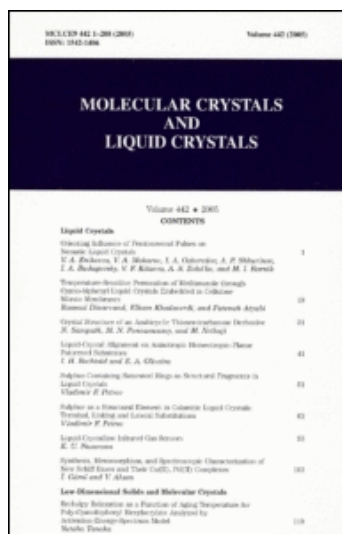


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Fast Electro-Optical Device Based on Chiral Liquid Crystals Encapsulated in Periodic Polymer Channels

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The chiral flexo-electro-optic effect produces a sub-millisecond, temperature independent in-plane rotation of the optical axis and is potentially interesting for the display industry. The main drawback in the exploitation of this effect is that it relies on a texture, the Uniform Lying Helix (ULH), which is intrinsically unstable since neither planar nor homeotropic surface conditions are compatible with it. We present a method, based on the use of periodic polymeric micro-channels, to create highly ordered and stable ULH structures. We show that the periodic structure, which is created holographically, naturally aligns the cholesteric helical superstructure along the micro-channels, without requiring any elaborate ad-hoc procedure, even when the size Λ of the micro-channels is much larger than the pitch P ($\Lambda > 20P$). Electro-optic measurements performed on the test-device show a large contrast ratio between bright and dark states (better than 100:1), short switching time (200 μ s) and large optical rotation ($>30^\circ$).

Keywords Electro-optic; flexoelectricity; liquid crystals application; polymer structure

1. Introduction

Flexoelectricity in a nematic liquid crystal, first predicted by Meyer [1] in 1969, is the coupling between the elastic distortion of the director and the electric polarization. It can be shown using symmetry arguments that only bend and splay distortion produce a non-zero electric polarization \vec{P} , which can be expressed as:

$$\vec{P} = e_1 \hat{n}(\nabla \cdot \hat{n}) - e_3 \hat{n} \times (\nabla \times \hat{n}) \quad (1)$$

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where \hat{n} is the nematic director and the coupling coefficients e_1 and e_3 are referred respectively as splay and bend flexoelectric coefficients.

An interesting consequence of the flexoelectric coupling is the so called chiral flexo-electro-optic effect (FEO) [2], which is observed in a short pitch cholesteric aligned in the uniform lying helix (ULH) texture. ULH refers to a uniformly aligned parallel-plate cell in which the helical axis of a cholesteric lies parallel to the confining surfaces in a unique direction. A short pitch cholesteric is optically uniaxial, with the optical axis parallel to the cholesteric helix such that an aligned ULH slab acts like a retardation plate.

In the FEO an electric field E , applied perpendicular to the helix induces an in-plane tilt of the optical axis of a short-pitch cholesteric aligned in the ULH (see Figure 1). The magnitude of the tilt is proportional to the electric field, and the tilt is inverted if the polarity of the electric field is reversed.

The flexo-electric nature of the tilt becomes apparent if one considers the projection of the nematic director on a plane Γ perpendicular to the optical axis. As shown in Figure 1, in the absence of an electric field the tilt is zero and the director projection on Γ is homogeneous, but for a tilt different from zero, one can easily identify a periodic splay bend distortion of the nematic director on Γ . The flexo-electric polarization arising from such distortion couples with the electric field causing the tilt.

If the tilt ϕ is assumed constant throughout the cell, it can be shown that:

$$\tan \phi = \frac{(e_1 - e_3)p}{2\pi(k_{11} + k_{33})} E \quad (2)$$

where p is the cholesteric pitch, k_{11} and k_{33} are the elastic constants associated respectively to the splay and bend deformation of the nematic director and E is the magnitude of the electric field.

