Liquid-crystal diffraction gratings using polarization holography alignment techniques

Gregory P. Crawford
Division of Engineering, Brown University, Providence, Rhode Island 02912
and Department of Physics, Brown University, Providence, Rhode Island 02912

James N. Eakin
Division of Engineering, Brown University, Providence, Rhode Island 02912

Marc D. Radcliffe
3M Display and Graphics Laboratory, 3M Center, St. Paul, Minnesota 55144

Andrew Callan-Jones and Robert A. Pelcovits
Department of Physics, Brown University, Providence, Rhode Island 02912

(Received 18 July 2005; accepted 6 November 2005; published online 27 December 2005)

A method of patterning surfaces for liquid-crystal alignment using a polarization holography exposure on a linear photopolymerizable polymer alignment layer is demonstrated. Three configurations are demonstrated which include registered planar-periodic surface boundary conditions on both surfaces (true polarization gratings), planar-periodic and uniform planar surface boundary conditions, and planar-periodic and homeotropic boundary conditions. Two-dimensional polarization gratings are also demonstrated by orientating planar-periodic alignment layers orthogonally. Passive polarization gratings are also demonstrated using reactive mesogens to capture the periodic order indefinitely. The underlying structure of the configuration is discussed, including the nature of their switching transition (threshold or thresholdless), for all three configurations. A simple phenomenological model is presented to describe the Freedericksz transition for the registered planar-periodic boundary condition case. © 2005 American Institute of Physics. [DOI: 10.1063/1.2146075]

I. INTRODUCTION

The patterning of surfaces for aligning liquid-crystal materials has received a significant amount of attention by scientists and engineers to create a plethora of hybrid geometries and multidomain configurations for wide-angle-viewing liquid-crystal displays, projection displays, and photonic devices for optical communications. A number of materials and techniques have been employed using polymer alignment layers or photosensitive materials to create multidomain configurations for liquid-crystal materials exhibiting unique electro-optic behavior compared to the well-described single-domain liquid-crystal alignment typically imposed by a polyimide layer. In this contribution, we focus on planar-periodic boundary to create various types of optical gratings including polarization gratings, which are diffractive optical elements with an anisotropic periodic index profile.1–6 Although the disclosed polarization gratings are “thin” gratings, the thin-screen approximation predicts high diffraction into the first diffraction orders making them attractive for optical devices.7,8 These gratings may have numerous applications in photonic systems and displays, including highly efficient projection displays, wide-viewing and direct-view displays, polarized beam splitting devices, multiplexing, and polarization dispersion applications. In this paper we investigate the structure of a number of gratings formed using polarization holography for liquid-crystal alignment.

The field of patterning surface alignment layers for liquid crystals is vast. Varghese and co-workers have demonstrated a four-domain liquid-crystal alignment using a microrubbing technique on homeotropic polyimide that allows for expanded viewing angles in full-color displays.5,10 Chen et al. have shown a hybrid liquid-crystal configuration (combining planar and twisted regions) using photolithography to create a double-rubbed polyimide layer for a high-efficiency polarization-independent grating.11 Honma and co-workers have demonstrated liquid-crystal gratings consisting of multidomain alignment regions fabricated through a microrubbing technique.12–14 Versteeg et al. have applied direct laser-writing methods to melt polyimide regions, creating multidomain configurations of planar and twisted alignments.15 Wen et al. have shown a dual-domain polarization grating configuration created from scribing a polyimide layer with an atomic force microscope tip that produces right- and left-handed twisted regions.2 Komitov et al. have demonstrated a uniform lying helix (ULH) alignment for short pitch cholesteric liquid crystals using periodic-planar and homeotropic anchoring conditions.3 Additionally, Rosenblatt and co-workers have published extensive results on anchoring effects, surface roughness, and electro-optic dynamics associated with liquid-crystal alignment on atomic-force-microscopy-scribed polyimide surfaces.4–6

In addition to modifying polyimide-coated surfaces to
manipulate the surface alignment of liquid crystals, photosensitive layers have been demonstrated as noncontact methods for establishing multidomain configurations. Schadt et al. created a four-domain twisted nematic display that exhibits wide-angle viewing using a coumarin photopolymer layer with polarized ultraviolet irradiation. Gibbons et al. have demonstrated a method of creating a liquid-crystal grating by exposing a dye-doped polymer layer to an ultraviolet interference pattern to create periodic regions of planar and twisted alignments. Lee et al. have functionalized a self-assembled monolayer (SAM) to create hybrid regions of planar and bend alignment upon irradiation of nonpolarized ultraviolet light.

