
Photo-aligned anisotropic optical thin films

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Abstract — Photo-alignment of anisotropic optical thin films enables realization of novel optical elements, which were not feasible up until now. Photo-aligned anisotropic thin films can be applied to rigid or flexible substrates, which may be flat or curved. The optical performance of such films can be tailored to application-specific requirements by introducing tilt profiles of the optical axis and/or generate patterned retarders with continuous or periodical in-plane variation of the optical axis.

Keywords — Photoalignment, optical retarders, thin film, multilayer.

1 Introduction

Many optical applications, which operate with polarized light, make use of anisotropic optical elements. Most often, stretched polymer films are used for large-area and large-volume applications. Because stretching induces non-equilibrium molecular distribution in the polymer film, relaxation starts once the temperature approaches the glass-transition temperature of the film. Consequences are mechanical shrinkage and a decrease in the optical anisotropy.

Since stretching can only be applied along or perpendicular to the web direction, the in-plane optical axis directions of stretched retarder films are restricted to either parallel or perpendicular to the edges of the film. For liquid-crystal-display (LCD) applications, retarder films are usually laminated directly onto the polarizers with an application-specific angle between the main axes of retarder and polarizer film. Because roll-to-roll lamination would only allow parallel or perpendicular orientation, polarizer and retarder films have to be cut at an angle and laminated together in a slow and labor-intensive batch process.

Over the last few years, coated anisotropic films were introduced to the market as they offer several attractive features. For example, twisted retarders¹ are used in double-cell supertwisted-nematic (D-STN) LCDs instead of the passive STN cell in order to compensate the intrinsic retardation of active STN-LCDs. Films consisting of splayed discotic materials are widely employed in thin-film-transistor (TFT) twisted-nematic LCDs (TN-LCDs) to improve the viewing-angle performance of computer and TV LCD monitors.²

2 Properties of photo-aligned anisotropic films

The combination of ROLIC's linear photo-polymerization (LPP) technology³ with specifically developed liquid-crystal polymer materials (LCP) offers a unique flexibility in

designing and processing of anisotropic thin films.⁴ For many applications, anisotropic films work as birefringent layers, such as quarter- or half-wave retarders. However, LCP materials may also comprise dichroic dyes to realize coatable linear polarizers or chiral dopants to achieve narrow or broadband cholesteric filters, twisted retarders, or negative c-plate retarders.

As a basic advantage, the LPP photo-alignment process is contact free and therefore not restricted to rigid and flat substrates, but can be applied to flexible and/or curved substrates as well. Furthermore, the alignment direction can be set to any angle just by rotation of the uv-polarization plane. Consequently, photo-alignment is the ideal solution for large-volume roll-to-roll processing of flexible film substrates. Contrary to mechanical brushing, photo-alignment does not scratch the surface of the substrate. Therefore, multiple anisotropic layers with individual optical axis orientation can easily be stacked on top of each other without damaging the lower layers.⁵

Both LPP and LCP materials can be designed such that the optical axis within the LCP layer is arbitrarily tilted at the LPP and/or at the air boundary according to the requirements of the respective application. Hence, novel viewing-angle-enhancement films for TN-LCDs can be realized which counteract the residual birefringence of the tilted liquid-crystal molecules of the LCD.

Contrary to the uniaxial optical axis orientation of conventional anisotropic films, ROLIC's LPP/LCP technology allows the generation of orientation patterns with submicron resolution for applications such as 3-D LCDs, patterned interference color filters, micro-optics, etc.

2.1 Preparation of photo-aligned anisotropic films

The process steps to manufacture photo-aligned anisotropic films involve wet-coating of an LPP layer, drying at elevated