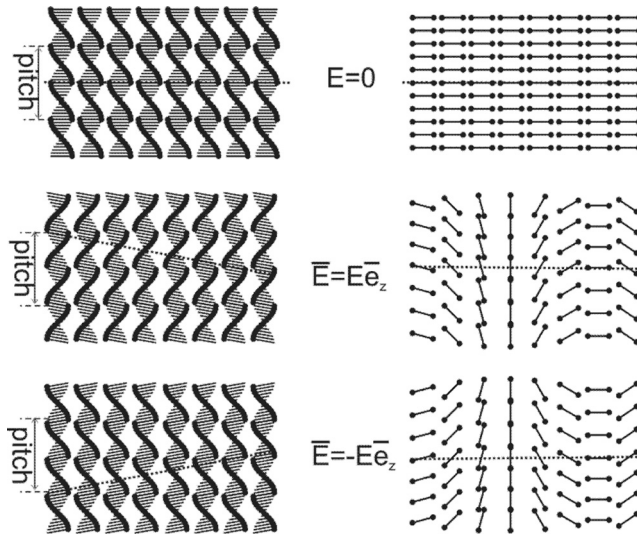


Figure 1. In the presence of an electric field, the projection of the director on a plane perpendicular to the optical axis describes a periodic bend-splay pattern which produces a flexoelectric polarization that couples with the electric field.

The simple behaviour described in equation 2 is in general perturbed by the dielectric coupling which tends to unwind the cholesteric helix, limiting the linear regime of the FEO [3], however, an appropriate design of the liquid crystal (LC) molecules can substantially increase the range of linearity of the FEO [4,5]. In the past 10 years, a considerable improvement has been made in this direction and new LC mixtures, specifically designed for the FEO, have been synthesized, based on the use of bimesogenic molecules [6–8]. These mixtures exhibit a large flexoelectric ratio $(e_1 - e_3)/(k_{11} + k_{33})$, and a relatively small dielectric constant $\Delta\epsilon$ extending the linear range of the flexoelectric effect to angles as large as 30° [9]. Thus full modulation between dark and bright states is possible within the linear regime.

Linear effects in LCs are quite rare and potentially very interesting, since they allow fast changes in the optical state [10]. The case of FEO is particularly interesting for many reasons. First of all, the response times are in the microsecond range for the short pitch materials (it has to be noted that the use of pitch of around 300 nm is also necessary to avoid diffractive effects). In addition, the FEO can show great temperature independence, since both the difference of the flexoelectric coefficients $(e_1 - e_3)$ and the elastic constants $(k_{11} + k_{33})$ are believed to scale quadratically with the nematic order parameter S [3]. By choosing a chiral LC mixtures with temperature compensated pitch, it is possible to obtain an electro-optic characteristic that is temperature independent [3,11].

This makes FEO, potentially, useful for applications like wide viewing angle displays with grey-scale possibility or sub-millisecond light shutters.

2. Uniform Lying Helix

In order to exploit the FEO in the display industry a few problems still need to be addressed, the most severe one is related to the ULH texture required. The ULH is intrinsically unstable since it is incompatible with both planar and homeotropic alignments. As a consequence, in order to accommodate the ULH close to a uniformly aligning surface a set of defect lines or a large periodic distortion need to be introduced close to the surface. For positive dielectric anisotropy materials, one can force the ULH texture into the system by cooling down the sample from the isotropic phase, while applying a moderate electric field, but the texture is not ideal and, when rotated under crossed polarizers, the contrast ratio between dark and

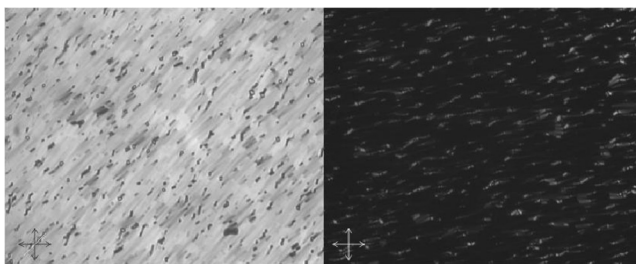


Figure 2. ULH alignment created by cooling a chiral nematic from the isotropic phase, while applying a moderate electric field. The optical axis is oriented (a) 45° and (b) 0° with respect to the polarizer.

bright states is, in general, quite poor (see Fig. 2). Moreover, when the electric field is turned off, the system naturally relaxes back to the lower energy Grandjean texture, where the cholesteric helix is perpendicular to the confining surfaces. So far, only a few methods have been proposed to overcome this problem but a viable solution has still to be found.

