Rewriteable Photoalignment of Liquid Crystals as a Route to High-Laser-Damage-Threshold Active Beam Shapers

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Abstract

The fabrication of rewriteable liquid crystal (LC) cells using high-damage-threshold photoswitchable azobenzene materials has been investigated as a route to all-optical active beam shapers. Beam shapers are used to modulate the intensity profile of OMEGA EP's petawatt-peak-power laser beams. All-optical photoswitchable beam shapers would allow intensity profile tailoring to occur at higher-fluence locations in OMEGA EP's beam lines than is currently possible using electro-optical LC beam shapers. Photoswitchable LC cells were assembled using one substrate coated with a non-switchable buffed polyimide alignment layer and a second substrate coated with an azobenzene-based photoswitchable alignment layer. The cells were filled with nematic E7 LC. Irradiating the cell with polarized UV light induced an azobenzene trans-to-cis isomerization, facilitating in-plane switching of LC alignment. The irradiation time was optimized to produce high-quality LC alignment. Different transmission functions were obtained after multiple alignment write cycles. During the fabrication of these devices, limitations in the azobenzene material's ability to continually rewrite were discovered, but high-quality rewritable behavior was obtained by exposing the photoswitchable layer to UV light through various masks.

1. Introduction

Beam shaping is required in high-peak-power laser systems such as the 60-beam, 40-TW, 351-nm OMEGA laser and the 4-beam, 1-petawatt 1054-nm OMEGA EP laser in order to ensure that the beams fill the entire aperture of the large-scale laser amplifiers and to compensate for spatial variations in energy across the laser beam clear aperture.[1] A beam shaper is an optical element that can control the spatial beam energy profile, either passively (at one fixed profile) or actively in real-time. In order to compensate for the lack of uniformity created by the amplification process, an inverse of the intensity profile of the amplified beam is used. After passing through the beam shaper and undergoing amplification, the high-energy beam entering the laser system's target chamber has a uniform intensity profile.

Laser beam shapers currently used in OMEGA and OMEGA EP are made up of patterns of metal pixels deposited on a fused silica substrate.[2] These metal pixels do not transmit the incident laser light, while the areas where the metal pixels are not present allow full transmission. Macroscopically, this distribution of pixels creates a smooth energy gradient and can be used to produce a beam with a tailored intensity profile. Such metal-mask beam shapers have a 1054-nm laser damage threshold of 0.2 J/cm^2 at 1-ns pulse width, which is relatively low compared to the output of a high-powered laser.

Liquid crystals (LC's) have also been used in the fabrication of beam shapers.[2] Nematic LC's display orientational order and have useful optical properties. On a molecular level, LC's are anisotropic in shape, like a rod or a disc, which causes them to exhibit optical properties whose values can change depending on what direction light is incident on the molecular structure. The combination of anisotropy and orientational order results in birefringence, which

is defined as the difference between the refractive index measured along the LC material's long molecular axis and the index measured at an angle to the molecular axis. Birefringence allows the polarization or the phase of incident light to be altered.

To make use of the optical properties of LC materials in devices such as beam shapers, the alignment direction of the long molecular axis must be controlled. Molecular alignment on the surface of a substrate used for LC devices is accomplished by inducing a preferred alignment direction in a thin polymer layer deposited on the substrate surfaces by either mechanical buffing or irradiation with polarized UV light ("photoalignment"). Buffing requires a velvet cloth to be rubbed against the polymer alignment coating, and the LC molecules align along the buffing direction. This system of alignment is photo-stable, so the alignment direction will not change from its original orientation. Because the surface of the substrate comes into contact with the fabric, the buffing process induces scratches and particulate contamination in the coating surface, which lowers the device's laser damage threshold. In the photoalignment process, a photoactive polymer is exposed to polarized UV light, causing the material to dimerize either parallel or perpendicular to the direction of polarization, depending on the photochemistry of the alignment layer. The LC molecules align along the dimerization direction. Because the process is non-contacting, it is inherently clean, and photoaligned LC devices typically show significantly higher laser damage thresholds than do buffed LC devices.

