Higher Order Bragg Reflection Colors in Polymer Stabilized Cholesteric Liquid Crystals

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Abstract:

We have previously reported on dynamic electro-optic (EO) response of polymer stabilized cholesteric liquid crystals prepared using unpolarized UV light (U-PSCLC), such as reflection bandwidth broadening and either red or blue tuning of the reflection peak. Here, we describe recent efforts to use a polarized single argon ion laser beam to create PSCLCs (L-PSCLCs) with higher order reflections. The L-PSCLCs exhibit a primary reflection peak in the near infrared (NIR) regime and a second order reflection band with a narrow bandwidth in the visible regime that results from a deformed in-plane CLC helical structure. The initial positions of the reflection bands are adjusted by the chiral dopant concentrations of the CLC.
mixture, and red, green and blue reflection colors from the second order Bragg reflection are demonstrated. The primary and the second order reflection bands can be shifted to longer wavelengths by application of a direct current (DC) electric field. The reflection efficiency of the higher order reflection notch increases with polymer concentration, which affects the degree of in-plane deformation and fixation of the CLC helix. Modeling is used to further explain the formation of the higher order reflection bands of PSCLCs observed experimentally.

**Introduction**

Cholesteric liquid crystals (CLC) are one-dimensional, photonic materials that exhibit selective Bragg-type reflection due to the self-organized, helicoidal superstructure.[1, 2] The periodic CLC phase has a pitch \( p_0 \) expressed as \( \lambda_0 = \bar{n} \cdot p_0 \) and \( \Delta \lambda = \Delta n \cdot p_0 \), where \( \lambda_0 \) is the center of the wavelength, \( \Delta \lambda \) is bandwidth, \( \bar{n} \) is the average refractive index of the liquid crystal, \( p_0 \) is the cholesteric pitch length, and \( \Delta n \) is the birefringence of the liquid crystal. CLCs can be prepared by mixing chiral dopants with a nematic medium, and the spectral position (pitch length) of the CLC can be adjusted by the chiral dopant concentration. The supramolecular chirality of the CLC is determined by the handedness of the chiral dopant, and circularly polarized (CP) light having the same handedness is reflected 100% and CP with the opposite handedness is transmitted 100% (unpolarized light - 50% transmitted, 50% reflected). For a left handed helix and left handed light, 100% is reflected.

Polymer stabilization of CLCs is a common method used to improve relaxation kinetics and control the phase behavior of CLC with positive dielectric anisotropy (\( \Delta \varepsilon > 0 \)).[3-14] Over the past several years, we extensively studied the dynamic EO response of PSCLCs with negative \( \Delta \varepsilon \), including bandwidth broadening,[6-8, 15] red tuning,[16-18] blue tuning,[19] and switching scattering.[20] The proposed mechanism of dynamic EO responses is ion mediated deformation of the polymer network used to stabilize the CLC. The application of a DC field
induces movement of ions, especially positive ions, trapped on and in the polymer network, causing the polymer network to deform towards the negative electrode. The small molecule LC host consequently moves with the polymer network which results in a pitch variation across the cell.\(^{[6, 8, 18]}\) Recently, we also demonstrated that reversible switching and red-shifting tuning responses in a single positive \(\Delta \varepsilon\) PSCLC sample.\(^{[21]}\) AC field application induces switching from reflective to transparent states by rotating the positive \(\Delta \varepsilon\) LC in the normal direction of the substrate, while the pitch gradient is induced by the deformation of the polymer network by an increase in the DC field and the position of the reflection notch is controlled.