We focus on creating periodic boundary conditions of liquid crystals using a linear photopolymerizable polymer (LPP) surface layer exposed to a polarization interference pattern. Unlike amplitude holography, which uses two interfering beams of the same polarization to create an intensity modulation as shown in Fig. 1(a), polarization holography uses two interfering beams of orthogonal linear or orthogonal circular polarization to create polarization modulations as shown in Figs. 1(b) and 1(c), respectively. When creating polarization interference patterns, as in Figs. 1(b) and 1(c), the intensity of the incident light is constant, or nearly constant for certain exposures. The pitch of the interference pattern, $\Lambda$, is also defined in Fig. 1. Although we show in Fig. 1(c) that no intensity modulation exists, there is in fact one. Cloutier et al. have shown that if the exposure angles are small, as in the case of this publication, the intensity interference pattern is very weak. In this publication we employ the arrangement shown in Fig. 1(c). The holographic exposure of the LPP using the configuration polymerizes the material in the incident polarization direction and creates planar-periodic boundary conditions of liquid crystal in the plane of the substrate. The photochemistry mechanism to achieve such alignment is analogous to that reported by Schadt et al. Using this holographic polarization alignment pattern shown in Fig. 1(c) on a pair of substrates, we have created a rotating planar alignment configuration of liquid crystal registered between the top and bottom substrates. Furthermore, we have combined the holographic polarization alignment pattern with conventional alignment layers to enforce uniform planar or homeotropic conditions on the opposite substrate.

II. MATERIAL PREPARATION

Linear photopolymerizable polymer (LPP F301), commercially available from ROLIC (ROLIC Ltd., Allschwil, Switzerland), was used as a photosensitive alignment layer. It was spin coated on indium tin oxide (ITO)-coated glass substrates to achieve a uniform film thickness of $\sim 100$ nm. After spin coating, the substrates were heated on a hot plate at 150 °C for 15 min to remove the residual solvent from the alignment layer. Cell configurations were prepared by sandwiching two substrates together with the alignment layers facing each other; a cell gap of 5 $\mu$m was maintained by glass fiber spacers (EM Industries, Hawthorne, NY). A single holographic exposure using an Ar$^+$ laser (Innova 70, Coherent, Santa Clara, CA) with an emission line of $\lambda=351$ nm to create a two-beam transmission interference pattern ensued; the beams had equal intensity and orthogonal circular polarizations to create the spatially rotating linear polarization pattern. After exposure to the interference pattern, liquid-crystalline material was infiltrated into the empty cell by capillary action at an elevated temperature and cooled to room temperature under ambient conditions. By sandwiching the substrates together prior to the holographic exposure, a liquid-crystal grating structure was created with periodic boundary conditions registered on both substrates.

Liquid-crystal gratings were also created using hybrid boundary conditions. These cell configurations were prepared using one LPP-polarization-patterned ($\Lambda=7.5$ $\mu$m) ITO glass substrate and one polyimide-coated ITO glass substrate. Commercially available polyimide precursors were used (Nissan SE-2170 and Nissan SE12-11, Brewer Science, Rolla, MO) to promote planar and homeotropic alignments.
respectively. The planar polyimide material was spin coated and then cured in an oven to achieve a film thickness of \( \sim 200 \text{ nm} \). After curing, the planar polyimide-coated substrate was mechanically rubbed with a velvet wheel to achieve uniform planar alignment in the rubbing direction. The homeotropic polyimide material was spin coated and then cured in an oven to achieve a uniform film thickness of \( \sim 100 \text{ nm} \). Following the exposure of the LPP-coated substrate to the polarization interference pattern, the hybrid cell fabrication was completed by sandwiching the LPP-polarization-patterned ITO glass and polyimide-coated ITO glass separated by 5 \( \mu \text{m} \) glass fiber spacers. In the hybrid configurations it was not necessary to sandwich both substrates together prior to the holographic exposure since the boundary conditions on the two substrates did not require registration. The liquid crystal (MLC 6292-100, EM Industries, Hawthorne, NY) was filled by capillary action into the empty cell at an elevated temperature and cooled to room temperature under ambient conditions.

Planar-periodic boundary conditions were also used to align reactive mesogen materials. These grating configurations were prepared by using one LPP-polarization-patterned glass substrate with a liquid-crystal polymer (LCP) (LCP CB 483, ROLIC Ltd., Allschwil, Switzerland) coated on the substrate using a Meyer rod after the holographic exposure. Prior to polymerization the LCP material exhibits similar surface alignment characteristics as a low molecular weight liquid-crystal material. Polymerization with UV light permanently “captures” the order of the liquid-crystal phase in a polymer form. This particular reactive mesogen was chosen since it was developed to work with the LPP material.