In order to stabilize the ULH texture, the use of reactive mesogens has been proposed to create a polymeric structure that locks the cholesteric into the ULH texture [12–14]. The use of an appropriate concentration of reactive mesogens leads to stability of the ULH without having deterioration in the tilt magnitude or the switching speed. Nevertheless, this method relies on the good quality of a pre-existing ULH texture. This is usually achieved by mechanical shearing of the substrates, in order to induce a flow, while applying a moderate electric field through the cell, to uniformly align the helix axis in the plane of the device. This strategy can only be applied to align chiral nematics with positive dielectric anisotropy. Moreover, it is not viable to align devices in this way for industrial applications.

The use of a surface showing periodic (horizontal/vertical) anchoring conditions has also been used to produce ULH alignment [15]. Even though the idea is very interesting and allows the creation of a ULH without complex procedures such as mechanical shearing, the resulting ULH structure does not show good stability during thermal cycling, which is essential for practical applications. Moreover, since an electric field needs to be used to favor the ULH over other less ordered textures (such as focal conics texture), it only can be used for positive dielectric anisotropy materials.

3. ULH Induced by Periodic Microchannels

In this paper we present a method to promote the ULH as the lowest energy texture, using periodic polymer walls to break the in-plane translational symmetry, while imposing homeotropic alignment to the LC molecules, as depicted in Figure 3. In this geometry, the cholesteric helix can only orient along the polymer channels.

The creation of the polymer micro-channel is based on the polymer liquid crystal polymer slides (POLICRYPS) [16] method and involves a physical-chemistry multi-step process, which will be reported elsewhere. This process results in a series of side-by-side microchannels with a very sharp morphology and within each channel

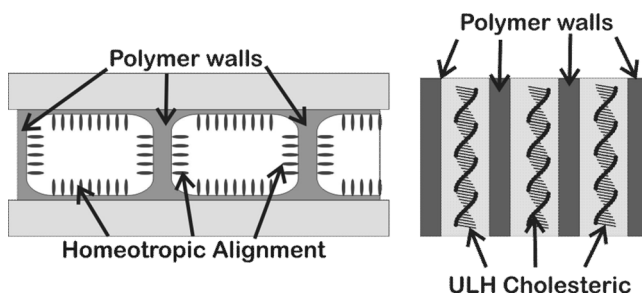


Figure 3. In order to promote a stable ULH alignment we break the in-plane translational symmetry by creating periodic polymer walls while imposing homeotropic alignment to the LC molecules.

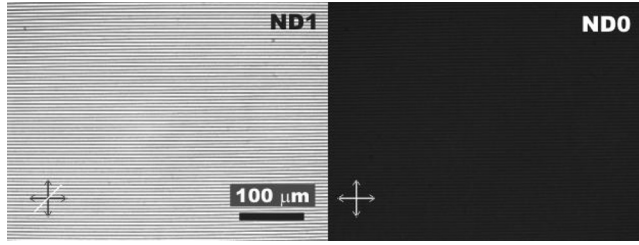


Figure 4. Cholesteric filled periodic polymeric microchannel observed with a polarizing microscope. The optical axis is oriented (a) 45° and (b) 0° with respect to the polarizer.

the cholesteric helix orients along the microchannels. Figure 4 is a crossed polarized micrograph of a short pitch cholesteric liquid crystal encapsulated in periodic polymer channels fabricated using this method. The picture shows a good quality of alignment and the large contrast between dark and bright states.

