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Exploiting soft organic optical resonant structure towards large range electro-optic tunable devices

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Abstract

Very fine molecular arrangement of cholesteric liquid crystal (CLC), can be compressed by applying an external electric field in the liquid crystal cell geometry. This enables very fine tuning of the wavelength of the reflected light. The compression of the periodic dielectric molecular arrangement continues till the chiral energy density meets its repulsive-type excluded volume interaction and hence the corresponding resonant wavelength jumps back to the original spectral position. Experimentally, the tuning range is observed to be highly affected by the concentration of the chiral dopant in the mixture. Analytically, it is shown that, use of chiral molecules with higher helical twisting power (HTP) can reduce the concentration of the chiral dopant for a desired pitch length in visible range and hence, the spectral tuning can be exploited over the entire visible range.

Keywords

Tunable filter; Electro-optic device; Cholesteric liquid crystal; Helical twisting power; Bragg's reflection

1. Introduction

Recent market trend in organic integrated devices such as organic thin film transistor, organic sensor, solar cell, OLED, organic photodetector (OPD), etc., is advancing in a rapid pace for enabling the portability, flexibility, ruggedness, and cost effective production purposes. Cholesteric liquid crystal (CLC) based 1-D organic photonic crystal is gaining significant research interest in this background. The salient features of CLC as a photonic crystal are: self-molecular assembling, high packing density, fluidity, compatibility with flexible substrate, easy spectral positioning of photonic bandgap, and controllability over bandwidth. However, to improve the potential applicability of CLC further, real time

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wavelength tunability has been paid a lot of interest in the last few years. There are few viable means of tuning with various external stimuli like mechanical stress [1], UV-light [2], temperature [3], electric field [4] etc. Electric field controlled tuning is more demanding because it is compatible with integrated electro-optic devices. Recently, few electric field induced real time tuning techniques for CLC have been reported: (1) by creating polymer network, which increases the structural restoring force against the external applied electric field [5], (2) untwisting of helix by in-plane switching, where the direction of the applied field is perpendicular to the helix axis [6], (3) inducing the surface dynamic of ferroelectric liquid crystal (FLC) onto a CLC layer [7], (4) using the advantage of low elastic bend constant, CLC shows a large range of bending deformation induced by electric field and hence wide tunable capability [8].

In this communication, an electrostatic actuation based tunability in a classical CLC system is endorsed. Analytically, it has also been shown that the proposed organic tunable optical resonant structure of CLC could have a very large tuning range over the visible spectrum, if we use less concentration of chiral dopant having very high helical twisting power (HTP).

2. Electro-optic properties of CLC molecular system

CLC is basically a mixture composite, consisting of two basic components: nematic liquid crystal (NLC) and chiral dopant, that is why it is also known as chiral nematic. In this phase, chiral molecule induces its chirality onto the NLC host. As a result, nematic directors rotate as a helix around the chiral/helix axis (*Z*-axis) with a pitch length *P* as shown in Fig. 1. It is noted that the pitch length *P* depends on the concentration of chiral dopant *c* as per the following relationship:

$$P = \frac{1}{\text{HTP} \cdot C} \quad (1)$$

where, HTP is the helical twisting power of the chiral dopant, having the unit of azimuthal rotation in $^{\circ}/\mu\text{m}$ along the helix axis-*Z*. The unique feature of CLC is that, by varying the concentration *c* of the chiral dopant of certain HTP, it is very much possible to make the structural pitch length *P* in the order of optical wavelength λ_0 , related as following:

$$\lambda_0 = \langle n \rangle \cdot P \quad (2)$$

where $\langle n \rangle$ is the average refractive index of liquid crystal. Because of this helical periodic structure of CLC, the incoming optical beam (Fig. 1) senses the helical periodicity parallel to the helix axis inside the CLC medium. As a result, one dimensional (1-D) photonic band gap (PBG) is formed, which is static over the time span in the absence of an external electric field. But, application of the external electric field leads to two possibilities, depending on the dielectric anisotropy ϵ (either +ve or -ve) of the host NLC material. Because in the case of $\epsilon = +ve$, i.e., $\epsilon_{\parallel} > \epsilon_{\perp}$, the resultant dielectric coupling in the presence of the

