

Liquid Crystal Beam Shaping Devices Incorporating Coumarin-Based Photoalignment Layers

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1. Abstract

Liquid crystal (LC) devices incorporating photosensitive polymers based on coumarin were investigated for laser beam shaping applications. Presently, the OMEGA EP laser system uses pixelated metal beam shaping devices, though their relatively low damage threshold (approximately 0.2 J/cm^2) makes them poor options for long-term usage. LC devices have the major advantages of a much higher damage threshold (9-18 J/cm^2). Previously, $10 \text{ }\mu\text{m}$ pixelated LC beam shapers were fabricated by patterning a commercially available cinnamate-based photopolymerizable LC alignment material (ROLIC) through a mask using polarized UV light to control the LCs' orientation. This project investigated fabrication and characterization of LC beam shaping devices employing a coumarin-based linearly photopolymerizable polymer (LPP) as an alternative to the cinnamate-based LPP. After developing an experimental procedure for creating coumarin-based LPP liquid crystal cells, analyzing cell transmission data, and evaluating LPP coating uniformity, it was determined that the coumarin-based LPP could be photolithographically patterned. However, more research is needed to improve coating uniformity and determine the UV irradiation conditions necessary to improve pixel resolution.

2. Introduction

Beam shaping devices are important to the design of high-energy laser systems. In order for the beam to reach the intensity of its final output, it must first pass through amplifiers. Laser beam amplifiers cause spatially varying gain in the beam, meaning that the resulting amplified beam is non-uniform. Beam shaping mechanisms precompensate for this spatially varying gain, thus producing uniform amplified beams and facilitating optimal energy extraction. Figure 1 portrays, in theory, the effectiveness of precompensating for spatially varying gain using beam shaping devices. The resulting output beam from the amplifier is of the desired uniform intensity.

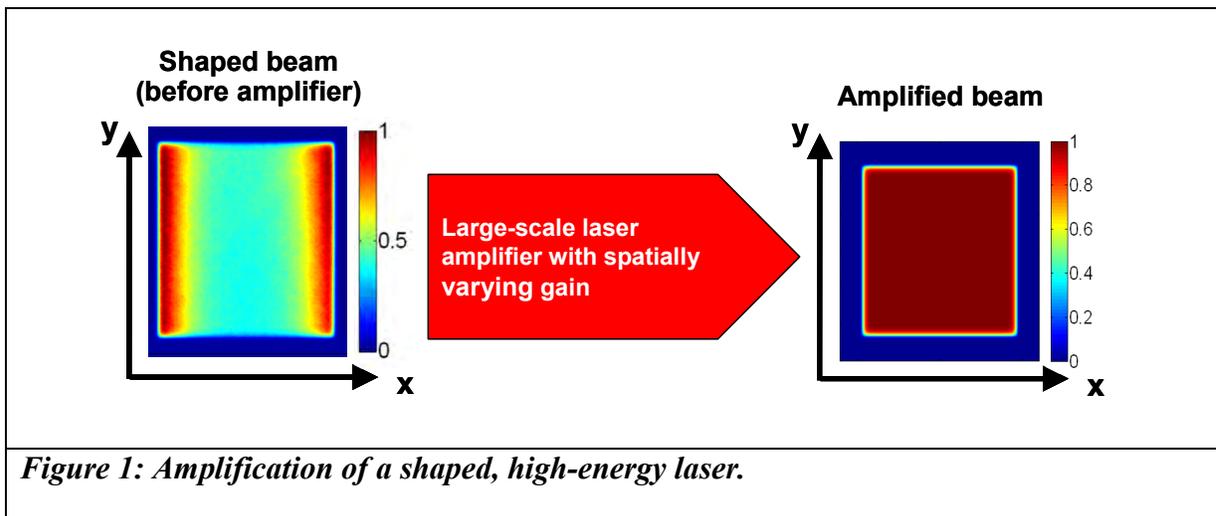


Figure 1: Amplification of a shaped, high-energy laser.