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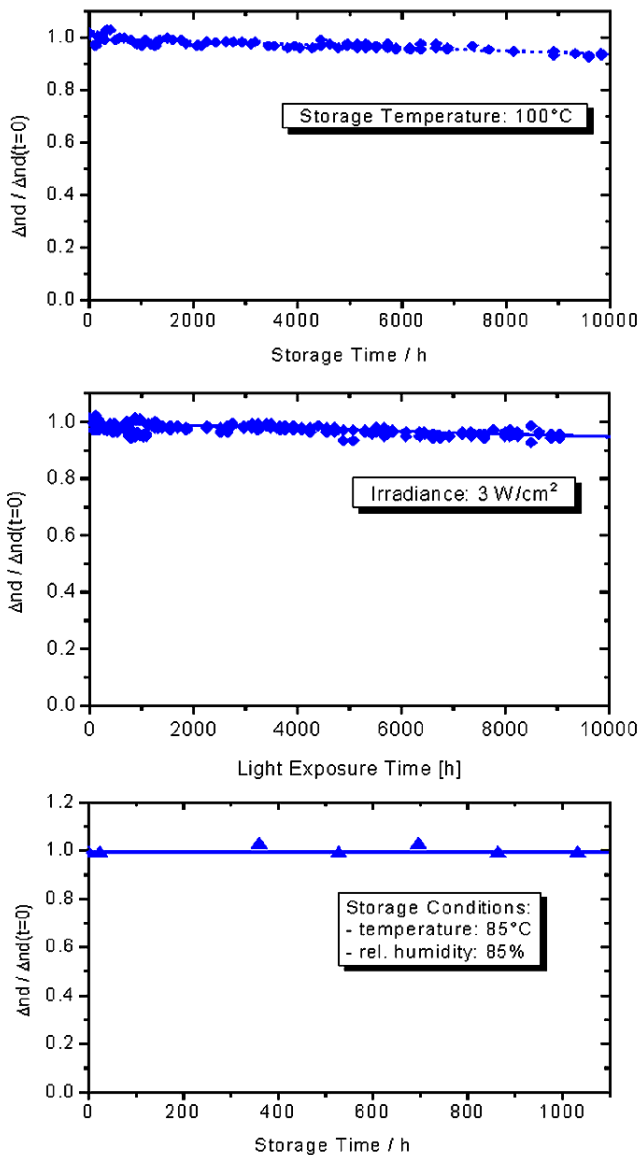


FIGURE 1 — Stability of LPP/LCP retarders. Top: Thermal stability at 100°C. Middle: Photo-stability (irradiance = 3 W/cm²). Bottom: Stability to humidity at 85°C/ 85% RH. LPP material: Staralign™ 2100, LCP material: Opalva™ 2130.

temperature, exposure to linearly polarized uv (LPUV) light, wet-coating of cross-linkable liquid-crystal materials, drying, annealing, and uv cross-linking. The thickness of LPP layers is typically between 20 and 50 nm, whereas the LCP layer thickness depends on the desired optical function. For example, the thickness of an LCP quarter-wave retarder is around 1 μm.

2.1.1 Coating

Standard coating and printing techniques such as spin-coating, gravure coating, slot dye coating, offset printing, *etc.*, are applicable to both LPP and LCP materials.

2.1.2 UV polarization

UV light is typically polarized using plastic-sheet polarizers, wire-grid polarizers (Moxtek), or Brewster-type polarizers. Brewster type polarizers are most stable to UV light but have some restrictions in the acceptance angle which requires a certain amount of collimation of the incident UV light. On the other hand, wire-grid polarizers can easily be combined with any kind of UV light source, and allow the construction of large-area tiled UV polarizers.

2.1.3 UV exposure

ROLIC has developed highly uv-sensitive LPP materials, which exhibit perfect alignment capability upon LPUV exposure with energies of only a few mJ/cm² (wavelength range, 280–340 nm). In addition to the alignment quality, the uv exposure energies applied to align LPP and to cross-link the LCP affect other layer properties including hardness, adhesion, surface energy, chemical resistance, photo- and thermal stability, *etc.* Therefore, the optimum exposure energies strongly dependent on the application and the materials which are in contact with LPP/LCP layers.

2.1.4 Drying and annealing

Drying temperatures highly depend on the solvents used for the LPP and LCP solutions. Additional pre- and/or post-baking steps improve material properties such as hardness, adhesion, surface energy, photo and thermal stability, alignment speed, alignment quality, *etc.*

2.2 Stability of LPP/LCP retarders

Because anisotropy is inherent to LCP materials, relaxation of the anisotropy does not occur upon thermal stress. As a consequence, LCP layers show high thermal stability of both mechanical and optical parameters. Furthermore, LCP retarders are extremely stable against exposure to intensive visible light and withstand high humidity. Consequently, LCP retarders are well suited for application in LCD projectors, where birefringent elements are required for contrast improvement. Data on the stability relative to heat, light, and humidity are shown in Fig. 1.