In summary, several processes to fabricate polarization gratings were disclosed using both low molecular weight liquid crystal and liquid-crystal polymer. The gratings formed using registered planar-periodic boundary conditions will, in principle, exhibit a larger refractive index modulation due to the continuous spatial rotation of the alignment. This may enhance electro-optic properties as well as functionalities for device potential. The other hybrid configurations disclosed in this work will exhibit a lower refractive index modulation caused by the constraints of the boundary conditions but may be applicable for specific device applications if larger refractive index modulations are not necessary.

III. PHASE HOLOGRAPHY

In 1984, Nikolova and Todorov reported on the polarization interference pattern of two coherent beams having opposite polarizations on a photorecordable material.\(^{19}\) In one example, a polarization interference pattern was recorded using two equal intensity, orthogonal, linear-polarized beams where the resulting light field \( E \) was described by 
\[
E = \frac{E}{E} \cos \delta \exp \frac{i E}{E} \sin \delta,
\]
where \( \delta \) represents the phase shift between the two beams and is defined as 
\[
\delta = 2(\xi)\lambda \sin \Theta,
\]
which is periodic along \( \xi \); \( \lambda \) is the wavelength and \( \Theta \) is the half angle between the two interfering beams. This interference pattern resulted in an uniform intensity with a periodic polarization pattern that varied spatially from \( 2\delta = 0 \rightarrow 2 \pi \) to create linear, elliptical, and circular polarization regions. The second example of polarization holography recorded the interference pattern generated from right- and left-handed circular-polarized light where the light field \( E \) was described by 
\[
E = \frac{E}{E} \cos \delta \exp \frac{i E}{E} \sin \delta.
\]
This type of polarization interference generated a uniform intensity with a spatially rotating and periodic linear polarization pattern from \( 2\delta = 0 \rightarrow 2 \pi \).

These two types of polarization holography have been applied to other photosensitive materials to create patterned alignment regions in azobenzenes,\(^{22}\) dye-doped liquid crystals,\(^{23}\) polymer liquid crystals,\(^{24}\) and polymer-dispersed liquid crystals.\(^{25-27}\) In our situation, the continuously varying polarization “imprint” on the alignment layers is most useful for aligning liquid crystal [Fig. 1(c)]. The polarization pattern shown in Fig. 1(b), where the polarization pattern transforms from linear to elliptical to circular, is less attractive for liquid crystal alignment. It is unclear how the liquid crystal would align on such a surface.

IV. POLARIZATION GRATINGS

A. Registered planar-periodic boundary conditions

An optical polarizing microscopy image of the polarization grating (\( \Lambda = 7.5 \mu \text{m} \)) between crossed polarizers is shown in Fig. 2, where \( \mathbf{n} \) is defined as the liquid-crystal director. The anchoring conditions of the LPP alignment layer create a uniform grating configuration without defects in zero field. The dark grating lines indicate regions where the liquid crystal is oriented at 0\(^\circ\) or 90\(^\circ\) with respect to the polarizer (P) or analyzer (A) while the lighter regions indicate where the liquid crystal is aligned at some other angle between the polarizer and analyzer. A schematic illustration of the top and cross-sectional views showing the anchoring alignment imposed by the polarization-patterned surfaces is represented in Fig. 2. Because the top and bottom patterned surfaces are registered during exposure, the planar alignment propagates unperturbed through the depth of the cell to create uniform alignment throughout the volume of the cell. As will be shown later, the propagation of the liquid-crystal
alignment through the cell can only happen if the grating pitch is comparable or larger than the cell gap \( (\Lambda \geq d) \).