Previously, passive LC beam shapers that function in a very similar manner to metal-mask beam shapers were fabricated at LLE using a coumarin photoalignment layer (Fig. 1). Such coumarin-based photoaligned beam shapers have 1054-nm laser damage thresholds upwards of 27 J/cm^2 (1-ns pulse) and show pixel resolution comparable to metal-mask beam shapers.[3] The high laser damage threshold of LC beam shapers allows the optic to be placed in different, possibly more convenient places after the beam has undergone more amplification.

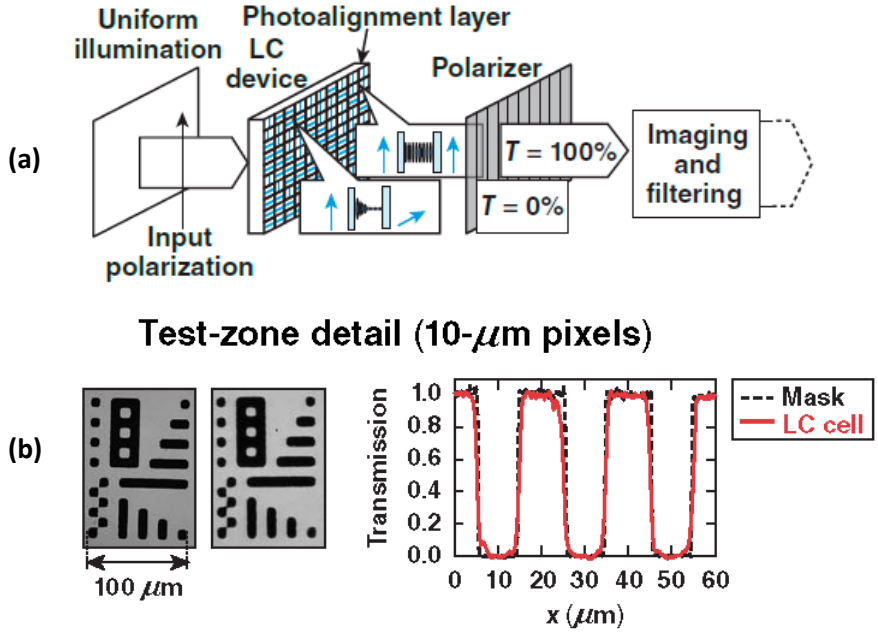

Fig. 1: (a) An illustration of the concept of the LC beam shaper. Polarized light passes through the LC device containing a distribution of pixels with parallel or twisted alignment states. The polarization of the light passing through each pixel changes accordingly and is either blocked by or transmitted through a second polarizer. (b) A test zone detail of a liquid crystal device on the right and a metal mask on the left along with the transmission graph comparing the two. The resolution and contrast of the liquid crystal device is on par with the metal masks currently in use.

Active laser beam shaping would allow the pattern of the pixels to be dynamically rewritten without replacing the optic. Currently, real-time, active beam shaping in OMEGA EP is conducted using electro-optical LC devices that contain a matrix of conductive-oxide electrodes deposited on the inner surfaces of the substrates, similar to what is used in LC displays in

computer monitors and flat-screen televisions.[3] The requirement for these conductive-oxide electrodes results in laser damage thresholds for electro-optical LC devices comparable to those of metal-mask beam shapers (< 1 J/cm^2 at 1054 nm, 1-ns pulse), limiting their use to low-fluence areas of the laser system. A photoaddressable, rewriteable alignment layer with sufficiently high laser damage threshold could be used to fabricate an optically addressed active LC beam shaper. Such materials are commercially available from Beam Engineering for Advanced Measurements