Once formed the ULH is extremely stable. We verified that the ULH naturally reforms when the sample is cooled down in the cholesteric phase after heating in to the isotropic phase. This shows the ULH is indeed the ground state texture of the system. The ULH was also stable to the application of external electric field. For large values of the electric field we observed an unwinding of the cholesteric helix. Nevertheless when compared to the threshold value E_{th} for unwinding the same cholesteric in a bulk system, the electric field required in our system is larger by 40%. Also, even when the electric field unwinds the helix, the ULH is restored as the voltage is decreased. Finally we verified that the texture is also stable against mechanical stress.

In order to verify the robustness of this method we created several devices varying the ratio Λ/p , between the period Λ of the polymeric structure and the pitch p of the cholesteric helix, from 5 to 30 and all systems resulted in a well ordered ULH texture.

Based on the latter approach, we engineered a device using a chiral nematic LC, which consists of a bimesogenic mixture (Merck) doped with $\sim 2\%$ of R5011 (Merck), a chiral dopant with large helical twist power. The use of dopant with large helical twisting power allowed us to produce a chiral nematic with a short pitch without using large amount of dopant, which could otherwise modify the flexoelectric ratio or the dielectric anisotropy of the mixture. The mixture resulted in a chiral nematic with a pitch $p \approx 300$ nm. This value of the pitch is the trade-off between large rotation of optical axis and fast switching speed. In fact, as predicted by Eq. (2), larger tilt angles ϕ are promoted by using long pitch. On the other hand, faster switching speeds are achieved using short values of the pitch:

$$\tau = \frac{\gamma p^2}{4\pi^2 k} \quad (3)$$

where τ is the switching time, k is an average elastic constant and γ is the effective viscosity associated with the tilt of the director within the helix and is approximately equal to the twist viscosity γ_1 [17]. Due to the small index contrast between polymer and LC, the device is expected to produce diffraction of the incident light. In the past,

a theoretical model [18] based on Kogelnik's idea [19] has been implemented to predict the diffraction efficiency η of the POLICRYPS gratings. The approach yields the expression $\eta = \sin^2 \Phi(T, L, \lambda)$, where the phase Φ is a function of the grating thickness L , the probe wavelength λ , and temperature T . η is therefore a periodic function of the phase Φ . The value of L has been chosen to obtain a very low efficiency grating since for this application, the grating should ideally transmit all the impinging light. Experimentally, we observed diffraction efficiency smaller than a few percent.

4. Electro-Optical Measurements

The test device exhibited a large optical contrast between dark and bright state (see Fig. 4). A quantitative measurement of the contrast was performed with the help of a polarized microscope. A photodiode was used to measure the intensity of the white noncollimated light transmitted through the sample inserted between the polarizers and with its optical axis parallel to one of the polarizers or oriented 45° with respect to them. In order to minimize the effect of nonlinearities of the photodiode, a neutral density filter ND2 was used to attenuate the intensity of the light transmitted in the bright state. The measured contrast was better than 100:1, which to our knowledge is the largest value reported for a ULH texture, using white noncollimated light.

In order to perform electro-optic characterization, the sample was placed in a micro-oven (instec HCS412 W, which allowed temperature control within 0.01°C) and then mounted on the rotating stage of the optical setup in Figure 5. The light of a He-Ne laser ($\lambda = 632.8 \text{ nm}$) passes through the polarizer A, the sample, the analyzer B, whose optical axis is crossed with respect to A, and is finally detected by a photodiode. A function generator (Wavetek 395) and a 10x power amplifier (FLC Electronic) were used to apply an ac voltage across the device. Both the voltage applied to the cell and the signal from the photodiode were monitored on a digital oscilloscope used to both visualize and record the signal.

A measurement of the maximum tilt ϕ of the optical axis as a function of the applied voltage V_{in} was performed. For each temperature T and each value of the applied voltage V_{in} , the sample was rotated so that the angle θ angle between