The OMEGA EP laser system, which was developed to provide ultrahigh intensities for advanced x-ray and proton-beam radiography of compressed targets, currently incorporates metal-mask fused-silica beam shapers. These masks have microscopic pixels allowing for either full transmission or no transmission of laser beam light. Despite these two transmission extremes, metal masks can produce a range of transmissions over the device dependent on the concentrations of the pixels. Using a

metal mask with a high concentration of full-transmission pixels (i.e., metal has been removed to expose the transmissive fused silica glass underneath) in one area, for example, will allow for high percentages of locally transmitted light.¹

While the metal masks are effective in shaping the beam, they decrease the optimal energy extraction of the system because of their low damage threshold (approximately 0.2 J/cm² at 1054 nm, 1 ns). That is, the beam's intensity is limited by the damage threshold of the beam shaping mechanism. Liquid crystal beam shaping devices have been investigated for use in high-energy laser beam shaping because their components—the LCs and LPP on which they align—have much higher damage thresholds (9-18 J/cm² at 1054 nm, 1 ns and up to 60 J/cm² at 1054 nm, 1 ns, respectively).²

An additional advantage to LC beam shapers is that, when they are paired with a polarizer, the LC molecules' orientations can allow for full transmission of polarized light, no transmission of polarized light, or a percentage of polarized light transmission. Full transmission of polarized light can be achieved by assembling the LC devices such that the LC molecules align parallel to each other throughout the device, and in the same direction of the polarized light. No transmission of polarized light can be achieved by assembling the LC devices such that the molecules' alignment "twists" 90°. Since the polarized light can only pass through LC molecules oriented in the same direction as the light's polarization, the 90° change in LC molecule orientation effectively blocks the transmission of light. Partial transmission of light is achieved with LC devices that have been assembled such that the molecules are arranged at any degree of orientation between parallel and perpendicular assembly.¹

Figure 2 is a schematic of the LC beam shaper's incorporation into a laser system. The beam shaping mechanism consists of pixels that allow either full transmission or no transmission of the light. The magnified diagrams of the pixels show the orientation of the LC molecules within the cell.