Higher order reflections in CLCs with positive dielectric anisotropy (\(\Delta \varepsilon > 0\)) have been previously observed in planar cells fabricated with interdigitated electrodes. In this case, the pitch of the CLC increases when an electric field is applied perpendicular to the helical axis of the CLC, but the planar texture is maintained.\(^{[22-24]}\) The increase in pitch causes a red shift of the reflection bands. At an electric field higher than the critical electric field, higher order reflection bands were induced in the CLCs due to the helix unwinding of the CLC.\(^{[25, 26]}\) While Blinov et al. observed a second order reflection band using this technique, Rumi et al. observed second and third order reflection bands of CLCs with \(\Delta \varepsilon > 0\) in planar cells with interdigitated electrodes. This report described high reflection efficiency of the second order reflection band (~80% of the main reflection band) whose position could be tuned by the magnitude of the applied electric field.

Broer and colleagues also reported higher order Bragg reflections in CLC polymers containing dichroic initiators photopolymerized by linearly polarized (LP) UV light.\(^{[27-30]}\) The higher absorption of the dichroic photoinitiator in the direction parallel to the direction of the light polarization leads to faster initiation and photopolymerization of the LC monomers in the CLC mixture. This selective, non-uniform photopolymerization causes inhomogeneous director rotation in the CLC helical structure through the cell thickness which leads to higher
order Bragg reflections. The reflection efficiency of the higher order reflection bands is proportional to the degree of deformation of the helical structure in the CLC polymers.

In this manuscript, we report on the formation of higher order reflections of PSCLCs prepared from a CLC mixture without the use of interdigitated electrodes or a dichroic initiator. Instead, a single linearly polarized argon laser beam is used to polymerize the liquid crystal monomers (LCMs) within the CLC mixture. The position of the second order reflection band of the CLC is controlled by the concentration of the chiral dopant, and the efficiency of the higher order Bragg reflection increases with polymer concentration. The position of the second order reflection and the main CLC reflection bands can be manipulated by applying a DC field. Liquid crystal monomers (LCMs) in a CLC mixture in the direction parallel to the laser polarization direction can absorb more UV light than LCMs in other directions, resulting in an inhomogeneously deformed in-plane CLC helix and higher order reflections. L-PSCLCs samples with electrically tunable higher order reflection color and the anisotropic ratio of reflection can be used for optical and display applications.

Results and Discussion

Two PSCLCs were prepared by irradiating with either unpolarized UV light (U-PSCLC) or a linearly polarized argon laser beam (L-PSCLC) and the transmission spectra were collected using right-handed circularly polarized light as the probe beam (Figure 1). The U-PSCLC and L-PSCLC samples were prepared from the same LC mixture containing 1% photoinitiator Omnirad 819 (IGM Resins), 13% liquid crystal monomer (C11M, AlphaMicron, Inc), 3.5% right handed chiral dopant (R1011, Merck) and the positive \( \Delta \varepsilon \) liquid crystal (E7, Merck).

The chemical structures for the component compounds are shown in Figure S1. As can be seen in Figure 1(a, b), both U-PSCLC and L-PSCLC exhibit a reflection peak at 1320 nm, but the L-PSCLC sample displays a second order reflection peak at 660 nm. The spectral position of this reflection notch is half of the position of the main reflection band. While the U-PSCLC
sample is visually transparent because the Bragg reflection is in the NIR, the L-PSCLC sample has exhibits red reflection (see inset in Figure 1(b)). Additional L-PSCLC samples containing 4wt% and 5wt% chiral dopant R1011 were prepared. Figure 1(c, d) depicts the transmission spectra and inset photographs of thin films of these two additional L-PSCLC samples. The spectral positions of the primary and secondary peaks are easily adjusted by the chiral dopant concentration in the CLC mixture, and red, green, and blue reflection colors are observed in L-PSCLC samples containing 3.5 wt%, 4 wt%, and 5 wt% R1011, respectively.

As a control experiment, a PSCLC sample was prepared from a CLC mixture without a photoinitiator exposed to the same argon laser beam under the same exposure conditions.[31] In Figure S2, higher order Bragg reflections are not observed in the PSCLC sample without a photoinitiator, whereas the second order Bragg reflection peak is observed in the PSCLC sample with 1wt% photoinitiator.