**B. Hybrid configured gratings**

1. **Planar periodic-planar boundary conditions**

The hybrid configuration of the planar substrate and LPP-polarization-patterned substrate resulted in a hybrid structure of alternating regions of planar and twisted alignments corresponding to the grating pitch (\( \Lambda \)). The pitch (\( \Lambda \)) of the planar-periodic boundary condition is \( \Lambda = 7.5 \mu \text{m} \). An optical polarizing microscopy image taken between crossed polarizers of this hybrid configuration is shown in Fig. 3. Regions where the polarization direction of the LPP-coated substrates were aligned parallel to the rubbing direction of the polyimide-coated substrate resulted in a planar alignment of the liquid crystal propagating through the depth of the cell. This can be seen as thick, dark grating lines in the optical polarizing microscopy image in Fig. 3. Schematic illustrations showing the surface and cross-sectional hybrid alignment views are also presented in Fig. 3. Regions where the polarization direction of the LPP-coated substrates were aligned parallel to the rubbing direction of the polyimide-coated substrate resulted in a planar alignment of the liquid crystal through the depth of the cell. This can be seen as thick, dark grating lines in the optical polarizing microscopy image in Fig. 3. Schematic illustrations showing the surface and cross-sectional hybrid alignment views are also presented in Fig. 3. Regions where the polarization direction of the LPP-coated substrates were aligned parallel to the rubbing direction of the polyimide-coated substrates, resulting in a continuous twisted alignment (\( <90^\circ \)) of the liquid crystal through the depth of the cell. This can be seen as light grey grating lines in the optical polarizing microscopy image due to the adiabatic waveguiding of light in the twist configuration. Because of the continuous and rotating nature of the polarization pattern, the amount of twist spatially varies across the grating pitch between 0° and 90°, as observed by the grey levels in the optical polarizing microscopy image. Defect lines are also present in this configuration due to a competition between twisting directions in the cell. The planar-periodic boundary conditions imposed by the polarization-patterned alignment layer and polyimide alignment layer create an inherent twist sense as described earlier. These acute twisted regions exhibit both right-handed and left-handed twisting directions, as shown in Fig. 3. These right- and left-handed twisted regions “collide” in the region where the liquid-crystal alignment imposed by the planar polyimide boundary condition is orthogonal to the liquid-crystal alignment imposed by the polarization-pattern boundary condition. Instead of forming a complete 90° twist, a sharp defect line is observed as shown in the optical polarizing microscopy image in Fig. 3. In Fig. 3, the defect line appears as a dark line within a white grating line. Because of competition between right-handed and left-handed twisted domains, it is not possible to suppress the appearance of the defect line for this particular hybrid geometry.

2. **Planar periodic-homeotropic boundary conditions**

The hybrid configuration from the homeotropic polyimide boundary conditions on one substrate and planar-periodic boundary conditions resulted in a structure that combined periodic regions of different bend geometries corresponding to the grating pitch (\( \Lambda \)). This is a defect-free configuration because the periodic regions do not generate a frustrated liquid-crystal alignment within the cell. An optical polarizing microscopy image of this structure between crossed polarizers is shown in Fig. 4. These bend structures appear as a grating texture due to the periodic and spatially rotating planar alignment established by the planar-periodic boundary conditions. The darker grating lines in the microscope image in Fig. 4 represent bend regions where the planar-aligned liquid crystal is at 0° or 90° with respect to the polarizer or analyzer. Lighter grey grating lines represent bend regions where the planar-aligned liquid crystal is at some other angle with respect to the polarizer and analyzer. Schematic illustrations of the alignment surfaces and cross-sectional views of the cell are also visualized in Fig. 4.
C. Comparisons

1. Polarization dependence

Polarization dependence of the hybrid configurations was investigated using a helium-neon laser to illuminate the patterned area of the cell at normal incidence while a photodetector connected to a wideband amplifier and digital multimeter recorded the first-order diffracted intensity. The first-order diffraction intensity was measured as a function of the incident linear polarization in 10° increments, with 0° representing the incident polarization perpendicular to the grating vector and 90° representing the incident polarization parallel to the grating vector. Plots of the first-order diffraction intensity as a function of the incident linear polarization direction for the three cell configurations are shown in Fig. 5. Polarization results for the polarization-patterned surfaces show a slight dependence. The plot in Fig. 5 shows the increase in diffracted intensity as the linear polarization is rotated from the incident s polarization (0°) to the p polarization (90°). Overall the change in intensity is ~3:1 which demonstrates that the polarization grating is slightly polarization dependent. This slight polarization dependence may be attributed to the existence of a small amplitude grating during exposure. This has been seen in other polarization grating technologies using azodye-doped photoresist.\textsuperscript{20} The first-order diffraction efficiency was measured for a single patterned substrate (without any liquid crystal) for the incident s and p polarizations. The first-order diffraction efficiency for the incident s polarization was ~0.06% and ~0.08% for the p polarization, confirming that the holographic exposure creates a small refractive index modulation in the LPP film itself.

