nm, θ = 0° and n = 1.6 into the

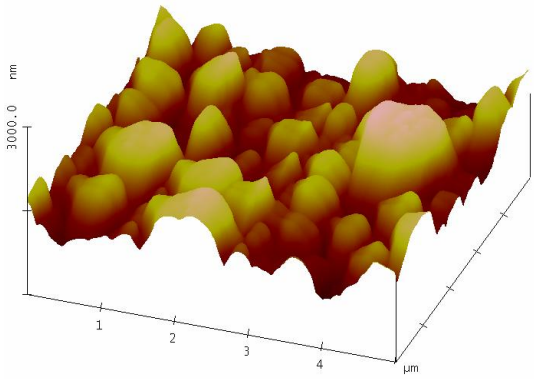


Figure 4 : The AFM image of the irregularly adsorbent structure on Sc.

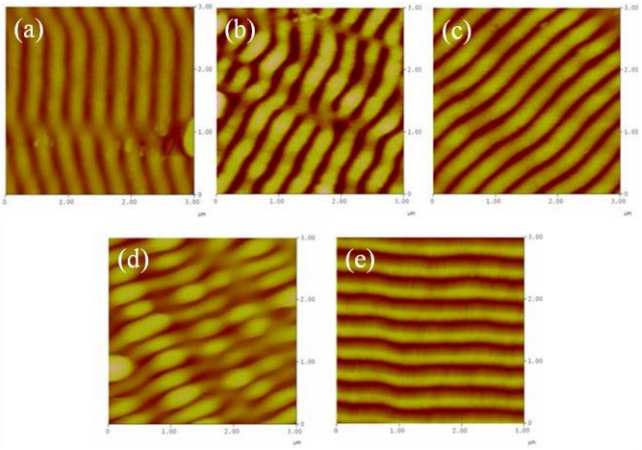


Figure 5 : Ripple structures generated by linear polarized pump beam (a) 0° (b)30° (c)45° (d)60° (e) 90° with respect to rubbing direction.

equation, the Λ is calculated to be about 0.33 μm which is consistent with the experimental value. Such ripple structures are believed to be generated by the interference between incoming polarized pump beam and the scattered light from the dye-adsorbed surface Sc [16]. Figure 5(b)~5(e) showed the resultant ripple structures generated by 30°, 45°,60° and 90°-linear polarized pump beam, respectively. We found that the orientations of the ripple structures were formed parallel to the linear polarization of pump beam. From the result of AFM measurement, we speculate that the direction of interference field between incoming pump beam and the scattered light is depending on the orientation of the adsorbed dye molecules. It is worth to mention that the depth of the resultant ripple structure which generated by 90°-linear-polarized exciting beam is lower than that generated by 0°-linear-polarized exciting beam under the same exciting intensity and duration. Additionally, the morphology of the resultant ripple structure, which generated by 90°-linear-polarized exciting beam, is more smooth than others relatively. The reason is that the polarized orientation of 90°-linear-polarized exciting beam was perpendicular to the major axis of the dichroic MR molecules. According to figure 6, the dichroic ratio of MR molecule is about 5 at the wavelength of 532 nm, so that the absorbed efficiency under irradiation by 90°-linear-polarized exciting beam was weaker than that irradiation by 0°-linear-polarized pump beam.

## 3.2 Elliptical Polarization

The ellipticity and inclination angle of the elliptical polarized pump beam were tuned by a polarizer and a quarter wave plate. The ellipticity is the intensity ratio of major and minor axes of the pump beam and the inclination angle is the azimuthal angle between major axis and rubbing direction. Figure 7 (a) and 7 (b) presents the AFM images of ripple structure which generated by 0° and

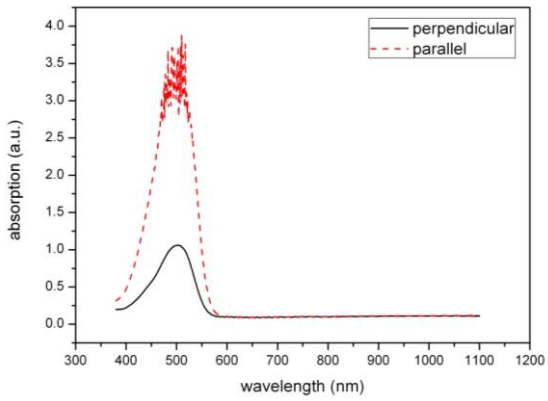


Figure 6 : The absorption spectrum of DDLC samples with dye molecules parallel and perpendicular to the pumped field.

45°-elliptical polarized pump beam respectively. The ellipticity of elliptical polarization is about 3 and the total intensity of pump beam is about 5mW/cm$^2$. From figure 7(a), the direction of ripple structure were obviously determined by Ey of the elliptical polarization field. Under irradiation by 0°- elliptically polarized pump beam, the MR molecules were adsorbed onto Sc and tend to reorient perpendicular to Ey. Due to the dichoric ratio of MR molecules, the polarization component Ey dominates the reorientation process which leads to more stable state. As the pumping period was extended, the ripple structure was generated and the groove direction was formed parallel to Ey. Similar to the case of 0°- elliptical polarization, under irradiation by 45°- elliptically polarized pump beam, the direction of resultant ripple structure was formed a angle 45° with respect to rubbing direction [ Figure 7(b) ]. Particularly, under irradiation by 90°- elliptically polarized pump beam (Ex>Ey), the intensity ratio Ix/Iy must be greater than some quantity. This is because that the dichoric ratio of MR molecules was about 5, the lower intensity ratio Ix/Iy will induce the MR molecules reorient perpendicular to the rubbing direction and the resultant ripple structure was formed parallel to Ey.

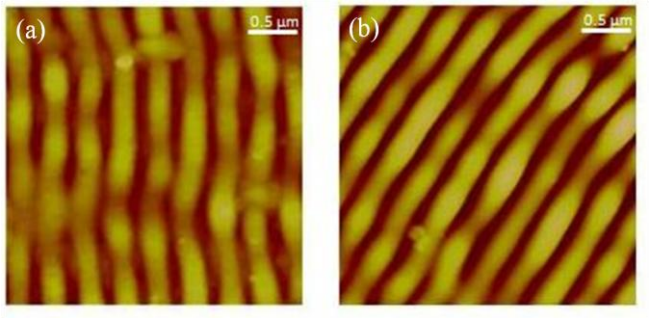


Figure 7: Ripple structures generated by elliptical polarized pump beam with azimuthal angle (a) 0° (b) 45° with respect to rubbing direction.

## 4 CONCLUSIONS

In summary, the generation of ripple structures have been demonstrated by various polarization of pump beam. Under irradiation by linear polarized and elliptical pump beam, the direction of ripple structures were formed parallel to the polarization of pump beam. Under irradiation by elliptical polarized pump beam, the direction of ripple structures were formed parallel to the major axis of elliptical polarization. The formation of ripple structures accompany with the permanent reorientation of liquid crystals. In the early stage, the liquid crystals were aligned perpendicular to the polarization of pump beam. In the later stage, the liquid crystals were re-aligned parallel to the polarization of pump beam.

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