external electric field leads to reorient the long axis of NLC molecules along the direction of the external electric field i.e., perpendicular to the ITO-glass surface (Fig. 1) and, results in the destruction of helical structure or, PBG. In contrast, in the case of $\epsilon = -ve$, i.e., $\epsilon \parallel < \epsilon_{\perp}$, the PBG remains intact even in the presence of the external electric field because, the resultant dielectric coupling is in the favorable direction of helical axis-Z. Here is the interesting phenomenon we have observed experimentally with this $\epsilon = -ve$ NLC host. The PBG starts spectral tuning towards the blue side to a certain spectral range when we apply an external ac field above the threshold value E_{th} . It is worthy to mention here that, after the PBG reaches its extreme spectral position, the intensity of the PBG vanishes and reappears to the original spectral position even though the external field is ON. The frequency of the ac field is kept constant at some moderate value (e.g., around 1 K Hz) during the experiment, because we observed the frequency has no impact on the tunability of the PBG but, only the variation of the ac field strength. In the next analytical section, a smart way has been adopted based on the experimentally measurable quantities such as the reflected optical intensity of the PBG as a function of wavelength, i.e., $I(\lambda)$, the external electric field E_{rms} and, the structural restoring energy/stiffness $S(q)$ as a function of structural periodicity $q = 2\pi/P$. This structural stiffness is further considered to be proportional to the product of HTP and concentration c of the chiral dopant, i.e.,

$$S(q) \propto \text{HTP} \cdot c. \quad (3)$$

3. Analytical treatment

When the static PBG of the CLC medium is subject to $E_{rms} > E_{th}$, the spectral tuning of the PBG is observed experimentally. This experimental phenomenon can be modeled based on the input-output behavior. The externally applied electric field E_{rms} serves as the input field, which interacts with the overall stiffness $S(q)$ of the CLC medium and, as a result we observe the output performance i.e., the spectral tuning, denoted as $I(\tilde{\lambda})$. It is noted that the overall stiffness $S(q)$ contains indeed all possible elastic energies corresponding to the various types of defects used in general Landau's free energy expansion for any LC system [5]. As per the linear elastic coefficient is concerned, the Hooke's law [1] is well valid to formulate the stress-strain relationship for our CLC system which is subject to the electrostatic actuation to compress its periodic structure linearly, i.e.,

$$I(\tilde{\lambda})\tilde{q} \cdot S(q) \propto E_{rms} \quad (4)$$

where $\tilde{q}$ symbolizes the variation in the pitch length (compressed) due to the electrostatic force. It could be a misleading term to use "electrostatic force" as we applied ac field in our experiment, but as far as the LC system is concerned, its response time of the orientational polarizability is very slow as compared to the applied frequency (few KHz), resulting in effective static field of magnitude E_{rms} to the LC.

In general, when $E_{rms} > E_{th}$ the reflected intensity $I(\lambda)$ from a static periodic structure of periodicity q confined within a finite thickness d can be written by using standard diffraction formula over a finite space integral, which reads as [9]:

$$I(\lambda) = F(q) \cdot F^*(q)$$

with,

$$F(q) = \int a(z) \exp(iqz) dz \quad (5)$$

where $F(q)$ is the diffraction amplitude and $a(z)$ is the complex amplitude of the optical wave inside the periodic medium. For the homogeneous distribution of chiral dopant in NLC host, we can imagine the optical wave can exist as a continuous 1-D sinusoidal function i.e., $a(z) = a_0 \cos(2\pi z/P)$, inside the periodic medium of CLC with a pitch length P (Fig. 2). However, for a finite thickness d of the CLC cell, the diffraction amplitude is integrated over the finite space:

$$F(q) = \int_{-d/2}^{d/2} a_0 \cos(2\pi z/P) \cdot \exp(iqz) dz. \quad (6)$$

Using the standard integral formula over the finite space, the intensity becomes a very well-known sinc function [9]:

$$I(\lambda) = F(q) \cdot F^*(q) = d^2 a_0^2 \cdot \text{sinc}^2 \left[\frac{d(q - q_0)}{2} \right]. \quad (7)$$

In order to have the control over the input-output behavior of the system, we impose the dynamic condition from Eq. (4) onto the static output reflected intensity in Eq. (7), which becomes:

$$I(\tilde{\lambda}) = d^2 a_0^2 \cdot \text{sinc}^2 \left[\frac{d(\tilde{q} - q_0)}{2} \right] = d^2 a_0^2 \cdot \text{sinc}^2 \left[\frac{d \left(K \cdot \frac{E_{rms}}{HTP \cdot C} - q_0 \right)}{2} \right] \quad (8)$$

Using $\tilde{q} \propto \frac{E_{rms}}{S(q)}$ and, $S(q) \propto HTP \cdot C$ from Eq. (4) and, Eq. (3) respectively, where K is the proportionality constant. Eq. (8) is a complete equation, carrying all the parameters to control the input-output behavior of the CLC system.