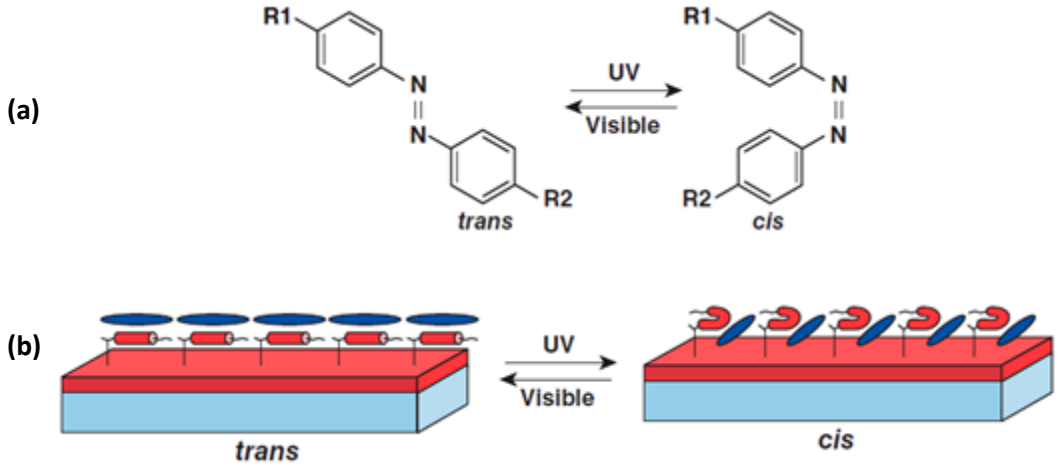

Fig. 2: (a) An illustration of the trans to cis isomerization of azobenzenes when exposed to UV light.(b) A representation of the in-plane switching of liquid crystal alignment when the azobenzenes undergo a trans to cis isomerization.

(BEAM) Company.[4] These materials are azobenzene based, and can therefore undergo a reversible trans- to cis- isomerization when exposed to polarized UV light. This change in shape results in reversible, in-plane switching of LC alignment (Fig. 2).[5] Recent work at LLE has shown that the BEAM Co PAAD photoswitchable azobenzene alignment layers have 1054-nm laser damage thresholds ranging from 28 to 67 J/cm^2 (1 ns pulse), depending on the alignment layer material composition.[3] These materials provide a potential route to an all-optical active LC laser beam shaper.

In order to determine whether these PAAD materials could potentially be used in the fabrication of an active beam shaper, LC devices were fabricated with an azobenzene-based photoalignment layer and a buffed polyimide (PI) coating. The PAAD photoalignment layer was photopatterned using a photolithographic UV lamp. These devices were characterized macroscopically and microscopically and assessed on their contrast between different alignment areas and the resolution sharpness of the transitional boundary. This work showed that PAAD alignment layers can rewrite and that a similar, high quality material has the potential to be used in the development of an active LC laser beam shaper.

2. Experimental

2.1 Device Fabrication

Two 50-mm-diameter fused silica substrates were cleaned via an aqueous cleaning process during which they were scrubbed with 0.05-µm MasterPrep® polishing suspension using a synthetic wiper (2 minutes), rinsed with 2 megohm-cm deionized (DI) water and nanopure DI water (1 minute each), and ultrasonically cleaned at 69°C in a solution of Extran detergent for 1 hr. The substrates were then rinsed with 18.5 megohm-cm DI water for one minute, before being blown dry with nitrogen to remove any visible moisture and placed on a hotplate to dry at 130°C for 30 minutes.

In this process, two metrics were used to ensure substrate cleanliness. The first was a seven-second water break test. While rinsing a substrate after the scrubbing step, each surface is flooded with DI water, and the substrate is held at an angle. The water should remain a continuous sheet across the substrate for at least seven seconds; breaks in the sheet of water indicate that hydrophobic contaminants are present, and the substrate should be scrubbed again.

The second metric was a visual inspection of the cleaned and dried substrate performed by looking through the substrate towards a fiber optic lamp. The substrate should appear clear (cloudiness is indicative of polishing abrasive dried on the surface) with no apparent particles on the surface. Substrates that do not pass this check must go through the entire cleaning process again. Additionally, substrates with obvious surface damage (e.g., scratches or pit marks), are discarded.

After the substrates were cool, one was coated with Nissan Sunever polyimide (PI) and buffed to provide UV-stable LC alignment and the other was coated with a photoalignment material (in this case PAAD-22). The first substrate was placed on a spin coater and flooded with the PI solution filtered through a 0.2 µm PTFE syringe filter. The solution was allowed to sit on the substrate for one minute before being spun off at 2000 rpm for 40 seconds. The coating was soft-baked on a hot plate at 80°C for 10 minutes, followed by hard-baking at 230°C for one hour.