**Figure 1.** Transmission spectra of PSCLCs with $\Delta\varepsilon > 0$ irradiated for 10 min by (a) unpolarized 365 nm wavelength UV light with an intensity of 100 mW cm$^{-2}$ and (b-d) a linearly polarized argon laser beam with an intensity of 70 mW cm$^{-2}$. The CLC mixtures contain 1wt% Omnirad 819 (photoinitiator), 13% C11M, various R1011 concentrations and a nematic LC mixture E7: (a, b) 3.5%, (c) 4%, and (d) 5% R1011. The polarization direction of
the laser beam is parallel to the rubbing direction of the cell. Inset is an optical microscope reflection image and the scale bar is 200 nm.

The exposure of the argon ion laser to the CLC mixture leads to in-plane deformation of the CLC helical structure. **Figure 2** shows the degree of deformation of the helical structure when photopolymerized in two different curing conditions, with the rubbing direction of the cell either parallel (∥) or perpendicular (⊥) to the polarization direction of the argon ion laser beam. The polar diagram of these L-PSCLCs measured using a UV-vis setup is summarized in Figure 2(c). The sample polymerized under parallel curing condition (∥) shows strong polarization dependence of the second order reflection band in Figure 2(a), and the maximum reflection efficiency of the second order reflection band is observed at 75-255° coordinate indicating a deformed in-plane helical CLC structure and polarization rotation. However, the L-PSCLC sample polymerized under perpendicular curing condition (⊥) shows a small polarization dependence of the second order reflection band and a small rotation of the helical axis as shown in Figure 2(b). During irradiation, an inhomogeneous director rotation in the helical polymer network structure through the cell thickness (in-plane helical deformation) is induced by the combination of reorientation and photopolymerization of the liquid crystal monomer.\textsuperscript{[27,28]} In Figure 2(c), the ratios of reflection ($R = R_{\text{max}}/R_{\text{min}}$) of 18 µm thick samples polymerized under parallel (∥) and perpendicular (⊥) curing conditions are 3.23 and 1.16, respectively. The anisotropic in-plane distortion of the helical structure is explained in **Scheme 1**. U-PSCLC shows a uniform twist angle ($\Phi$) of the average in-plane molar rotation, while L-PSCLC exhibits a non-uniform rotation of the average in-plane molar rotation due to a deformed in-plane helical structure. U-PSCLC exhibits a uniform twist angle ($\Phi$) of the average in-plane molar rotation, while L-PSCLC shows a non-uniform rotation of the average in-plane molar rotation.

This indicates that the ratio of reflection of the L-PSCLC sample can be controlled by the curing direction of the CLC mixture.\textsuperscript{[27,28]}

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Figure 2. Transmission spectra of the second order reflection band of L-PSCLC with a polymer concentration of 15 wt% irradiated by argon ion laser in (a) a parallel direction and (b) a perpendicular direction to the rubbing direction of the cell. (c) A polar diagram of the transmittance of the second-order reflection peak as a function of the polarized probe light. R is the rubbing direction.

Scheme 1. Schematic illustration of PSCLCs prepared upon exposure of (a, b) unpolarized 365 nm UV light (U-PSCLC) and (c, d) 363.8 nm Argon laser beam (L-PSCLC): (a, c) top-down view and (b, d) side view. Φ is the twist angle of the average molar rotation.
Modeling of higher order Bragg reflections for L-PSCLC was performed and is shown in Figure 3 and Figure S3. The higher order Bragg reflection of L-PSCLC is modeled with an inhomogeneously twisted director through the CLC helix using the various nondimensional products ($\bar{a}I_0$) from 0.05 to 0.5. A good agreement between the experimental data (Figure 1(d)) and the calculated results is observed in Figure 3. Since photopolymerization occurs under a linearly polarized light, the network density is higher in the places where the initial director is parallel to the light polarization. This network does not want the LC director to be twisted and compete with chiral dopant induced twisting power. This induces the deformed in-plane CLC helical structure. In an ideal cholesteric liquid crystal, the twist of the CLC director through the CLC helix is uniform and is given by \[ \frac{d\phi(z)}{dz} = \frac{2\pi}{P_0} \] or \[ \phi(z) = \frac{2\pi z}{P_0} \]. In the inhomogeneously twisted sample, the twist angle dependence on $z$-coordinate is described by a similar formula \[ \frac{d\phi(z)}{dz} = \frac{2\pi}{P(z)} \]. The pitch change induced by light at a given point $z$ is proportional to the absorption of light at this point

