Electrothermal Color Tuning of Cholesteric Liquid Crystals Using Interdigitated Electrode Patterns

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A transparent color-tunable device is presented based on an electrothermal response by using interdigitated electrode patterns. The response is generated by applying an in-plane AC electric field that heats up a thermosensitive cholesteric liquid crystal mixture. The induced temperature elevations cause band gap shifting (Δλ > 350 nm) up until a colorless state is reached, corresponding to the isotropic phase. Color shifting can be tuned manually by varying the electric field or autonomously by the surrounding temperature. Broadband dielectric spectroscopy reveals that the electrothermal response originates from resistive heating of the transparent electrode pattern in conjunction with the cell capacitance and is therefore largely dependent on the electrode configuration. Hence, the electrothermal response can be easily modified by changing the electrode pattern, frequency and/or voltage, dependent on the user’s requirements. Therefore, the ability of this technique to manipulate the autonomous thermal response by an electric field, using only one conductive substrate, shows promise in the field of optoelectronics, sensors, and smart windows.

1. Introduction

Inspired by stimuli-responsive color changes found in nature, researchers have developed dynamic photonic materials that can change their optical properties (ir) reversibly upon environmental changes. Most of these photonic materials operate autonomously, triggered by an external stimulus (e.g., temperature, humidity, light etc.) thereby providing feedback for the user of the current conditions. Despite the ability to operate without the need for human interference, in specific situations, a manual response is required, overcoming the initial autonomous response. For that matter, the introduction of a second stimulus allows for a more specific control dependent on the user’s needs.

Cholesteric liquid crystals (CLCs) are an example of dynamic photonic materials that reflect a certain wavelength of light due to their periodic helical configuration, defined by the pitch. External stimuli can alter the pitch length, resulting in a shift of the initial reflection band toward higher or lower wavelengths. Among these stimuli, electricity is favorable for technological applications since the response can be readily tuned by modifying the applied electric field or the electrode design. Liquid crystals (LCs) exhibit dielectric anisotropy due to their asymmetry that allows them to be reoriented under influence of an electrical stimulus dependent on its direction. In most cases, electrically induced CLC band gap shifting has been achieved in a photonic cell by applying a direct current (DC) electric field between two conductive glass substrates parallel to the helical axis. As such, the electric field caused a contraction or elongation of the pitch, resulting in band gap shifting and/or broadening. A drawback of electrically induced band gap tuning via this method is the usage of an undesired DC electric field, accompanied by transmission losses upon enhancing the amplitude of the electric field due to increased scattering inside the cell. Therefore, it would be beneficial if an alternating current (AC) electric field could be used to enact band gap shifting without any loss of optical quality. Recently, color tuning was demonstrated by reorienting heliconical CLCs upon applying an AC electric field along the cell gap. However, the need of two conductive substrates limits the device applications and complicates the fabrication process. Reducing the electric circuit to a single conductive substrate could not only simplify optoelectronic devices but also render more flexibility over cell-based systems when using flexible conductive substrates.
In-plane switching of CLCs, having positive dielectric anisotropy, can be realized by applying an in-plane AC electric field perpendicular to the helical axis. A single substrate with top an interdigitated electrode (IDE) pattern can be used for this purpose. The applied lateral electric field causes unwinding of the helices, resulting in a shift of the initial reflection band at low frequency (typically 1 kHz). However, limited band gap shifts of only a few nanometers were obtained in all cases at high voltage amplitudes with substantial transmission losses due to the inevitable formation of focal conic structures above the electrode pattern attributed to the non-homogeneous electric field distribution. Hitherto, the challenge to achieve precise color tuning while preserving high optical quality by using CLCs in combination with an in-plane electric field remains.

Besides the above-mentioned electrically responsive LC systems, an electric field can also act as an indirect stimulus, generating a second stimulus that distorts the pitch length. Some of the latest progress on color-tunable devices has been demonstrated by using an electrochemical or electromechanical or electrothermal response in combination with (transparent) conductive substrates.