3 Multiple-layer retarders

Due to the contact-free photo-alignment method, multiple LPP/LCP layers can directly be coated on top of each other without the scratching defects which would be caused by alignment through rubbing. This eliminates additional laminating steps, which would be required if individual retarder films would have to be combined.

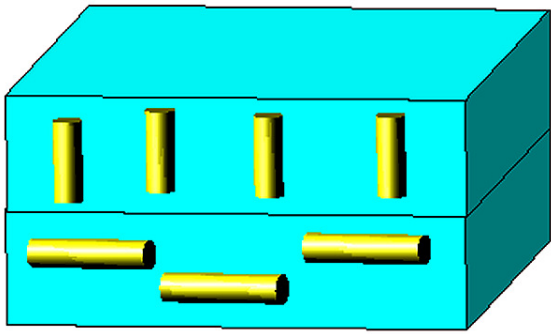


FIGURE 2 — Biaxial retarder consisting of a planar and a vertical-aligned LCP retarder.

3.1 Biaxial retarders

The most straightforward way to generate biaxial retarders from uniaxial LCP materials is to apply a c-axis retarder on top of a planar retarder (Fig. 2).

3.1.1 Viewing-angle improvement of polarizers

Biaxial retarders can be applied to improve the angular performance of crossed polarizers, which normally appear very bright for oblique angles at 45° azimuth to the polarizer axes. Biaxial retarders were proposed to drastically reduce this light leakage.⁶ Biaxial LPP/LCP retarders can directly be coated onto one of the polarizers which adds only about $2\mu\text{m}$ to the thickness of the polarizer. The strong depression of the light leakage is demonstrated in Fig. 3. Both the in-plane retardance of the planar LCP layer and the out-of-plane retardance of the vertical-aligned LCP layer were adjusted to 140 nm .

3.1.2 Angular independent retardance

The optical retardation of uniaxial retarders is strongly dependent on the incident angle of light. Contrary, the retardance of biaxial retarders is angular independent if $n_z = (n_x + n_y)/2$,⁷ where n_z is the refractive index along the thickness direction and n_x and n_y are the in-plane refractive indi-

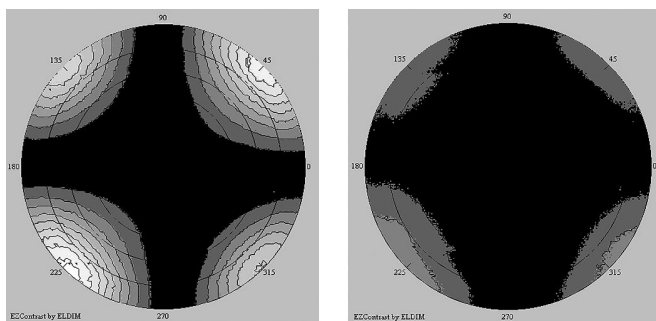


FIGURE 3 — Left: Light leakage through conventional crossed sheet polarizers. Right: Depression of the light leakage through crossed polarizers due to a biaxial retarder coated onto one of the polarizers.

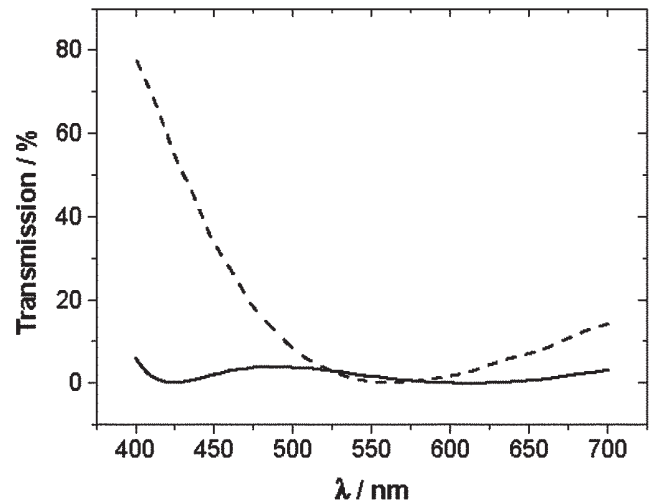


FIGURE 4 — Transmission of conventional (dashed) and achromatic (solid) LPP/LCP half-wave retarder between parallel polarizers.

ces. Since the biaxiality of stacked LPP/LCP retarders can be tailored by the thickness of both LCP layers, angularly-independent retarders of any retardation can easily be realized.