First of all, it is clear from Eq. (8) that, the spectral tuning of the reflected intensity completely depends on E_{rms} . If there is no applied field, i.e., $E_{rms} = 0$, we can immediately find the variable part in the argument of *sinc function* vanishes and, we have some reflected intensity $I(\lambda_0)$ which is centered at some static spectral position q_0 or, equivalent to λ_0 . With $E_{rms} > E_{thr}$, we can expect some spectral tuning of the reflected intensity $I(\lambda)$ from the CLC medium. The limiting range of the spectral tuning can be found by imposing the boundary condition when *sinc function* becomes zero, i.e.,

$$\text{Sinc}(BZ) = \begin{cases} 0, & z = \frac{\pi}{B} \\ 1, & z = 0 \end{cases}$$

Putting this condition in Eq. (8), we can find the maximum spectral tunability in reflected intensity $I(\tilde{\lambda})$ occurred corresponding to a new value of structural periodicity, say at q_0 :

$$q_0 = K \cdot \frac{E_{rms}}{\text{HTP} \cdot C} - \frac{2\pi}{d} \quad (9)$$

and the tuning range $\Delta q = K \cdot \frac{E_{rms}}{\text{HTP} \cdot C}$, by equating the above Eq. (9) with the general form, $q_0 = q - q_0$. Eq. (9) is very important because, it tells us the way to improve the tuning range of the device. We can see the tuning range Δq depends on a few intrinsic parameters of the material, like HTP and the concentration c . It is clear from Eq. (9) that for a chiral dopant of fixed value of HTP, by decreasing the concentration c of the chiral dopant, we can achieve larger tuning range Δq . Basically, by decreasing the concentration of the chiral dopant, the tuning range Δq of the system can be increased because it is not limited by the excluded volume interaction of the chiral molecules as shown schematically in Fig. 2.

4. Experiment

In the experiment, negative dielectric anisotropy liquid crystal MLC-2081 (*Merck*, $n=0.25$, $\epsilon=-5.6$) and the right handed chiral dopant R-811 (*Merck*, $\text{HTP}=22\mu\text{m}^{-1}$) is used. In order to transmit the maximum mechanical stress onto the CLC film, special care has to be attributed on making the uniform thickness of the CLC film between two.

ITO-glass substrates. Macroscopic uniformity in CLC film depends on the uniformity in the cell thickness and microscopic homogeneity depends on the mixture homogeneity and the alignment layer over the ITO-glass substrate. Very uniform $4\mu\text{m}$ -thick is fabricated with two ITO-glass substrate. Each of the substrates has a surface area $20\text{ mm} \times 20\text{ mm}$ and a homogeneous planar alignment layer of polyimide (Fig. 1). The cell is filled at isotropic temperature (65°C) with the chiral-nematic mixture, containing MLC-2081: 82% and, chiral dopant R-811: 18 wt%. After that the cell is cooled down naturally to room temperature.

5. Results

The cell is subject to an ac electric field (1KHz, 15 V/ μ m), parallel to the helical axis–Z, and immediately the wavelength tuning starts, which is observed with a polarizing microscope in transmission mode (Fig. 3) as well as with a spectrometer (Ocean Optics-2000) in reflection mode (Fig. 4a) at the normal direction to the cell substrate. It is observed in the spectrometer that the reflected intensity from the CLC mixture with a central wavelength $\lambda_c = 720$ nm starts moving continuously towards the blue side, and at the farthest extreme with $\lambda_c = 570$ nm, the reflected intensity becomes zero and reappears further at the original position 720 nm, though the electric field remains ON with same value. The effective tuning range $\lambda = (720 - 570)$ nm = 150 nm, shown in Fig. 4a, is considered from the original spectral position to the position just before where the reflected spectra becomes zero (this zero values happens at the extreme end towards the blue side of the spectral tuning, not shown in Fig. 4a). It is observed that the threshold field E_{th} has a strong dependence on the rigidity of the ITO-glass substrate as per the mechanical stress is concerned. With reduction in thickness of the ITO-glass substrate, the rigidity of the substrate is reduced and electrostatic actuation commences even at lower threshold field but, the total tuning range almost remains unaltered (Fig. 4b). Tuning range was observed to be affected only by chiral concentration as shown in Fig. 5a, or equivalently with the central wavelength of various chiral-nematic mixtures (Fig. 5b). It is worthy to mention here that, the tuning range in the case of a very uniform cell thickness but without any alignment layer is almost negligible (5–10 nm only). Because the surface roughness induces microscopic inhomogeneity, which reduces the microscopic ordering of the CLC periodic structure, as a result there is no net electrostatic force developed along the depth (Z-axis) of the cell.