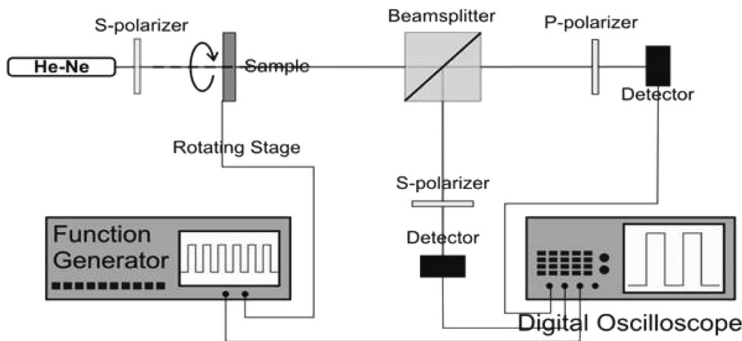


Figure 5. Optical setup used to carry out the electro-optical characterization of the samples. The sample is temperature controlled and can be rotated with respect to the direction of the laser beam.

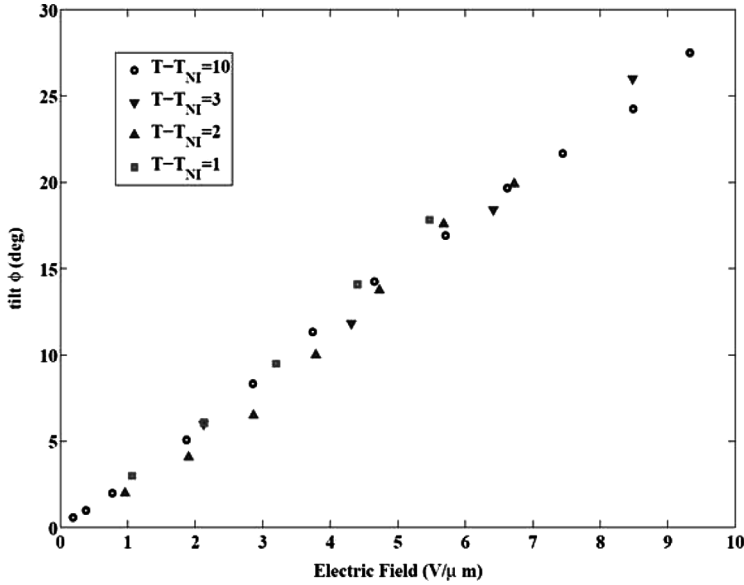


Figure 6. The tilt ϕ of the optical axis as a function of the applied voltage for four shifted temperatures. The tilts show linear increase with the applied field without temperature dependence.

the polarizer and optical axis of the device was set to $\theta = 22.5^\circ$, for this value of θ the mean intensity \bar{I} measured by the photodiode was recorded. Because of the ac electric field applied to the device, the optical axis oscillates around its direction at zero field and the intensity measured by the photo diode is an ac signal oscillating between I_{max} and I_{min} . If θ is rotated, I_{max} and I_{min} change. In order to measure ϕ , θ was first rotated so that I_{max} reaches the value measured for \bar{I} at some value of $\theta = \theta_{max}$ and then in the opposite direction so that the I_{min} reaches the value measured for \bar{I} at some value of $\theta = \theta_{min}$. The tilt ϕ of the optical axis was then evaluated as $2\phi = \theta_{max} - \theta_{min}$.

Figure 6 shows the tilt ϕ as a function of amplitude of the electric field for several shifted temperatures. As expected from equation 2, the tilt increases linearly with the applied field and no temperature dependence is observed. As discussed in paragraph 1, the flexoelectric ratio $(e_1 - e_3)/(k_{11} + k_{33})$ is temperature independent. As a result, the temperature independence of the tilt reflects a temperature independence of the helical pitch.

The values of the tilt are smaller than expected for this material. Figure 7 shows the tilt ϕ as a function of E obtained in a standard planar cell, filled with the same cholesteric mixture as the test device. The ULH alignment in this case is created by cooling down the LC from the isotropic phase while applying a moderate electric field. By comparing the ratio ϕ/E for the two systems, one can notice that ϕ/E is smaller in the system containing the microchannel, by 30%. This is probably due to the constrained geometry created by the microchannels. Even for a regular cell, it is well known that anchoring and confinement effects tend to decrease the flexoelectric tilt [9,20]. A detailed study of the effect of confinement in these systems will be carried out elsewhere.