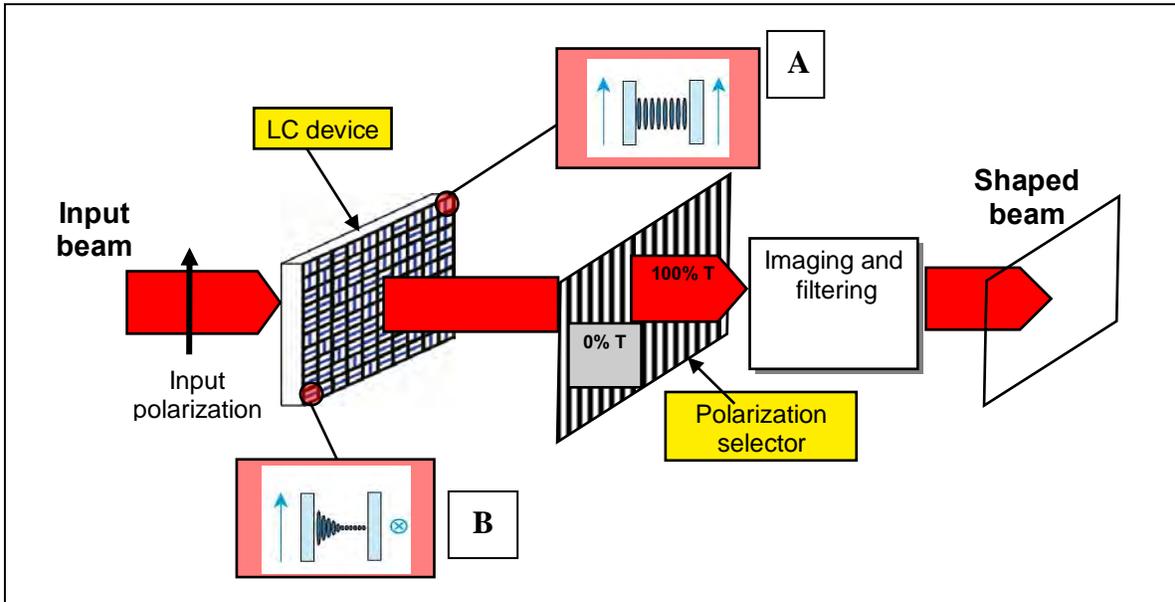


Figure 2: Incorporation of a liquid crystal beam shaper into a laser system.

(A) Parallel orientation of LC molecules produces 100% transmission;

(B) Twisted LC orientation produces 0% transmission.

The two current methods of liquid crystal alignment are mechanical buffing and the cleaner, non-contact, preferred process of “photoalignment”.⁴ Photoalignment was the method used in this experiment. Photoalignment facilitates LC alignment by using polarized UV light, rather than physical contact, to align a linearly photopolymerizable polymer (LPP) that has been deposited on the surface of a substrate. In the process, irradiation by polarized UV light causes the polymer chains of the coumarin-based LPP to align on the surface of the substrate in the same direction as the polarization of the UV

light. UV irradiation causes the partially polymerized LPP to become cross-linked, enabling the alignment of liquid crystal molecules along it.² Patterning of the LPP is possible when irradiation first occurs through a photomask substrate, before the mask is removed and the substrate rotated 90° and re-irradiated. Van der Waals forces enable the LCs to align along the patterned LPP when the device is assembled.^{4,5} The photoalignment and LC cell assembly process is depicted in Figure 3.