A commercial buffing machine was used to buff the PI coating. The buffing wheel and motorized stage were vacuumed to remove loose fibers. Then, the PI coated substrate was placed on the stage near the buffing wheel, and the height of the wheel was adjusted to just graze the substrate. The wheel was turned on, and the substrate was held in place by vacuum. The substrate passed under the buffing wheel 8 times. After it was removed from the stage, the substrate was rinsed with 8.5 megohm-cm DI water and blown dry with nitrogen to remove any buffing fibers. This rinsing and drying were repeated until all fibers were removed. The substrate was then allowed to dry on a hot plate at 130°C for half an hour.

To deposit the PAAD-22 photoalignment material, the second substrate was first conditioned by placing it on the spin coater and flooding the surface with isopropanol, which was spun off immediately at 2000 rpm for 30 seconds. The substrate was then flooded with PAAD-22

filtered through a 0.2-μm PTFE syringe filter. The solution was spun off immediately with the same spin parameters. The coated substrate was placed in a clean hood to air dry for 10 minutes at room temperature.

The LC cell was then assembled with the coated substrates. Glass microspheres (8 μm dia.) were dispersed into Epotek UV curing epoxy and the mixture deposited as small spots onto the PI coated side of the substrate in three locations 120° apart using a syringe needle. The PAAD coated substrate was placed, coated side down, on top of the PI coated substrate, and the epoxy cured under a UV lamp for 10 minutes. The empty cell was placed on a hot plate along with a surface thermometer and the hotplate temperature was raised to at least 70°C (past the clearing point of the LC). The inter-substrate gap was filled by capillary action with the LC mixture, E7 containing 0.07 weight-percent of the chiral twisting agent CB15 in its isotropic phase. The filled cell was cooled at a rate of 10°C per hour to ambient temperature.

2.2 Device photopatterning

To complete the device, the fluid-filled cell was irradiated with polarized UV light in a photolithography setup as shown in Fig. 3.

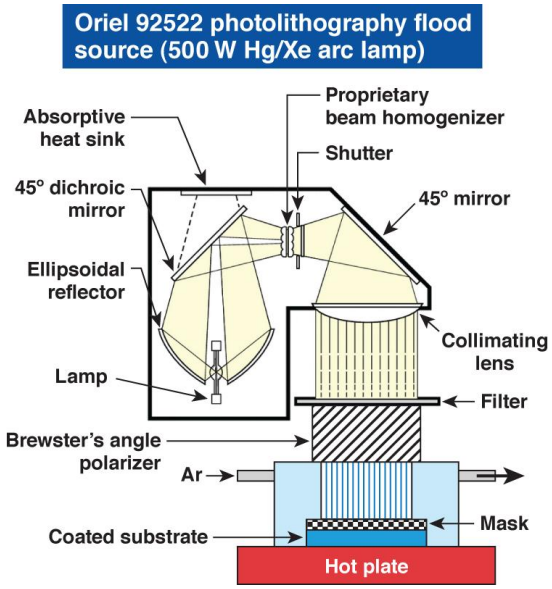

Fig. 3: The photolithography setup used to pattern and expose photoalignment layers for the LC beam-shaper device. Pictured at the bottom is a coated substrate that is exposed to the filtered, polarized light coming from the 500 watt lamp.

The cell was placed in the irradiation chamber with the buffing direction of the polyimide-coated substrate either parallel or perpendicular to the UV polarization direction and with the top substrate having the photoalignment layer. To create a patterned device, the cell was exposed to UV light, then rotated 90° and irradiated again through a mask. This action resulted in a rewritten portion of alignment in the unmasked area. The cell was continually rewritten by changing the orientation of the substrates with respect to the polarization direction and using different masks.

2.3 Device characterization

Devices were characterized both macroscopically and microscopically (Figure 4). Macroscopic characterization was conducted by viewing the devices between crossed and parallel polarizers to evaluate the overall alignment quality. High quality alignment was indicated by high contrast between different alignment areas and by sharp resolution in transition areas. The boundary resolution between adjacent alignment zones and the level of coating contamination were determined by examination of the devices in transmission with a polarizing microscope at 100 x magnification.