In comparison, the hybrid configuration formed from the polarization-patterned LPP and planar polyimide shows no dependence on the incident polarization as it is rotated from 0° to 90°, as shown in Fig. 5. The diffracted intensity remains constant, thus creating a completely polarization-independent grating. This is attributed to a refractive index modulation between the planar and twisted alignment regions independent of the incident polarization direction. Regardless of the incident linear polarization direction, the refractive index modulation will be the difference between the extraordinary \(n_x(=1.5991)\) and ordinary \(n_o(=1.4845)\) indices of refraction for the liquid crystal due to the periodic and orthogonal alignment configuration established by the boundary conditions \(n_x-n_o=0.1146\), MLC-6292.\textsuperscript{20}

The hybrid configuration formed from the polarization-patterned LPP and homeotropic polyimide also shows no dependence on the polarization as it is rotated from 0° to 90°, as presented in Fig. 5. The measured diffracted intensity is less than the hybrid configuration using planar polyimide, due to a lower refractive index modulation in the grating caused by the bend configuration. For the incident s polarization or \(p\) polarization the index modulation between alternating bend configurations is the difference between an effective \(n_{\text{eff}}\) index and the ordinary \(n_o\) index for the liquid crystal. The effective index \(n_{\text{eff}}\) can be defined as

\[
n_{\text{eff}} = \frac{-1}{d} \int_0^d \frac{n_o n_x}{n_x^2 \cos(\theta(z)) + n_o^2 \sin(\theta(z))} dz,
\]

where \(d\) is the thickness of the cell, \(n_o\) and \(n_x\) are the ordinary and extraordinary indices of refraction for the liquid crystal, and \(\theta(z)\) is the angle between the nematic director and the x-y plane. The effective index can be approximated as \(~\sqrt{n_x n_o}\) to give \(n_{\text{eff}}=1.5407\), which supports a lower index modulation \(n_{\text{eff}}-n_o=0.0562\) in the periodic-bend configurations.

D. Electro-optic switching

1. Registered planar-periodic boundary conditions

The electro-optic switching behavior was investigated for the three hybrid configurations with the same grating pitch and thickness \((\Lambda=7.5 \, \mu\text{m}, \, d=5 \, \mu\text{m})\). A 1 kHz square wave was applied across the sample while a helium-neon laser probed the sample and a photodetector measured the change in the first-order diffraction intensity as a function of the voltage. A plot of the first-order diffraction versus voltage curve for the cell configuration with periodic boundary conditions is shown in Fig. 6, having a threshold voltage of...
1.4 V. The shape of the first-order diffraction versus voltage curve indicates, as voltage is increased to 2 V, that the liquid-crystal alignment in the field essentially erases the index modulation. A 10% first-order diffraction peak persists slightly above 2 V. After 2 V, the diffraction intensity begins to grow again. As can be observed in the microscope images (see inset Fig. 6), the grating again becomes more visually prominent at 3 V before disappearing at 10 V. The phenomenon observed in Fig. 6 is speculated to be the result of reverse tilt disclinations arising due to the absence of pretilt. At 3 V a very strong grating reappears with distinct boundaries between the adjacent domains. We are currently in the process of modeling these structures using Monte Carlo simulations. The calculations involving these structures are not trivial, especially when surface tilt angles are included, and are the subject of a future paper.28 Response times (on and off) were also measured using a 10 V modulated square wave applied across the sample; on time was measured as 4 ms (±0.2 ms) with a relaxation time of 8 ms (±1 ms). The pitch-to-cell gap ratio was not optimized in this sample so the diffraction efficiency of the first-order peak was only ∼8%. Using the thin-screen approximation, the pitch-to-cell gap ratio can be optimized to achieve high diffraction efficiency.7,8

2. Planar periodic-planar boundary conditions

The first-order diffraction versus voltage curve for the periodic-planar (periodic-twist) configuration is shown in Fig. 7. The planar periodic-planar configuration has a threshold voltage of ∼1 V. As voltage is increased to 2 V the liquid crystal begins to untwist and align in the field; the increased diffraction intensity is attributed to a larger index modulation for this partially aligned state. The planar regions of this configuration have a lower threshold voltage than the twisted regions. Therefore, as observed under the microscope (see inset to Fig. 7), the planar regions begin to switch before the twisted regions do, thereby increasing the index modulation initially. During this voltage range between 0 and ∼2 V the effective index of the planar regions decreases while the twisted regions remain the same. When the twisted regions also begin to align, the index modulation between the planar and twisted regions starts to decrease thereby reducing the diffraction intensity as observed in Fig. 7. The on time was measured at 5 ms (±0.5 ms) with a relaxation time of 20 ms (±1 ms). The pitch-to-cell gap ratio was not optimized in this cell so the diffraction efficiency was ∼3.5%. To achieve high diffraction efficiency the pitch-to-cell gap ratio can be optimized using the thin-screen approximation.7,8

3. Planar periodic-homeotropic boundary conditions

The first-order diffraction versus voltage curve for the periodic-homeotropic (periodic-bend) configuration presented in Fig. 8 resulted in a thresholdless voltage behavior. The thresholdless behavior results from an out-of-plane liquid-crystal alignment established from the homeotropic substrate. As the electric field is applied across the sample, the liquid crystal is further aligned in the direction of the field, decreasing the index modulation. Optical polarizing microscopy images of the planar periodic-homeotropic sample shown in the inset of Fig. 8 verify the alignment of the liquid crystal in the presence of an applied field. The on time was measured at 3 ms (±0.5 ms) with a relaxation time of 15 ms (±1 ms). The diffraction efficiency was ∼1% and was attributed to the lower refractive index modulation across the grating for this particular configuration as discussed in Sec. IV C. The pitch-to-cell gap ratio was not optimized for this cell configuration.