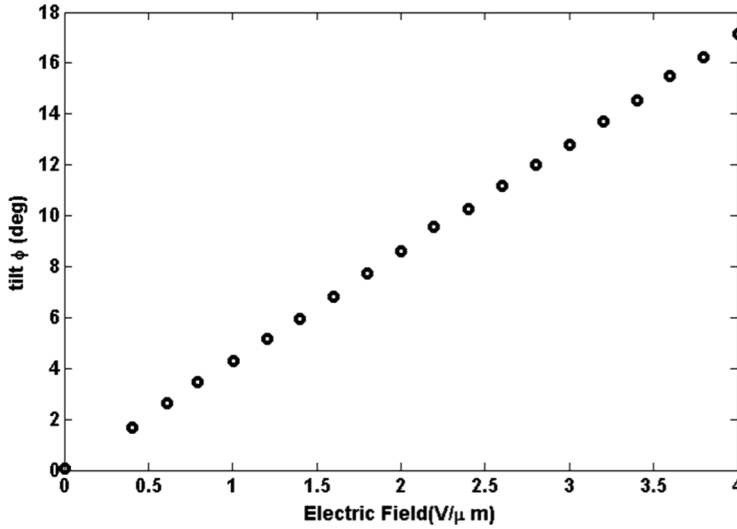


Figure 7. The tilt Φ of the optical axis as a function of the applied voltage for a standard planar device filled with the same chiral nematic mixture used in the test device.

The switching times τ as a function of the applied field for different shifted temperatures are shown in Figure 8. τ was measured as the time required for the transmitted intensity to change from 10% to 90% of the peak to peak amplitude after field polarity reversal of an square wave with $\theta = 22.5^\circ$. The values of τ show a strong dependence on temperature, mainly due to the temperature dependence of γ . From Figure 8 it is clear that τ also depends on the electric field. Even though this is not predicted by the theory [1], it is often observed in experiments [6,14,20].

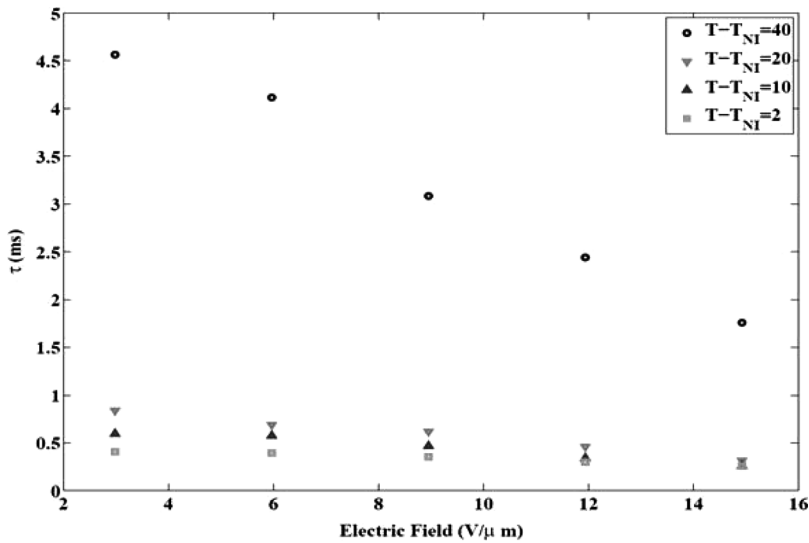


Figure 8. Switching time τ as a function of the applied field for different shifted temperatures.

5. Conclusions

To conclude, we have presented a method based on the use of periodic polymeric microchannels, which allows the creation of highly ordered and stable ULH structures. The ULH is shown to be stable under the application of large electric fields and temperature cycling through phase transitions.

A unique characteristic of this method is that it has the potential to be applicable to any cholesteric material regardless of the dielectric anisotropy since the creation of the ULH structure does not rely on dielectric properties but rather on surface coupling. The electro-optic characterization of a device constructed using this method shows tilts of the optical axis that are large enough to produce full modulation between dark and bright states, switching times on the order of $\sim 100 \mu\text{s}$, and a large contrast ratio between dark and bright states.

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