3.2 Achromatic retarders

Conventional retarders exhibit significant wavelength dependence of the phase retardance $\Delta nd/\lambda$. Consequently, every broad band application, where retarders are, for example, applied to rotate the plane of polarization of light or to convert linearly to circularly polarized light suffer from coloration and reduced brightness or contrast. Achromaticity can be achieved by stacking two planar retarders according to the method proposed by Pancharatnam.⁸ The efficiency of an achromatic half-wave retarder consisting of two photoaligned planar LCP retarders is shown in Fig. 4. Compared to conventional chromatic retarders, most of the light is blocked when arranged between parallel polarizers, which demonstrates the wavelength independent 90° rotation of the polarization plane.

4 Retarders with tilted optical axis

Tilt angles in LCP retarders can be generated by oblique LPUV exposure of the subjacent LPP layer. The resulting tilt angles mainly depend on material properties of both LPP and LCP as well as on exposure conditions. Because the LCP molecules interact differently at the air interface and at the LPP interface, tilt profiles along the LCP thickness direction can be induced. In general, the tilt angle of the optical axis may either increase, decrease, or is maintained from one side to the other (Fig. 5). All of these three cases can be realized by proper LPP- and LCP-material design. The evaluation of the tilt profile can be performed by ellipsometric measurements.⁹

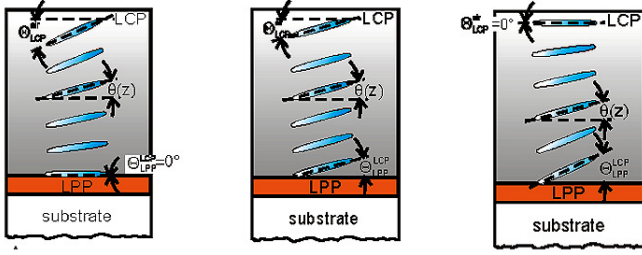


FIGURE 5 — Different tilt profiles in LPP-aligned LCP layers.

4.1 Dependence of tilt angles on exposure energies

LPP materials exhibit characteristic tilt-angle dependencies on the LPUV exposure energy. For example, Staralign™ 2110 generates an average tilt angle in a layer of LCP Opalva™ 2130 which reaches a maximum of 28° at LPUV energies of about 20 mJ/cm². Since the required LPUV energy often determines the maximum speed in roll-to-roll processing, it would be advantageous to shift the maximum tilt angle to lower LPUV doses. This can be achieved by subsequent exposure of the LPUV-exposed LPP layer to non-polarized uv light as shown by the results in Fig. 6. Because non-polarized uv light sources are already installed in many existing roll-to-roll coating lines, the production speed can easily be accelerated by at least a factor of 2 without additional investment in stronger LPUV light sources.

4.2 Viewing-angle enhancement of TN-LCDs

Retarders with tilted optical axis (o-plate) allow for the compensation of the residual off-axis retardation of TN-LCDs, thus enhancing the viewing-angle range. Extensive numerical modeling has to be used to find the optimum-wide-view film configuration for specific application requirements.

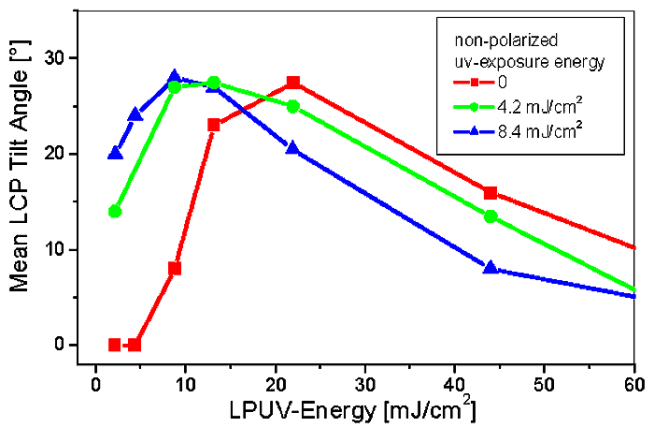


FIGURE 6 — Shift of the characteristic tilt angle vs. LPUV energy curve by additional exposure of the LPP layer to non-polarized uv light. Squares: only LPUV-exposed; circles: subsequent exposure to 4.2 mJ/cm² non-polarized uv light; triangles: subsequent exposure to 8.4 mJ/cm² non-polarized uv light; LPP material: Staralign™ 2110, LCP material: Opalva™ 2130.