\[ \frac{dP(z)}{dz} = -\alpha I_0 \cos^2 \phi(z) \]  

(1a)

where parameter $\alpha$ is the probability of the light absorption and monomers polymerization, $I_0$ is the light intensity magnitude, and the angle $\phi(z)$ is the angle between the light polarization and the director. The angle $\phi(z)$ in its turn satisfies the equation

\[ \frac{d\phi(z)}{dz} = \frac{2\pi}{P_0 + P_1(z)} \]  

(1b)

The approximate solution to the system of equations (1a) and (1b) is obtained by successive approximations. To find the first order approximation for $P(z)$ from the eqn. (1a), we used
zero order approximation for \( \phi(z) \) in the eqn. (1b), \( \frac{d\phi(z)}{dz} \approx \frac{2\pi}{P_0} \), and correspondingly

\[
\phi(z) = \frac{2\pi z}{P_0},
\]

then

\[
\frac{dP_1(z)}{dz} = -\alpha I_0 \cos^2\left( \frac{2\pi z}{P_0} \right)
\]

(2)

It is convenient to use the non-dimensional coordinate \( x = \frac{4z}{P_0} - 1 \),\(^{[27, 28]} \) which varies from -1 to 1, when \( z \) changes from 0 to \( P_0 / 2 \). Then the eqn. (2) becomes

\[
\frac{dP_1(x)}{dx} = \frac{P_0 \alpha I_0}{4} \cos^2\left( \frac{\pi}{2} (x+1) \right) = -2\tilde{\alpha} I_0 P_0 \cos^2\left( \frac{\pi}{2} (x+1) \right)
\]

(3)

Solution to this equation subject to \( P_1(x) = 0 \) when \( I_0 = 0 \) reads

\[
P_1(x) = -\tilde{\alpha} I_0 P_0 \left[ \text{const} + x + \frac{\sin \pi (x+1)}{\pi} \right]
\]

(4)

The first order approximation for the CLC director twisted within one period should be found from the eqn. (1b), which now gives

\[
\phi(x) = \frac{P_0}{4} \int_{-1}^{x} \frac{2\pi}{P(x)} dx \approx \frac{\pi}{2} \left[ x + 1 - \frac{1}{P_0} \int_{-1}^{x} P_1(x) dx \right].
\]

When conditioned by the imposed periodicity \( P_0 \), \( \phi(-1) = 0 \), \( \phi(1) = \pi \), the solution reads

\[
\phi(x) = \frac{\pi}{2} \left[ x + 1 + \tilde{\alpha} I_0 \left( \frac{x^2 - 1}{2} - \frac{\cos \pi (x+1) - 1}{\pi^2} \right) \right]
\]

(5)

Figure S3 shows calculated transmittance and reflectance of the L-PSCLC film with the inhomogeneous director twist given by eqn. (5) when the nondimensional product \( \tilde{\alpha} I_0 = 0.5, \) 0.3, 0.25 and 0.05; the other parameters used in the modeling are \( P_0 = 554nm \) (corresponds to the \( \lambda_{refl} = 900nm \)), LC refractive indices \( n_o = 1.5357, \) \( n_e = 1.7144 \), a film thickness \( L = 20 \mu m \), a bottom substrate refractive index \( n_s = 1.52 \) and air \((n_{top} = 1)\) above the film.
Comparing Figure 1(d) and Figure 3 shows that the proposed mechanism for the in-plane pitch distortion due to inhomogeneous photopolymerization yields qualitatively good agreement between modeling and experimental observations. The slight discrepancy in the position of the second order reflection peak between the experimental and modeled transmittance is probably due to the incomplete normal incidence of the UV laser beam on the CLC sample.