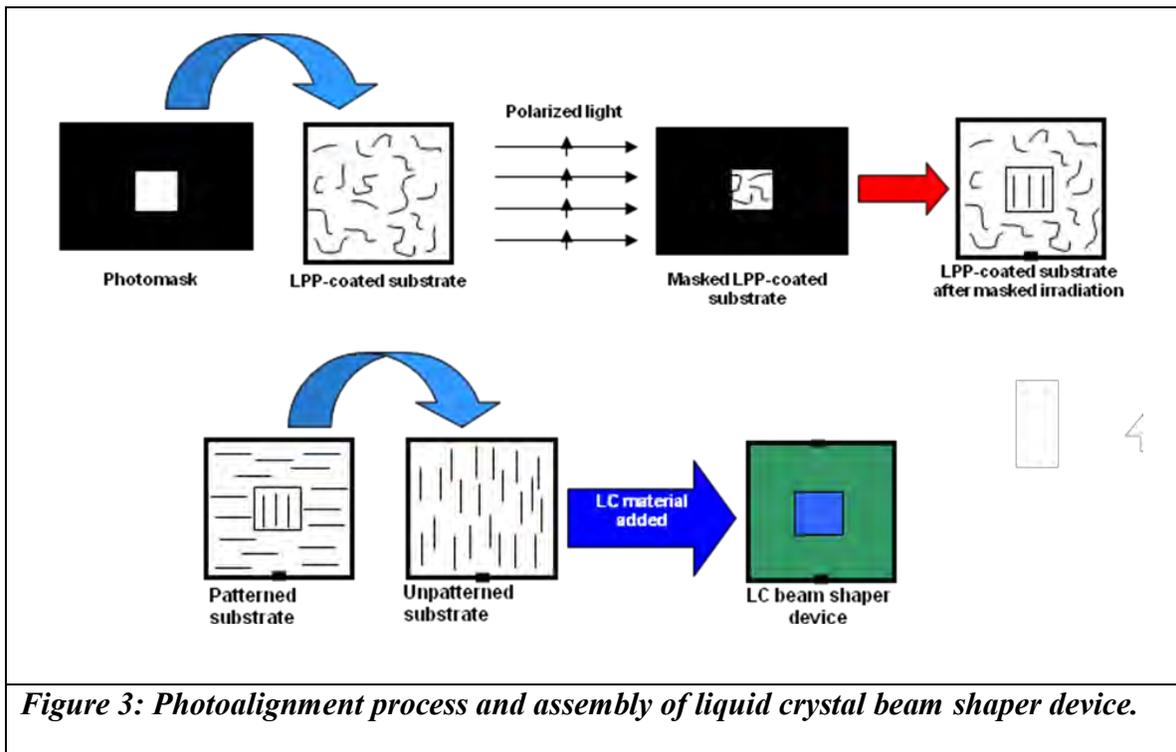


Figure 3: Photoalignment process and assembly of liquid crystal beam shaper device.

The center of the assembled device shown in Figure 3 would allow for full transmission of light, as did the LC molecule orientation in case A of Figure 2. The rest of the LC beam shaper device, colored green in Figure 3, would allow for 0% transmission of light, as in case B of Figure 2.

Previous research on LC devices has incorporated the cinnamate-based LPP ROP-103/2CP, produced by the company ROLIC. Using this LPP, LC devices have been

patterned with high resolution 10 μm pixels.¹ However, ROP-103/2CP is no longer available commercially, leading to the investigation of a new coumarin-based LPP invented and synthesized at the Laboratory for Laser Energetics. In addition to its greater availability, coumarin presents the advantage of a higher damage threshold than that of ROP-103/2CP. This project involved the development of a photoalignment and LC cell preparation procedure for beam shaper devices incorporating the novel coumarin photoalignment material rather than the formerly used ROP-103/2CP.

3. Experiment

Pre-cut fused-silica glass substrates were used for the assembly of the liquid crystal devices. The substrates were wetted with water, scrubbed with 0.05 μm deagglomerated alumina micropolish, and rinsed with filtered deionized water before being subjected to the seven-second water break test. This test served to reveal any organic surface contaminants that were not removed by the scrubbing. Each substrate was flooded with water and observed for seven seconds to see if the surface tension of the water on the substrate was broken, which would indicate a need for additional manual cleaning prior to the substrate's placement in an ultrasonic bath. After the water break test, the substrates were cleaned with approximately 8 ml of Extran detergent in the ultrasonic bath at 69°C for 60 minutes. After removal from the ultrasonic bath, the substrates were rinsed with tap water for 3-5 minutes followed by filtered deionized water for 3-5 minutes. The substrates were then dried with a nitrogen air gun and put on a hotplate to dry at 130°C for 15 minutes. The dry substrates were allowed to cool to room temperature before spin-coating.

The cleaned substrates were placed on a spin-coater, covered with a glass dish to prevent evaporation of the chlorobenzene base coat and allow it to equilibrate with the glass substrate and produce a saturated vapor atmosphere. The glass cover dish had multiple holes around its circumference that were taped over to create a closed atmosphere in which any chlorobenzene vapors would remain during spin-coating. The substrates were flooded with pure chlorobenzene using a 0.2 μm PTFE 13 mm hydrophobic syringe filter, clean needle, and glass syringe inserted through one of the taped-over holes. The chlorobenzene-flooded substrate was spun at 3000 rpm for 60 seconds to evenly coat the surface of the substrate before the LPP was applied. The chlorobenzene-coated substrate was then flooded with 0.3wt% diluted coumarin (LPP) in chlorobenzene, which was deposited on the substrate through a 0.2 μm PTFE 13 mm hydrophobic syringe filter using a second clean needle and glass syringe. The substrate was immediately spun at 3000 rpm for 120 seconds. The coated substrates were allowed to air-dry for 10 minutes.