6. Discussions

Analytically two aspects of the experimental results are emphasized, firstly to show how the output spectral tuning performance of the reflected intensity $I(\tilde{\lambda})$ from the periodic CLC medium depends on the real time applied external electric field E_{rms} as in Eq. (8), and, Secondly, to show how the tuning range is limited to a certain maximum spectral position q_0 as defined in Eq. (9). The reason of the limited spectral range is described based on the intrinsic parameters including the chiral dopant concentration c and HTP as in Eq. (9). According to Eq. (9), increasing the chiral concentration reduces the tuning range for a fixed value of HTP of chiral dopant. To realize this situation, a schematic diagram is drawn in Fig. 2, which shows that when the chiral concentration is increased, the electrostatic actuation based compression range of the periodic structure of CLC is limited because of the fact that, the repulsive force is dominant due to the excluded volume interaction energy from higher number of chiral molecules present in the mixture. Experimentally, the chiral concentration dependence of the tuning range is shown in Fig. 5a, where the nonlinearity in the graph might be very logical as it is of *sinc function*.

7. Conclusion

Here we have presented a very demanding aspect of the soft organic CLC material, i.e., real time control of the spectral tunability based on its electrostatic actuation. The entire

phenomenon is explained analytically with a very simple and effective way and based on the analytical finding it is proposed that the present tuning range can be enlarged further by using high HTP value of the chiral dopant. In that case, even by using less concentration of chiral dopant it would be possible to form pitch length corresponding to visible wavelength range. This helps to compress the periodic structure to a larger extent without the system being affected strongly by the excluded volume interaction energy of chiral molecules itself.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Fig. 1.

Planar arrangement of CLC molecules in between two ITO-glass substrates, which serve as parallel plate capacitor and provide the necessary electrostatic force to actuate the CLC system accordingly. It is noted that, we have used LC host of negative dielectric anisotropy i.e., $\epsilon_{\perp} > \epsilon_{\parallel}$ purposely, such that the dielectric coupling does not lead to any change in the periodic structure of LC molecules even in the presence of an external ac electric field.

Fig. 2.

Schematically illustrates two cases of different chiral dopant concentrations, *top figure*: $c_2(z) > c_1(z)$; *bottom figure*. It is shown that the spanning of excluded volume interaction energy corresponds to the chiral concentration $c_1(z)$ is larger than that of the concentration $c_2(z)$. As a result the available space for structural tuning is large in the case of the less chiral concentration i.e., $c_1(z)$. But for the higher chiral concentration the tuning range is limited due to strong repulsive force from the excluded volume interaction.

Fig. 3. Micrographs from a polarizing optical microscope in transmission mode of the CLC mixture inside an electro-optic cell under external electric field, produces continuous blue shift from central wavelength $\lambda_{c1}=720 \text{ nm}$ to $\lambda_{c2}=570 \text{ nm}$.

Fig. 4.

(a) Experimental results showing the tuning range $\lambda = 150$ nm (in 30 s) of the reflected intensity for the CLC mixture of central $\lambda_c = 720$ nm inside the 4 μm cell (b) Threshold field dependency tuning and the three different thresholds corresponding to three different substrate thicknesses; blue: 1.5 mm, red: 500 μm , and green: 100 μm , in all the cases $\lambda = 150$ nm (in 30 sec).

Fig. 5. Tuning range ($\Delta\lambda$ in nm) dependency on (a) chiral dopant concentration (wt%) and (b) central wavelengths (λ_c).