**Figure 3.** Transmission spectra of (i) experimental and (ii) modeled L-PSLC sample formulated with 1 wt% Omnirad 819, 13 wt% C11M, 5 wt% R1011, and 81 wt% E7. The higher order Bragg reflection of L-PSLC is modeled with an inhomogeneously twisted director through the CLC helix with the nondimensional product $\tilde{\alpha}l_0 = 0.4$.

Figure 4 shows the transmission spectra of L-PSLC samples prepared under parallel (∥) and perpendicular (⊥) curing conditions probed with unpolarized, right-handed (RH) and left-handed circularized polarized light (LH-CPL). The reflection efficiency of the main and the second order CLC bands depends on the polarization conditions of the probe beam. Both L-PSLCs exhibit selective reflection ($\leq 50\%$ transmission) when unpolarized light is used as the probe beam. When the LH-CPL is used as a probe light, no main and second-order reflection band are observed, but the higher reflection efficiency of the L-PSLCs is observed in the right-handed circularly polarized probe light. Similar reflection behavior of the main
reflection band of L-PSCLCs prepared two different curing directions is observed in various probe beams, but the reflection behavior of the second-order reflection band is greatly affected by the curing directions. The L-PSCLC sample prepared parallel curing direction has higher reflection efficiency of the second-order reflection band when unpolarized light or RH CPL is used as a probe beam.

Figure 4. Transmission spectra of L-PSCLCs polymerized in (a, b) parallel and (c, d) perpendicular directions between the laser polarization direction and the rubbing direction of the cell. (b, d) Transmission spectra of the second-order reflection bands of L-PSCLCs. The samples are probed with (i) unpolarized, (ii) right-handed circularly polarized, or (iii) left-handed circularly polarized light. The sample was prepared by exposure to a 363.8 nm wavelength argon ion laser beam with an intensity of 100 mW cm$^{-2}$ for 10 min. A cell with 18$\mu$m thickness was filled with a CLC mixture of 1wt% Omnirad 819, 20wt% C3M, 4wt% R1011, and 75wt% E7.

The reflection efficiency of L-PSCLC samples depends on the degree of stabilization of polymer network in the CLC medium. Figure 5 shows the reflection efficiency of the second order reflection band as a function of polymer concentration from 6 wt% to 20 wt%. No higher order reflection peaks are observed in samples with a polymer concentration of less than 5 wt%. The reflection efficiency of the main CLC peak at ~1080 nm is constant regardless of the polymer concentration (Figure 5(a)), but the reflection efficiency of the
second order reflection peak at ~540 nm is strongly dependent on the polymer concentration, as summarized in Figure 5(b and c). Exposure to the laser beam induces the LC in-plane rotation, resulting in the distortion of CLC helix, and which is stabilized by the polymerization process. PSCLC samples with higher polymer concentrations show higher reflection efficiency of the second order reflection band, whereas PSCLCs with a polymer concentration of less than 5wt% does not stabilize the distorted CLC. Samples with polymer concentrations higher than 15wt% show maximum reflection efficiency, indicating that the deformed CLC helix is completely fixed.

Figure 5. (a, b) Transmission spectra of L-PSCLCs with various polymer concentrations (6-20wt% C6M). (c) Summary of the reflection efficiency of the second order reflection peak as a function of polymer concentration. For all CLC mixtures, 4wt% R1011 is used.