Recently, Lee et al. demonstrated electrothermal color tuning in a fully transparent cell without the need of additional heat dissipators by applying a high-frequency AC electric field between two conductive substrates. A blue shift over the entire visible spectrum was demonstrated at high frequencies due to heat dissipation generated inside a CLC cell. The induced electrothermal response was attributed to the pseudo-dielectric relaxation of the indium tin oxide (ITO) electrodes under these high frequencies. Inspired by this work, we report now the electrothermal band gap shifting of CLCs by using an in-plane AC electric field. We achieved color tuning over the entire visible spectrum with high optical quality by using a thermosensitive CLC mixture in combination with an IDE pattern. The extent of band gap shifting could be easily tuned by varying the applied frequency and/or voltage amplitude. Furthermore, the device could also operate autonomously in the absence of an electric field due to changes of the surrounding temperature. The presented dual-responsive device can operate with or without electrical stimulation, emphasizing that the user can distort the response at any time dependent on his/her needs. Therefore, the described method shows promise in the field of optoelectronics, actuators, and smart windows.

2. Results and Discussion

A thermosensitive CLC mixture was chosen as responsive system having a smectic to cholesteric phase transition around room temperature before becoming isotropic above 45 °C (Figure S1, Supporting Information). The smectic to cholesteric transition point of the CLC mixture was intentionally chosen to be around ambient temperature to promote a large blue shift upon small temperature elevations. Subsequently, the electro-optical device (Figure 1), equipped with an interdigitated ITO electrode pattern on top of the bottom glass substrate, was filled with this mixture. In the absence of an electric field, the device had a faint red color at room temperature indicating that the initial reflection band was located at the end of the visible spectrum. A color shift was observed as soon as the IDE pattern provided a high-frequency in-plane AC electric field to the system (Video S1, Supporting Information). We gave the device a voltage sweep from 0–70 V while the frequency was kept constant (Figure 2), rendering color tuning from red to green and blue before finally becoming transparent. By holding the electric field constant, no color shift was measured after 5 min, excluding possible temperature fluctuations. Transmission measurements revealed a blue shift over the entire visible spectrum (Figure 3a) upon increasing the voltage amplitude with a maximal achievable band gap shift Δλ > 350 nm. Even the isotropic state could be reached at high voltage amplitudes (U = 70 V), which is illustrated by the loss of the reflection band (Figure S2, Supporting Information). Remarkably the intensity of the reflection band is close to the theoretical 50% and remains constant upon changing the voltage. Furthermore, the scattering is low during the applied voltage sweep, exhibiting high optical quality at any moment. Color reversibility

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**Figure 1.** a) Schematic depiction (side view) of the electrothermal responsive CLC (red inner layer) cell with cell gap d = 6 µm, width w = 13 µm, and spacing s = 10 µm. An impression of the in-plane AC electric field was pictured by the dashed lines. b) Top view of the bottom glass substrate and IDE pattern. The analyzed responsive region is indicated by the dashed square. c) Photo of the actual device with the IDE region highlighted by the dashed square.
was demonstrated by switching the electric field off while being in the cholesteric state, resulting in the reappearance of the initial state (Video S2, Supporting Information).

The displayed color change was further investigated by measuring the shift of the initial reflection band as a function of frequency at a constant voltage \((U = 15 \text{ V})\). An analogous response could be observed upon increasing the frequency (Figure 3b). However, a maximum blue shift was attained at \(f = 10 \text{ MHz}\). At higher frequencies, a red shift was observed, which will be discussed in more detail later (Figure S3, Supporting Information). The optical quality of the device is less in this case due to an increase in scattering. This could imply that a stronger electric field (Figure 3a) helps to preserve the cholesteric state. The ability to shift the reflection band by altering the applied AC field allows for manual control of the device dependent on the user’s needs (Figure S4, Supporting Information).

As mentioned before, S811 containing CLC mixtures undergo a temperature-dependent band gap shift which was verified by heating the developed cell with an external temperature control stage (Figure S5, Supporting Information). A similar blue shift is observed as seen for the electrically induced color changes, proving that the systems could also operate autonomously in the absence of an electrical stimulus. Consequently, the reported color changes in the cell, caused by the applied in-plane electric field, should originate from a delivered electrothermal response. Thus, the applied in-plane electric field acts as an indirect stimulus, generating heat inside the device. To verify that, the cell temperature was measured in the presence of a high-frequency in-plane AC electric field. A significant temperature increase was observed upon increasing the applied frequency and voltage, showing the existence of an electrothermal response (Figure 4).