V. LIQUID-CRYSTAL POLYMER GRATINGS

It is a natural step to also make passive grating structures using reactive mesogen materials which can also be aligned by the planar-periodic boundary conditions. The planar-periodic order can then be captured by photopolymerization. A schematic representation of the fabrication steps used to create the passive polarization grating is shown in Fig. 9(a). The planar-periodic alignment layer was created as described earlier using LPP and the polarization holography. A liquid-crystal polymer (LCP CB 853) was coated down on the substrate with a Meyer bar and subsequently polymerized in a nitrogen-rich environment with a UV light. An optical polarizing microscopy image of the liquid-crystal polymer (LCP) grating between crossed polarizers is shown in Fig. 9(b). The
grating structure of this image indicates that the polarization-patterned alignment of the LPP creates the same patterned arrangement in the polymerized LCP layer as it does for low molecular weight liquid crystal disclosed in the earlier sections. The dark regions of the grating correspond to LCP alignment parallel or perpendicular to the polarizer or analyzer; the lighter regions correspond to the LCP alignment at some angle with respect to the polarizer or analyzer. Since this LCP layer is completely polymerized on a single substrate, it cannot be electrically switched; it can only act as a passive diffractive optical element.

Inspection of the microscope photograph in Fig. 9(b) shows a significant number of defects. These defects are attributed to the top air surface, which does not enforce planar anchoring conditions. One can of course use reactive mesogen materials and two aligning surfaces as described in Sec. IV A, to achieve near perfect alignment as observed in low molecular weight systems. In this contribution, we simply chose to use the liquid-crystal polymer material obtained from ROLIC since it was developed to work with the ROLIC LPP alignment layer. Other reactive mesogens should also work.29

VI. TWO-DIMENSIONAL STRUCTURES

Two-dimensional structures were created by exposing LPP-coated ITO glass substrates to the orthogonal circular polarization interference pattern (A=15 μm) individually and assembling an empty cell configuration with one polarization-patterned substrate rotated 90° with respect to the other. The resulting configuration is a two-dimensional array of planar and various twists (≤90°) of liquid-crystal alignment created by the overlapping planar-periodic patterned surfaces. A schematic illustration showing the two-dimensional alignment array between parallel and crossed polarizers is presented in Figs. 10(a) and 10(b). The dashed arrows represent the liquid-crystal director alignment on the top substrate while the solid arrows represent the liquid-crystal director alignment on the bottom substrate. Regions with parallel arrows indicate planar alignment propagating through the depth of the cell. Regions with orthogonal arrows represent 90° twisted configurations. Finally, regions with arrow directions between 0° and 90° represent twisted configurations coinciding with the angle between the two arrows. When this configuration is placed between parallel polarizers of an optical microscope as shown by the illustration in Fig. 10(a) and optical polarizing microscopy image in Fig. 10(c), the light regions coincide with planar alignment that is parallel or perpendicular to the polarizer or analyzer. Regions producing a twist <90° can be seen as grey regions in the illustration in Fig. 10(a) and optical polarizing microscopy image in Fig. 10(b). Likewise, 90° twisted regions appear as dark regions between parallel polarizers in Figs. 10(a) and 10(b). As the polarizer and analyzer are crossed, a contrast inversion appears spatially in the sample. The planar-aligned regions that were light regions between parallel polarizers appear as dark regions; the 90° twisted regions that were dark between parallel polarizers appear as light regions. This is observed in optical polarizing microscopy images in Figs. 10(c) and 10(d). An example of two-dimensional visible diffraction pattern created from this grating is shown in Fig. 10(e) using a helium-neon laser (λ=633 nm). The diffraction efficiency from one first-order diffraction peak in Fig. 10 was measured to be ~8%. 

FIG. 9. Optical polarizing microscopy image of a liquid-crystal polymer (LCP) layer polymerized on a LPP-polarization-patterned substrate.