After they had air-dried, the substrates were pre-baked at 50°C for 15 minutes. The pre-bake was developed after a comparison of pre-baked photoaligned coatings and non-pre-baked photoaligned coatings concluded that pre-baking the coated substrates prior to photoalignment resulted in a more uniform coating. After the pre-bake, the substrates were vacuum dried for one hour at room temperature and a pressure of approximately 0.1 torr to drive out the residual chlorobenzene. Photoalignment was accomplished by placing the substrates beneath a 500-watt mercury xenon UV lamp at 325 nm. They were placed beneath a “pile of plates” polarizer made of fused-silica microscope slides arranged at Brewster’s angle of 56.1° (Figure 4).

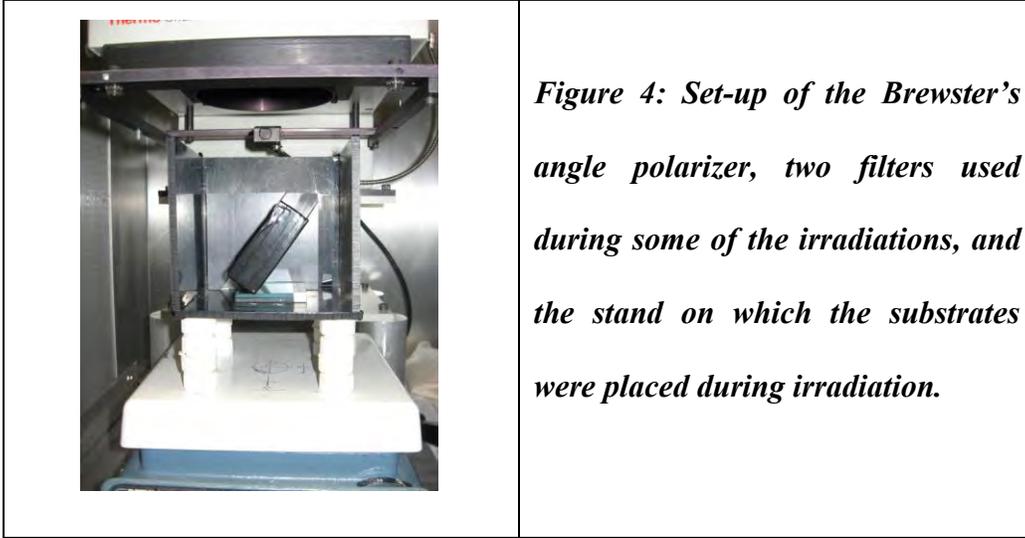


Figure 4: Set-up of the Brewster's angle polarizer, two filters used during some of the irradiations, and the stand on which the substrates were placed during irradiation.

Photopatterning of the alignment layer was accomplished by irradiating one substrate at 0° rotation with no mask. The second substrate was irradiated twice: first using a patterned mask, and second with the mask removed and the substrate rotated 90° . Irradiation times varied based on photomask substrate composition and the transmission of filters used.

Substrates were assembled into cells using Epo-Tek 5-minute epoxy. The gap between each cell's constituent substrates was controlled by dispersing $8 \mu\text{m}$ glass microspheres into the epoxy. A small amount of this spacer-containing adhesive was placed on each of the four corners of the first substrate. The second substrate was placed on the first one in such a way that the initial direction of 0° of rotation in the first substrate matched that of the second substrate (parallel assembly). The assembled cells were placed on a hotplate at 72°C and filled with a mixture of nematic E7 and 0.07wt% CB15 (an anti-reverse twist agent used to prevent random incorrect orientations of LC molecules) using a third clean needle, glass syringe, and $0.2 \mu\text{m}$ PTFE 13 mm

hydrophobic syringe filter. The completed cells were allowed to cool to room temperature on a hotplate at a rate of 10°C per hour. All operations except substrate cleaning and photoalignment took place in class 100 hoods.