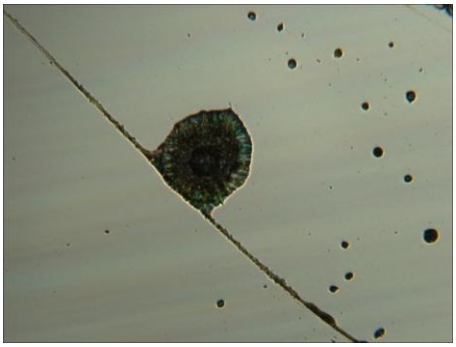

Figure 4a: A microscopic view of phase separation. This concentration of dye in the coatings is detrimental to the alignment quality of LC devices

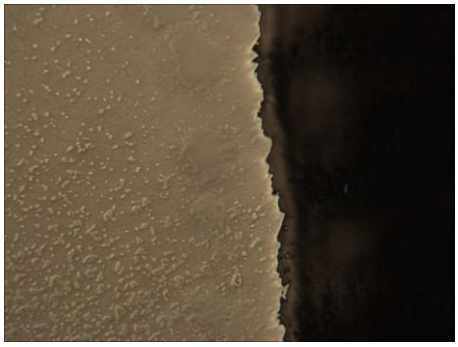

Figure 4b: A microscopic view between crossed polarizers of the boundary between two alignment areas. The resolution of this boundary is quite good, but is not on par with metal mask beam shapers.

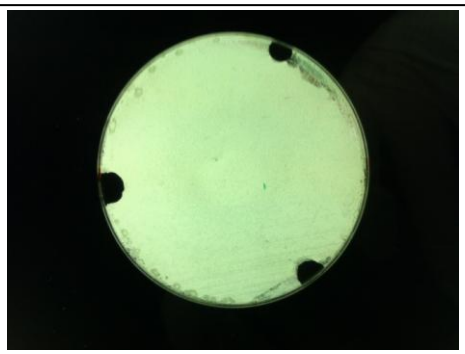

Figure 4c: A macroscopic view of a device with twisted alignment between crossed polarizers. This example shows good alignment and very few features.

Figure 4d: A microscopic view between crossed polarizers showing the texture of the coating. There are few features and the texture is mostly smooth, which shows good coating quality.

3. Results and discussion

An initial experiment determined that the PAAD-22 produced LC alignment perpendicular to the direction of the polarized light used to irradiate the device. In this experiment a device was irradiated, keeping track of the buffing direction, and then viewed between crossed and parallel polarizers. When the buffing direction was aligned with the direction of polarization of the UV light, a twisted cell was produced. Twisted cells look bright between crossed polarizers, and dark between parallel polarizers.

The quality of LC alignment varied with irradiation time. In order to determine the optimal irradiation time, a bracketed exposure experiment was conducted where a device was exposed to UV light for 30 seconds at a time. The device was first irradiated to give a twisted cell. One half of the device was rewritten to a parallel aligned configuration. For every 30 seconds of irradiation in the parallel aligned orientation, the device was characterized macroscopically to see if the quality of alignment improved. It was discovered that the contrast between two sections of alignment continued to improve until 4 minutes of irradiation had gone by. After that amount of time, the quality no long showed any substantial changes. From here on, exposure time was set to 4 minutes.

The patterned device produced as shown in Fig. 5 demonstrated the ability to write and rewrite a pattern. First, the device was written with twisted alignment, then rotated and masked to produce the first pattern. That pattern was rewritten by irradiating in the same orientation without the mask. The second pattern was written by turning the device 90°, masking it, and irradiating it. Each irradiation lasted 4 minutes. Although the first pattern was sharp, rewriting proved to be tricky and the resulting device was of a lower quality.

A memory effect was also observed while using these materials. After rewriting a pattern, remnants of the original would sometimes be visible. Sometimes, this effect would appear immediately after the initial erasing or rewriting of the pattern (as in Fig. 5), while on other occasions, the original pattern would disappear and then reappear in later irradiations. To determine if the LC was *remembering* its original pattern, the cell was heated to the isotropic phase and then cooled at 10° C per hour. This treatment had no noticeable effect on the memory. This confirmed that the memory effect was rooted in the photoalignment layer. In an effort to mitigate memory effects, devices were irradiated for longer periods in order to more fully expose the azobenzenes. Unfortunately, this method also proved to be inadequate.