FIG. 10. Schematic illustrations showing two-dimensional alignment array between parallel and crossed polarizers of an optical microscope.
time was measured at 5 ms (±0.1 ms) with a relaxation time of 15 ms (±1 ms). The response time measurements for the two-dimensional gratings are comparable to the one-dimensional grating response times.

In all cases (one-dimensional and two-dimensional gratings) the electro-optic response times were roughly equivalent to those of liquid-crystal cells with uniform alignment. The feature sizes of our periodic alignment condition were on the order of ~7 μm, which is relatively large. Larger deviations in switching times are expected for smaller feature sizes (i.e., <1 μm) when elastic distortion effects can play a bigger role.

VII. THEORETICAL CONSIDERATION

A. Periodic boundary conditions

Polarization gratings were created with different pitch lengths (Λ=5, 7.5, 10, 12.5, and 15 μm) with uniform 5 μm cell gaps to investigate and model the threshold voltage behavior associated with the polarization pattern. Since the grating pitch Λ is comparable to or larger than the cell gap, the periodic boundary conditions imposed on the surface alignment layer completely propagate through the depth of the cell to minimize the free energy. In order to derive an expression for the free energy of this configuration, we impose strong anchoring at the alignment surfaces with a zero tilt angle and use a single elastic constant approximation.

This has been shown in an earlier publication:  

$$ F = K \int_{-\Lambda/2}^{\Lambda/2} dy \int_0^d \left( \sin^2 \theta \left( \frac{d\phi}{dy} \right)^2 + \left( \frac{d\theta}{dz} \right)^2 \right) dy + 2 \sin^2 \theta \sin \phi \frac{d\phi}{dy} \sin \theta \frac{d\theta}{dz} - \frac{\Delta \varepsilon \varepsilon_0}{K} (E \cos \theta)^2, \quad (2) $$

where $F$ is the free energy, $\Lambda$ is the grating pitch, $K$ is the one-elastic-constant approximation, $d$ is the cell gap, $\theta$ is the angle between the director $n$ and the $z$ axis, $\phi$ is the angle between the director $n$ and the $y$ axis, $\Delta \varepsilon$ is the dielectric anisotropy of the liquid crystal ($\Delta \varepsilon = +6.9$ for MLC-6292-100), $\varepsilon_0$ is the permittivity of free space, and $E$ is the external electric field. Since the director varies spatially across the sample, the director profile is given by $n = [\sin(\pi y/\Lambda), \cos(\pi y/\Lambda), 0]$ through the depth of the sample. By minimizing the free-energy equation and taking into account the boundary conditions imposed at the photoalignment surfaces, the threshold voltage becomes:

$$ V_{th} = \pi \sqrt{\frac{K}{\Delta \varepsilon \varepsilon_0} \left( 1 - \frac{d^2}{\Lambda^2} \right)}, \quad (3) $$

where the threshold voltage is a function of grating pitch $\Lambda$ and cell gap $d$. In cases where the grating pitch $\Lambda$ is equal to the cell gap $d$, the model predicts a thresholdless voltage. The model and experimental results for threshold voltage as a function of grating pitch for $d=5$ μm is shown in Fig. 12. The inset of Fig. 12 shows the visible diffraction patterns for $\Lambda=5, 7.5, 10, 12.5,$ and 15 μm.

The boundary conditions imposed at the surfaces create nonuniform director orientation through the depth of the cell in the zero-electric-field state. By applying an electric field

---

**FIG. 10.** (a) A schematic representation of the liquid-crystal director orientation in a two-dimensional grating between parallel polarizers, arrows indicate director orientation on the alignment surfaces. (b) Schematic representation of the liquid-crystal director orientation in a two-dimensional grating between crossed polarizers. (c) Optical polarizing microscopy image of a two-dimensional grating between parallel polarizers. (d) Optical polarizing microscopy image of a two-dimensional grating between crossed polarizers. (e) Visible diffraction pattern created from two-dimensional polarization grating.

**FIG. 11.** Plots of transmission vs voltage for a two-dimensional planar-periodic cell configuration ($\Lambda=15$ μm, $d=5$ μm); the threshold voltage was 0.5 V.
across the sample (above the threshold voltage), the director is uniformly aligned in the direction of the field. This creates a gain in the elastic energy due to the electric and nonelectric terms. Note that this does not happen in the ordinary Fredericksz transition where at zero electric field the sample is uniformly aligned; by applying an electric field (above the threshold voltage) the director orientation remains uniformly aligned resulting in zero gain in the elastic free energy.