4. Discussion

Coumarin-based photoalignment materials have been photolithographically patterned with high-resolution masks for the first time, though further research is needed to achieve sharp, high-contrast, and well-resolved pixels of 10 μm or less. Much of this project was focused on determining the proper irradiation conditions for the coumarin polymer.

As prior research with the ROP-103/2CP LPP had shown, there is a correlation between photoalignment resolution and irradiation time.¹ The coumarin polymer was found to be highly sensitive to both the wavelength and intensity of the UV irradiation. LC cells prepared with masked LPP-coated substrates that had been irradiated with the highly transmissive fused-silica photomask for five seconds or more were found to be overexposed; that is, the pixels in the devices were larger and darker than desired (Figure 5). In contrast, the LPP was found to be underexposed after a one second irradiation time. This was concluded after the completed cell failed to reach full extinction, blocking all light transmission, when viewed under crossed polarizers.

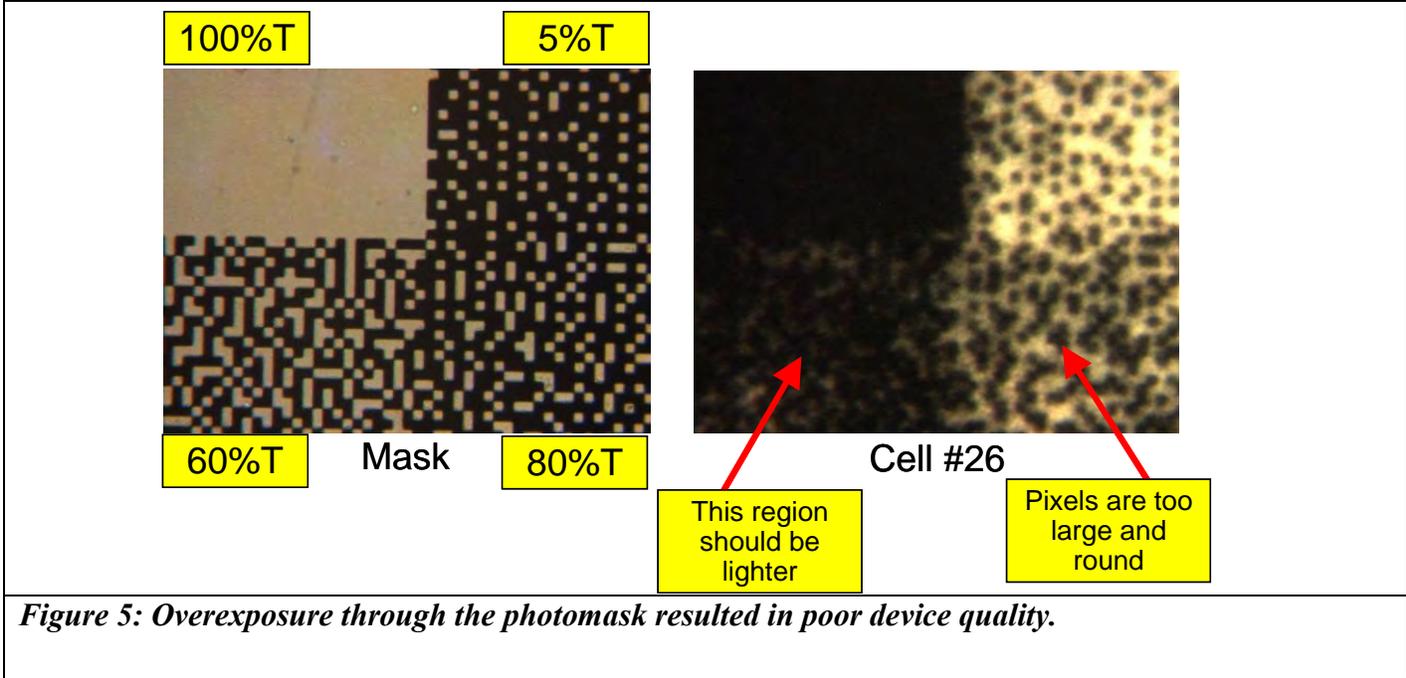
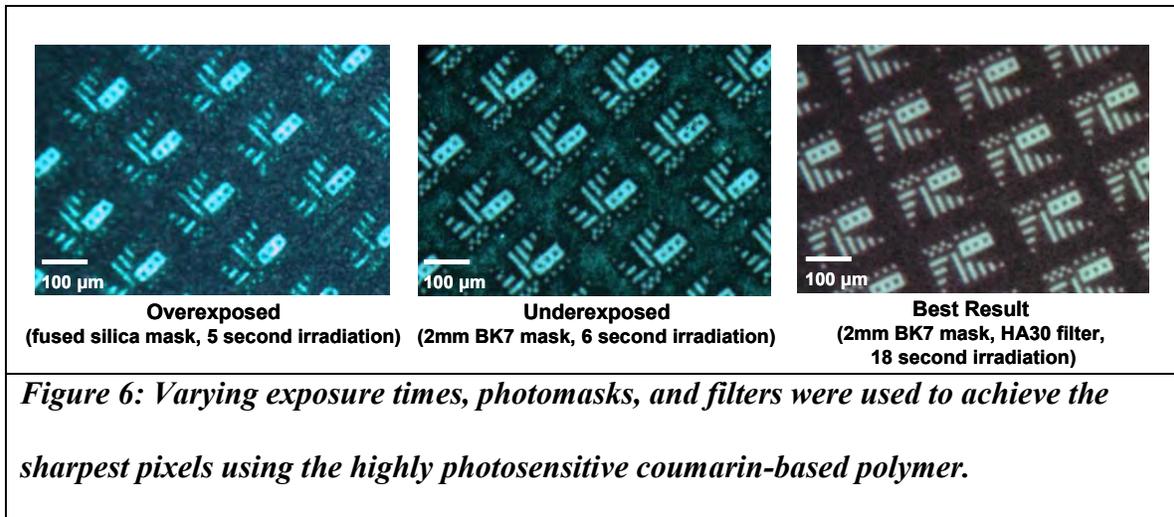