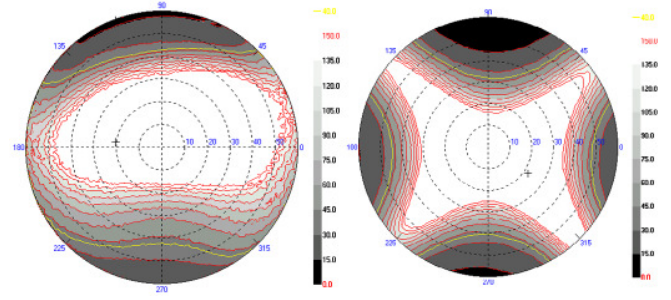


FIGURE 7 — Measured contrast coscopes of a TFT-TN display with the novel ROLIC compensator (left) and of a TFT-TN display with the Fuji WVF (right).

Recently, ROLIC has designed a wide-view film configuration for TFT-TN-LCDs which significantly enhances the horizontal viewing angle and avoids the yellow color shift, which is a well-known drawback of existing commercial products.¹⁰ A prototype of the novel wide-view film was realized with appropriate LPP and LCP materials.

The performance characteristics of a commercial TFT-TN LCD, to which the new wide-view film was attached, are demonstrated by measurements of the contrast coscope in Fig. 7 and of the color coordinates in Fig. 8.

5 Generation of alignment patterns

There are different ways to generate alignment patterns in LPP layers. Among them are the use of photomasks, alignment masters, laser scanning and synchronized rotation and/or movement of polarizer and substrate during uv exposure.

In the simplest method, the LPP layer is LPUV-exposed through a photomask in the first exposure step. For the subsequent LPUV exposure step without a photomask, the polarizer and substrate are rotated by the desired angle (typically 45° or 90°). The result is a patterned alignment layer exhibiting two different alignment directions. A LCP layer coated on top is then aligned according to the align-

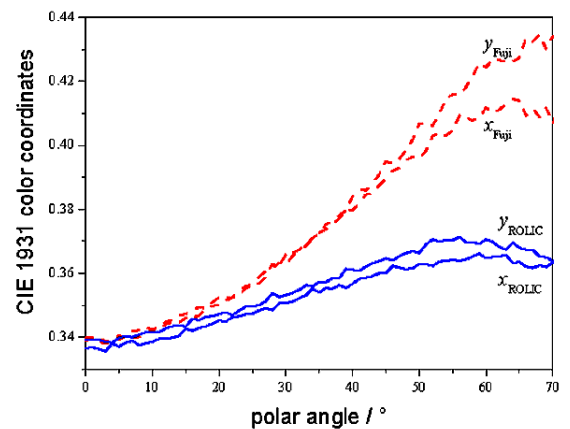


FIGURE 8 — Comparison of the measured horizontal color coordinates vs. polar angle for a gray level of 80%.

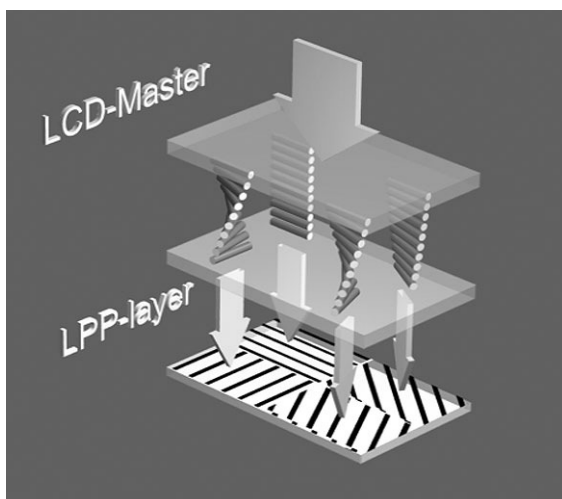


FIGURE 9 — Alignment master for the generation of complex alignment patterns in a single exposure step.

ment pattern stored in the LPP layer. Depending on the optical function of the LCP material, the resulting optical device may be a patterned retarder or a patterned polarizer. Both of them can, for example, be used to realize 3-D LCDs which distinguish the optical information for the left and right eye by the plane of polarization.