The periodic surface alignment created through the holographic exposure acts as an effective field with a magnitude \((d\phi/dy)^2 \approx 1/\Lambda^2\). For a grating pitch \(\Lambda\) comparable to the cell gap \(d\), it is energetically favorable to have a Fredericksz transition to a uniformly aligned state at zero electric field to minimize the cost in elastic energy.

In cases where the grating pitch \(\Lambda\) is less than the cell gap \(d\), the model breaks down. This is due to the prohibitively high cost in elastic energy for the periodic director orientation to propagate through the depth of the cell. In this case, the director orientation in the interior part of the cell will not correspond to the director orientation at the surfaces.

B. Hybrid boundary conditions

1. Planar periodic-planar boundary conditions

The planar periodic-planar alignment configuration formed using a planar-periodic patterned surface and planar polyimide-coated surface exhibits a lower threshold voltage behavior than the one predicted and observed in the periodic boundary alignment configuration. In this hybrid configuration, the periodic regions of planar and variable twist create an additional amount of elastic free energy in the zero-electric-field state that is not present in the periodic boundary alignment condition. As we described for the initial periodic boundary condition state, applying an electric field across the sample uniformly aligns the director, causing a decrease in the elastic energy due to the electric and nonelectric terms. We believe the additional twist component of this configuration attributes to a lower threshold voltage behavior.

2. Planar periodic-homeotropic boundary conditions

The periodic-bend configuration that is formed using a planar-periodic patterned surface and a homeotropic polyimide-coated surface exhibits a thresholdless voltage behavior in the case when the grating pitch \(\Lambda\) is larger than the cell gap \(d\). This behavior is described simply by virtue of the boundary conditions imposed at \(z=0\) and \(z=d\), where the director field must bend out of the \(x-y\) plane between the substrates, as depicted in Fig. 4. As a result, applying a field perpendicular to the substrates, while causing further bending out of the \(x-y\) plane, does not qualitatively change the zero-field arrangement (which is independent of the grating pitch \(\Lambda\) and thickness \(d\)). In this case, a thresholdless voltage response is expected and observed, since even a strong field only changes the degree of bend rather than the type of director configuration.

VIII. APPLICATIONS

There are potentially many applications for the periodic grating structures discussed in this paper, in particular. In this contribution we focused on the underlying physics of unique liquid-crystal configurations created using periodic alignment layers. We did not optimize these samples for any particular applications. Rather, we focused on the underlying physical phenomena of the periodic structure. There are several technologies in the literature that produce electrically switchable gratings. Holographic polymer-dispersed liquid crystals (H-PDLCS) are created through a photoinduced diffusion process where alternating layers of liquid-crystal-rich and polymer-rich layers are formed.\(^{30-32}\) Because of the nature of the phase separation,\(^{33,34}\) the switching voltages can be quite high due to the polymer confinement. Another approach for polarization grating technology is to add azodye compounds in polymers to create an index modulation.\(^{20,35}\) However, azodye-doped polymers tend to produce very small diffraction efficiencies of a few percent.\(^{20,35}\) The grating approach presented in this contribution overcomes these few issues since it can be switched at relatively low voltages and the aligned liquid crystal can produce large index modulations.

The potential to create highly efficient polarization gratings has been established by various groups.\(^{7,8,36,37}\) Sarkissian and co-workers have demonstrated that a thin-film approximation results in high diffraction of normally incident light into the first orders. Highly efficient thin gratings have potential applications in the display,\(^{38}\) optical communication,\(^{39}\) and optical security industries.\(^{40}\) Example applications that may capitalize on the polarization gratings discussed in this paper include projection,\(^{7,8}\) beam splitting devices,\(^{41}\) polarization dispersion control,\(^{42}\) and compensation devices.\(^{43}\) It is a subject of future study to optimize the diffraction efficiency within the framework of thin-screen approximation and to target specific applications.

IX. CONCLUSIONS

In summary, we have disclosed a number of switchable liquid-crystal gratings based on planar-periodic boundary conditions achieved through holographically (polarization
holography) patterned alignment layers. The underlying physics was studied for various periodic structures. These structures are attractive for applications because they offer much lower switching voltages than other material combinations that involve patterned phase separation of liquid-crystal/polymer dispersions. Furthermore, thin polarization gratings have high diffraction efficiency potential. This work was extended to two-dimensional and polymeric structures.

ACKNOWLEDGMENTS

The authors acknowledge the support from the National Science Foundation (NSF ECS 03-22878). One of the authors (J.N.E.) acknowledges the support from the NASA GSRP. Two other authors (A.C.-J.) and (R.A.P.) acknowledge the support from the National Science Foundation (NSF DMR 01-31573).

23G. Skacej, S. Zumer, and G. P. Crawford (unpublished)