When the temperature increase \(\Delta T\) is plotted against the applied voltage (Figure 4a), a quadratic relationship is observed. A deviation from this response occurred at high voltage amplitudes \((U > 50 \text{ V})\) that might be caused by LC realignment effects under influence of the strong electric field due to two-frequency addressing, thereby suppressing the electrothermal effect.\(^{[37,38]}\) Figure 4b shows that \(\Delta T\) can be divided into three regions upon enhancing the frequency: a linear response \((f = 0–3 \text{ MHz})\), a flattening \((f = 3–10 \text{ MHz})\) and a decrease \((f = 10–20 \text{ MHz})\) of \(\Delta T\) with respect to the applied frequency. At 10 MHz, the highest temperature was recorded, corresponding to the maximum blue shift (Figure 3b) that was attained at the same frequency. As previously mentioned, a red shift was observed at higher frequencies (Figure S3, Supporting Information) corresponding to the measured decrease of the temperature. The flattening and decrease of \(\Delta T\) can be explained in terms of ohmic losses above \(f > 3 \text{ MHz}\). The oppositely charged electrodes separated by the CLC mixture can be represented as a capacitor, which is known to experience ohmic losses at high frequencies due to a decreasing capacitive reactance \((X_C)\) and an increasing inductive reactance \((X_L)\).\(^{[39,40]}\)

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When reaching a specific frequency, characterized by \(X_L = X_C\), the circuit shows resonance behavior, which manifests as a maximum in the electrothermal response \((\Delta T_{\text{max}})\).

At even higher frequencies \((X_L > X_C)\), the inductive component of the CLC circuit dominates the impedance resulting in a loss in dissipated power and hence a temperature drop. Correlating \(\Delta T\) to \(\Delta \lambda\) and the temperature-dependent response (Figure S5, Supporting Information) affirmed that the color tuning is induced by an electrothermal response. Additionally, the profiles of the associated electrothermally induced wavelength shifts, shown in Figure 4, are similar to those of previous reports supporting the fact that the observed color shifts could be related to generated heat.\(^{[35,41]}\)

Once having established the scaling behavior of the electrothermal response with respect to the voltage amplitude and...
frequency of the driving AC field, it is evident that \( \Delta T \) should also depend on the design of the CLC cell, particularly the electrode pattern will influence the electrothermal response. Although not being studied here, one could argue that different IDE patterns could be used to fine-tune the response dependent on the final requirements.

Lastly, to reveal the origin of the electrothermal response we have performed a reference experiment on studying the dielectric response of an empty and filled CLC cell to an in-plane AC electric field using broadband dielectric spectroscopy (BDS).\(^{[42,43]} \) Here, the dielectric losses of a CLC device were measured over a broad frequency range to investigate the influence of the cell design and the presence of a dielectric liquid (the CLC mixture) on the frequency-dependent capacitance, and dielectric loss of the CLC cell that will alter the response.\(^{[44]} \) Based on the dielectric spectrum of the empty CLC cell, we have deduced an equivalent electrical circuit (lumped circuit) (Figure 5) that accounts for the non-zero contact resistance \( R \) of the ITO structure, the true capacity of the empty IDE structure \( C_\varepsilon \), an additional, parallel capacitance \( \Delta C_{\text{LC}} \) caused by filling the device with the CLC mixture, and a general stray capacitance \( C_{\text{stray}} \), which appeared to be small and thus negligible. From the measured real part of the dielectric spectrum \( C_\varepsilon '(f) \) of the empty cell (Figure S6, Supporting Information), we have derived the low-frequency (“static”) capacitance \( C_\varepsilon \) and the time constant \( (\tau = 150 \text{ ns}) \) using a Debye model (RC series model), which yielded the electrode resistance \( (R = 104 \Omega) \) via the relation \( \tau = R C_\varepsilon \) (details can be found in the Supporting Information).