5.1 Exposure through alignment master

For alignment patterns with more than two different alignment directions, the number of photomasks and exposure steps increase correspondingly. An elegant method to generate complex alignment patterns in a single exposure step is the use of an alignment master.¹¹ The function of alignment masters is to provide LPUV light with a spatial variation of the polarization plane, which directly generates an alignment pattern when it hits the LPP layer. For example, a liquid-crystal cell in which one of the alignment layers is patterned can be used as an alignment master (Fig. 9). The polarization plane of the LPUV light, which is incident on the uniaxially aligned side of the cell, is then rotated due to wave guiding by the local angle between the alignment directions of upper and lower substrates. Figure 10 shows a

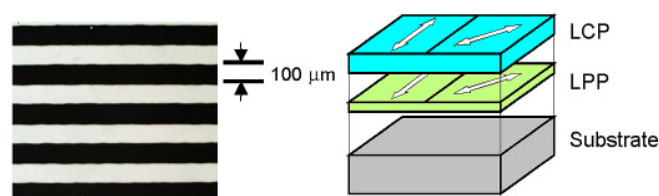


FIGURE 10 — Left: Patterned half-wave retarder between crossed polarizers. The alignment pattern was generated in a single exposure step via an alignment master. The optical axis direction in the black-and-white stripes differ by 45°. Right: Geometry of the patterned retarder.

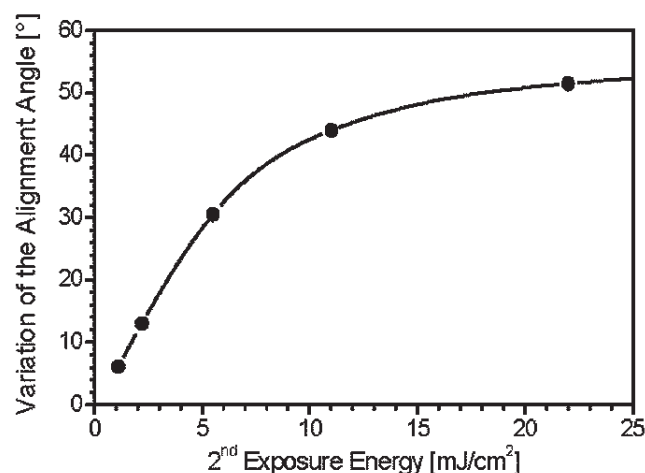


FIGURE 11 — Variation of the LPP-alignment direction upon second LPUV exposure. LPUV energy of first exposure: 5.5 mJ/cm². Angle between LPUV planes of first and second exposure: 60°.

patterned half-wave retarder, the alignment of which results from LPP exposure *via* an alignment master.

5.2 Continuous alignment variation

An alignment pattern with a continuous azimuthal variation of the alignment direction can for example be achieved by synchronized movement and rotation of LPP substrate and polarizer, respectively, while the LPP layer is exposed through a slit mask. However, only limited types of patterns can be generated by this technique, particularly for large-area LPUV exposure systems where it is quite complicated to rotate the polarizer.

Another technique, which makes use of the dynamics of the LPP photo-crosslinking process, allows the generation of complex patterns with continuous azimuthal alignment variation. The technique involves two exposure steps with different planes of LPUV polarization in each step. If the LPUV energy in the first exposure step is low enough so that the LPP material is not fully cross-linked, the second exposure causes a rotation of the alignment direction. Continuous spatial alignment variation is achieved by continuously changing the local LPUV exposure energy in the first and/or second exposure step. An example of the rotation of the alignment direction as a function of the second LPUV dose is shown in Fig. 11 for the commercial LPP material Staralign™ 2100. The maximum variation of the alignment angle correlates with the angle between the LPUV planes of the first and second exposures and can get very close to 90°.

Continuous local variation of the exposure energy can, for example, be achieved by moving the LPP substrate with varying speed during first or second LPUV exposure through a slit mask (Fig. 12). Complex patterns can be achieved if this process is applied in both exposure steps, but with different moving directions.