Figure 5: Overexposure through the photomask resulted in poor device quality.

In order to lengthen the LPP irradiation time and increase the range from underexposed to the ideal exposure time, various filters and a lower-transmission BK7 photomask were used to reduce the intensity of the UV light irradiating the coumarin polymer. Irradiation times using the filters were determined by analyzing a series of cells at different exposure levels, each of which had been irradiated through the BK7 mask. By multiplying the known value of the UV lamp's energy flux, or the rate of transfer of energy per unit area, by the exposure time each cell had been irradiated, the upper and lower irradiation fluence limits were determined. Using this data, the proper irradiation times for multiple filters were determined. The clearest pixels and most successful coumarin LPP patterning was achieved using the BK7 photomask and HA30 filter. Figure 6 shows the varying quality and irradiation conditions for an overexposed cell, an underexposed cell, and the best result.



5. Conclusion

The purpose of this project was to investigate the feasibility of using coumarin-based photoalignment layers in liquid crystal beam-shaping devices. While coumarin-based LPPs have been patterned for the first time, further research is needed to achieve higher-quality pixels.

In even the highest quality devices that have been made, the dark areas appeared to have a grainy texture; the device does not achieve zero transmission of light in the intended areas. Coating smoothness was confirmed using atomic force microscopy and a Nomarski microscope, but the source of the graininess is yet unknown. Further research is needed to investigate and correct the issue of graininess in the LC cells incorporating coumarin-based LPPs.

With further research, coumarin-based LPPs could be used in beam shaping devices of large-scale laser systems, eventually replacing the metal beam shapers. Achievement of this long-term goal would greatly increase the damage threshold of the

laser system, as the beam will be adequately shaped using optical materials capable of withstanding highly damaging beams.

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