In a second step, we have separated the true capacitive response of the CLC cell by using the same Debye model, which yielded the corrected complex capacitance spectra \( C_\varepsilon '^*(f) = C_\varepsilon '(f) - i C_\varepsilon ''(f) \) consisting of the real part and imaginary part. These “corrected” spectra are displayed in Figure 6a (loss spectrum \( C_\varepsilon ''(f) \)) and Figure 6b (real part capacitance \( C_\varepsilon '(f) \)) and allow a comparison between the empty and the filled CLC cell response. As expected, filling the CLC structure increases the capacitance by merely 80% because of the dielectric permittivity of the LC being larger than unity. However, and this becomes more clear in the loss spectrum (Figure 6a), the liquid crystal shows a pronounced intrinsic relaxation process with a loss peak maximum around 500 kHz that can be ascribed to slow reorientation dynamics of the mesogenic units (flip process around the short molecular axis).\(^{[45]} \) At room temperature, this additional time scale \( (\tau = 370 \text{ ns}) \) is readily separated from the RC time constant of the empty CLC cell, however, the actual frequency dependence of the permittivity of the LC needs to be taken into account for modelling the electrothermal response of filled CLC cells as discussed below.

Revisiting Figure 6b, we notice that the response in \( C_\varepsilon '(f) \) for the empty CLC cell, even after elimination of contact resistor, is not completely frequency independent as predicted by the equivalent circuit. Toward very low frequencies, the capacitance slightly increases, indicating some residual surface conductivity of the glass substrate next to IDE structure. At the high-frequency side, there is a rise in \( C_\varepsilon '(f) \) above 1 MHz, which can be attributed to inductive contributions that have not been explicitly considered in the equivalent circuit.

Based on our simplified equivalent circuit (according to Figure 5), we were able to model the dissipated electrical power \( (P) \) (being approximately proportional to \( \Delta T \)) as a function of the amplitude and frequency of the AC electric field. The result is given in Figure 7 showing the frequency dependence of \( P \) for different voltage amplitudes applied to the empty CLC device (details can be found in the Supporting Information). While at lower frequencies (below 3 MHz) the power scales with \( P(f) \propto f^2 \) as indicated by the linear rise in the double-log representation, this regime turns to a power saturation regime above 10 MHz. This crossover actually means that at low frequencies, the power increase is controlled by the dominant capacitive reactance \( (X_\varepsilon (f) \gg R) \), while above \( \approx 10 \text{ MHz} \), the ohmic resistance limits the total impedance resulting in a frequency independent power depicted by the plateau region. Consequently, though operation of the CLC cell in this plateau region

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**Figure 4.** a) Temperature increase \( (\Delta T) \) of the CLC device as a function of the applied voltage with \( f = 250 \text{ kHz} \) or b) as a function of the frequency with \( U = 15 \text{ V} \). A waiting time of 5 min between each measurement was respected to achieve equilibrium conditions.

**Figure 5.** Simplified equivalent electrical circuit of the empty \((C_\varepsilon \text{ and } R)\) and filled \((C_\varepsilon + \Delta C_{\text{LC}} \text{ and } R)\) electrothermal CLC device.
would be most efficient regarding maximum power dissipation, the existence of the crossover behavior (0.1–10 MHz) allows to tune the electrothermal heating by variation of the applied frequency, voltage and/or IDE pattern.

3. Conclusion

An electrothermal responsive CLC device showing color tuning with high optical quality over the entire visible spectrum has been presented. The reflection band can be tuned autonomously via variations in the surrounding temperature or manually by applying an electric field to the system. This allows for easy and precise tuning of the response at any moment, offering a user-friendly device that functions with or without a power input. Unique to the induced response is the use of an in-plane electric field provided by the IDE pattern. Enhancing the amplitude and/or frequency of the AC electric field caused significant temperature increases inside the cell, which resulted in band gap shifting up until the isotropic phase was reached. Our findings derived with BDS confirmed that the initiated electrothermal response is predominantly caused by resistance heating at the IDE pattern. The dissipated power, being related to the maximum achievable temperature increase, is also depending on the voltage amplitude and frequency by virtue of capacitive coupling via the cell capacitance, showing that the electrothermal response can be enhanced up until a specific frequency is reached. In addition, the interdigitated ITO pattern can be replaced by different conductive materials and fabricated in all sorts of patterns (e.g., meandering pattern) which could improve the electrothermal response and allows color patterning.