The complexity of patterns may be even higher if the LPP layer is exposed through an intensity mask, which

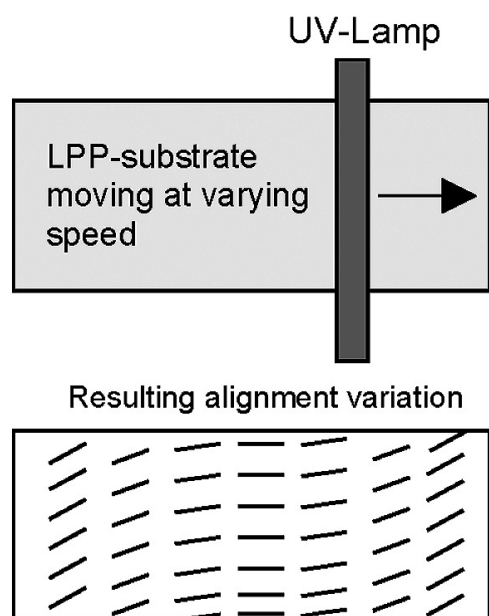


FIGURE 12 — Generation of a continuous alignment pattern by moving the LPP substrate at different velocities through the uv-illuminated area during one of the LPUV exposure steps.

directly provides a spatial energy distribution. For highly sophisticated applications, the use of an LCD as an intensity mask, laser writing, or time-sequential projection using digital micromirror devices (DMD) provide very high flexibility in pattern generation.

6 Conclusion

ROLIC's LPP/LCP technology provides a high level of flexibility in design and manufacturing of anisotropic layers. In particular, the process compatibility with flexible substrates and the possibility to adjust the optical axis to any azimuthal and polar angle makes it ideal for large-volume roll-to-roll production of retarders and wide-view films for LCDs. Stacking of LCP layers with individual optical functionality leads to compact films with new optical features.

References

- 1 M Bosma, *SID Intl Symp Digest Tech Papers* **98**, 850 (1998).
- 2 H Mori *et al*, *Jpn J Appl Phys* **36**, 143 (1997).
- 3 M Schadt, H Seiberle, and A Schuster, *Nature* **381**, 212 (1996).
- 4 M Schadt, H Seiberle, A Schuster, and S M Kelly, *Jpn J Appl Phys* **34**, L764 (1995).
- 5 M Schadt, H Seiberle, and F Moia, *Proc IDW '99*, 1013 (1999).
- 6 J Chen *et al*, *SID Intl Symp Digest Tech Papers* **98**, 315 (1998).
- 7 Y Fujimura *et al*, *SID Intl Symp Digest Tech Papers*, 739 (1991).
- 8 S Pancharatnam, *Proc Indian Acad Sci* **A44**, 247 (1956).
- 9 C Benecke *et al*, *Jpn J Appl Phys* **39**, 525 (2000).
- 10 T Bachel's *et al*, *Proc EuroDisplay '02*, 183 (2002).
- 11 H Seiberle, K Schmitt, and M Schadt, *Proc EuroDisplay '99*, 121 (1999).



Hubert Seiberle received his Ph.D. degree in polymer physics from the University of Freiburg, Germany, in 1989, where he investigated molecular motions in mixtures of liquid-crystal polymers. In 1990, he joined the liquid-crystal department of Dr. Schadt at Hoffmann-La Roche where he initially was involved in the design of nematic liquid-crystal mixtures. Since 1995, Dr. Seiberle has been working at ROLIC, a spin-off company of Hoffman-La Roche, which develops technology for liquid-crystal displays. At ROLIC, he is engaged in the development of photo-alignment and liquid-crystal polymer applications. Dr. Seiberle is head of the physics group at ROLIC.



Carsten Benecke received his Ph.D. degree in 1989 from the Technical University of Berlin in Germany where he performed research on the luminescence of semi-magnetic semiconductors. In 1990, he joined the liquid-crystal department of Dr. Schadt at Hoffmann-La Roche where he investigated wave guiding in Langmuir-Blodgett films for non-linear optics application. Currently, he is responsible for the design, development, and evaluation of novel photo-aligned crystal polymer material and related optical thin-film devices.



Thomas Bachel's graduated in condensed matter physics in 1997 from the Technical University Aachen in Germany. In 1999, he received his Ph.D. degree from the University of Basel, Switzerland, where he studied the thermochemical properties of metal and semiconductor clusters. He joined ROLIC in 2000 as a physics researcher. His current work comprises the development of various applications for polymerizable liquid-crystal materials.