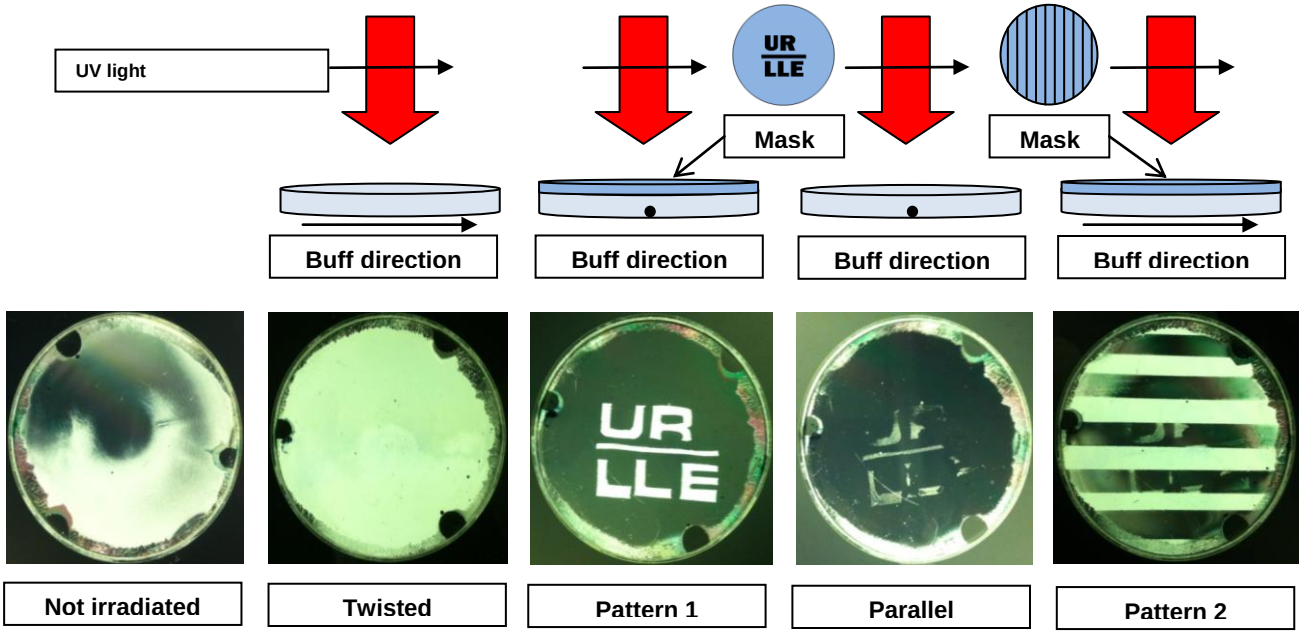

***Fig.5**: A schematic showing the irradiation procedure for rewriting an LC device. Each irradiation lasted 4 minutes and all images show the device through crossed polarizers. First, the device was written with twisted alignment, then rotated and masked to produce the first pattern. That pattern was rewritten by irradiating in the same orientation without the mask. The second pattern was written by twisting the device 90°, masking it and irradiating it.*

Another device was tested to determine the maximum number of times the PAAD material can be rewritten (Fig. 6). The irradiation of this device did not utilize a mask – instead

the entire device was rewritten with each exposure by changing its orientation with respect to the polarization direction. Each irradiation lasted 4 minutes and was accompanied by a decreasing alignment quality and an increase in pretilt, where the LC molecules do not lay flat on the alignment layer. With each irradiation, the increased pretilt caused the response of the device to applied pressure to increase. This effect is seen as the alignment becomes less homogeneous and there are no longer two different alignment orientations, but many. One possible cause of the increased pretilt may have been residual dimethylformamide (DMF) escaping from the coating. Attempts to cure the coating either by air drying for a longer period of time before cell assembly or by gently heating the coated substrate (65°C) showed no improvement, but DMF has not been ruled out as a potential cause. Increased phase separation in the photoalignment coating was also seen in multiple devices after many irradiations had taken place. Phase separation was observed as the formation of orange droplets in the coating (Fig. 4a). Another source of phase separation was the spin-coating process. During the coatings when less than 2 mL of PAAD material was used, phase separation was immediately and macroscopically visible, while when 2 mL or more was used, even microscopically there was little or no phase separation.