The use of a single conductive substrate to induce CLC color tuning could simplify future devices. Another possibility is the development of flexible electro-optical devices by using flexible conductive substrates. Introducing flexibility to the current system could expand the application range and is for instance crucial when working with curved surfaces. These features of manual control over an otherwise autonomous response, combined with a large potential for device optimization, shows the potential of this technique in the field of optoelectronics, actuators and smart windows.

4. Experimental Section

Materials: Nematic liquid crystalline mixture MLC-2138 and chiral dopant S811 were purchased from Merck and used without any further purification. A thermal responsive CLC mixture, containing MLC-2138 (65 wt%) and S811 (35 wt%), was stirred at 70 °C to obtain a homogeneous CLC mixture. Interdigitated ITO patterned glass plates with electrode width \( w = 13 \mu m \) and spacing \( s = 10 \mu m \) were purchased from Suzhou Walthy Co., Ltd.

Sample Preparation: ITO patterned \((2.5 \times 2 \text{ cm}^2)\) and pristine glass slides \((3 \times 3 \text{ cm}^2)\) were used, respectively, as bottom and top substrate for the final cell structure. The substrates were cleaned by ultrasonication in isopropanol for 20 min and subjected to UV-ozone treatment (UV Products, PR-100) for an additional 20 min. Polyimide (Optimer AL 1051, JSR Corporation) layers were spin-coated on top of the substrates. A dual-step spin coat procedure was carried out by rotating the sample at 800 rpm for 5 s, followed by rotating at 5000 rpm for 40 s. The coated polyimide layer was cured on a hotplate at 100 °C for 15 min and subsequent curing was done in an oven at 180 °C for 90 min. The ITO patterned and normal glass plates were rubbed with a velvet cloth before assembly. The substrates were placed...
with rubbing direction perpendicular to each other to obtain planar alignment. Both glass slides were glued together with UV curable glue (UVS 91, Norland Products Inc.) containing 6 µm spacer beads to control the cell thickness. The glue was cured for 10 min using a low-intensity UV-lamp (4 Philips CLEO 15W lamps). The cells were capillary filled with the CLC mixture at 35 °C and cooled to room temperature before usage.

Characterization: Thermal transition points of the CLC mixture were determined via differential scanning calorimetry (DSC) using a TA instrument (DSC Q2000). DSC measurements consisting of three repeating cycles were performed in a temperature range from −10 °C to 90 °C with heating and cooling rates of 2 °C min⁻¹. An in-plane AC electric field was applied to the CLC cell via the IDE pattern with an electrically activated area of 1 × 1 cm² and was generated as a square pulse function by a function generator (33220A, Agilent). All studies with constant voltage and increasing frequency were carried out by using the function generator, while the studies with constant frequency and increasing voltage were performed by coupling the function generator to a voltage amplifier (F20A, FLC Electronics) to reach higher voltage amplitudes. The output voltage and frequency of the applied in-plane AC electric field were measured with an oscilloscope (Tektronik TDS 210). The voltage amplitudes were expressed as AC root mean square (RMS) value V_{RMS}. Transmission spectra were performed at room temperature and measured by a Shimadzu UV-3102 PC UV/vis/NIR spectrophotometer equipped with an MPC-3100 sample compartment. A Linkam TMS93/LMP93 temperature control stage was installed in the spectrophotometer to analyze the thermal behavior of the CLC cells. A single ITO patterned glass plate was used as a baseline. Temperature changes were recorded by a visual IR thermometer (Ti32 Infrared camera, Fluke). Broadband dielectric spectroscopy measurements (BDS) were performed at room temperature employing a high-precision impedance analyzer (ALPHA ANB, Novocontrol Technologies). Spectra of the complex capacitance C''(f) = C'(f) – iC''(f) were acquired in the frequency range from 1 Hz to 20 MHz and fitted to common model response functions such as the Debye model or the Havriliak–Negami function; more details are given in the Supporting Information or in additional references.[45,46]

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

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Conflict of Interest
The authors declare no conflict of interest.

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