To investigate the electrooptic response of the higher order reflection peak, the L-PSCLC samples with 15-20 wt% polymer concentrations are used due to their high reflection efficiency. Samples with a polymer concentration higher than 20 wt% require a very high electric field to control the reflection band or form a polymer-dispersed liquid crystal (PDLC) that exhibits switching behaviour. Figure 6 show the dynamic response of the L-PSCLC sample with a polymer concentration of 15 wt% as the DC field increases. The main and the second-order reflection bands shift to longer wave length by increasing the DC voltage to 150V. The main CLC band shows a notch shift of 300 nm from an initial notch position of 1150 nm, while the second-order reflection band exhibits a ~150 nm shift. The initial green reflection color of the second-order reflection band can be tuned to red with 50V. Another L-PSCLC sample with a higher polymer concentration (20 wt% C11M) shows a similar red-
tuning response (Figure S4). When the applied DC voltage is removed, both reflection bands return to the initial notch position. The spectral position, tuning range, and normalized tuning range of the reflection bands of L-PSCLC with 20 wt% polymer concentration (C11M) are summarized in Figure S4(c, d, e). The primary reflection band displays twice the tuning range of the second order reflection band (Figure S4d), but the tuning ranges of the two peaks overlap nicely when the data is normalized (Figure S4e).

**Figure 6.** (a) Red notch tuning of the main and the second-order reflection bands and (b) second-order reflection bands of PSCLC samples with 15 wt% polymer contents (C11M) by increasing DC voltage to 150V. The inset is a reflective image of the sample at 0V, 10V, 30V, and 50V, respectively.

**Conclusion**

The formation of higher order reflection bands and electro-optic response of L-PSCLCs have been reported. The cholesteric liquid crystal mixture exposed to an argon ion laser beam creates a deformed in-plane helical structure of CLC. The deformed inhomogeneous CLC helical structure is fixed via photopolymerization of LCM and leads to higher order reflection bands. The main and the second order reflection bands of these materials can be shifted to longer wavelengths (red tuning) by applying a DC field. The electrically controllable reflection colors and polarization dependence of the main and higher reflection bands in L-PSCLCs can be used in several optical applications.

**Experimental Section**

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Preparation of higher order reflections in polymer stabilized cholesteric liquid crystals:

Alignment cells were prepared from ITO-coated glass slides (Colorado Concepts). The glass substrates were cleansed in acetone and methanol, and treated with air plasma for 30 s. The substrates were spin-coated with a polyimide solution (PI2551, HD Microsystems) and baked at 200 °C for 1 hr. The alignment layers were rubbed with a cloth, and the cell was constructed to ensure a planar alignment condition. The cell gap was controlled by mixing 10-20 µm thick glass rod spacers into an optical adhesive. Samples were prepared by mixing 1% Omnidrad 819 (IGM Resins), 6-20% liquid crystal monomer (C11M, AlphaMicron, Inc or C6M, Merck), 3.5%-5wt% right handed chiral dopant (R1011, Merck) and positive Δε nematic liquid crystal E7 (TNI = 58 °C, Δε = 13.8, Δn = 0.2253 at λ =589 nm, Merck). The polymer stabilizing network was formed within the samples by photoinitiated polymerization with 70~250 mW cm⁻² of unpolarized 365 nm light (Exfo) or a single 363.8 nm Argon ion laser beam (SABRE Innova, Coherent) for 10 min. All materials were used as received without any purification.

Experimental Setup and Measurements: Transmission spectra were collected with a fiber optic spectrometer, and unpolarized, linearly polarized, left-handed or right handed circularly polarized light was used as a probe beam. Transmission spectra were collected before, during, and after application of electric fields with the scanning rate of 1 V s⁻¹ or directly applied to the target voltage.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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An electrically tunable second order reflection color is demonstrated in the positive $\Delta \varepsilon$ PSCLCs. The second order reflection peak shifts from 570 nm to 740 nm as the DC voltage increases to 150V and a color change from green to red is observed.

Keyword: higher order Bragg reflections, cholesteric liquid crystals, polymer stabilizations, color control

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