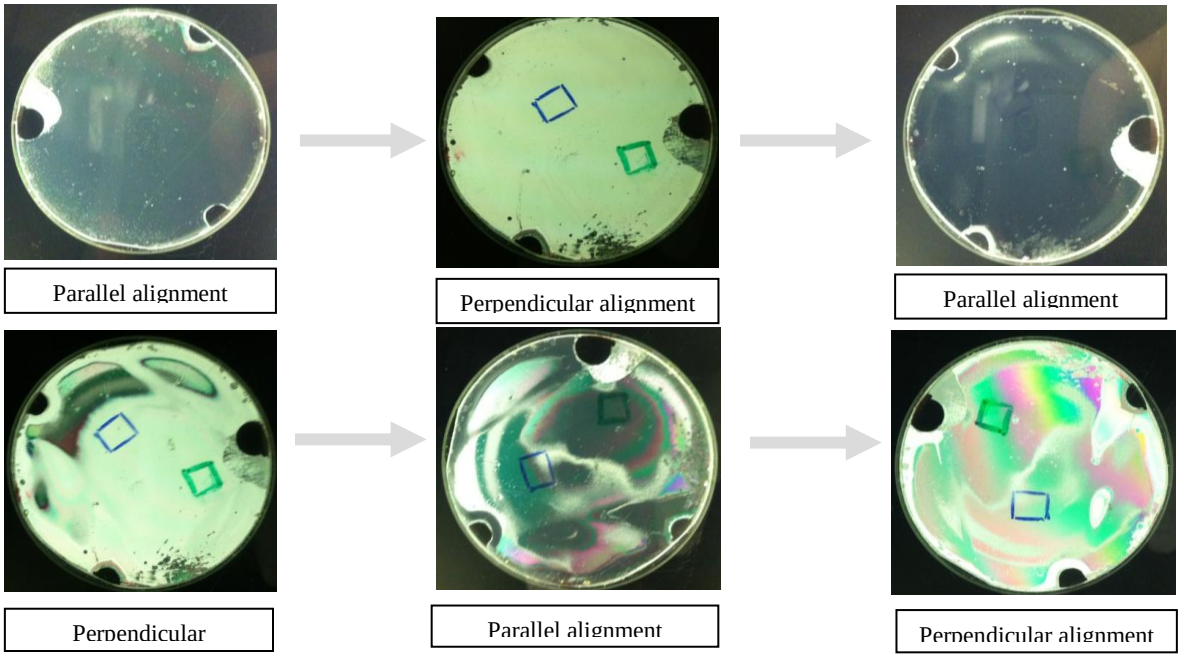

***Fig.6**: Images of an LC device that was rewritten until the alignment became indistinguishable. This experiment shows that after approximately three irradiation alignment cycles, there is markedly more pre-tilt and instability in the device. The deterioration of the alignment is a concern for the feasibility of the fabrication of an active laser beam shaper.*

4. Conclusions

Using PAAD-22, a photoalignment material, rewriteable LC devices were fabricated. These devices demonstrated that it is possible to rewrite patterned liquid crystal cells. An optimal irradiation time of four minutes was discovered alongside various rewriteability limitations. Procedural improvements were also made in order to reduce the amount of phase separation in the PAAD coating. Although these materials were shown to be rewriteable, the quality of the alignment is simply not yet good enough for fabrication of laser beam shapers. Future work will look into improving the quality of the coating and minimizing the memory effects. Ultimately, a new photoalignment material will mostly likely need to be developed in order to make an active laser beam shaper feasible. The concept of rewriteability has been proven possible and will see valuable applications if improved upon in the future.

5. Acknowledgements

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6. References

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