

# Facile Anisotropic Deswelling Method for Realizing Large-Area Cholesteric Liquid Crystal Elastomers with Uniform Structural Color and Broad-Range Mechanochromic Response

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Cholesteric liquid crystal elastomers (CLCEs) are soft and dynamic photonic elements that couple the circularly polarized structural color from the cholesteric helix to the viscoelasticity of rubbers: the reflection color is mechanically tunable (mechanochromic response) over a broad range. This requires uniform helix orientation, previously realized by long-term centrifugation to ensure anisotropic deswelling, or using sacrificial substrates or external fields. The present paper presents a simple, reproducible, and scalable method to fabricate highly elastic, large-area, millimeters thick CLCE sheets with intense uniform reflection color that is repeatably, rapidly, and continuously tunable across the full visible spectrum by stretching or compressing. A precursor solution is poured onto a substrate and allowed to polymerize into a 3D network during solvent evaporation. Pinning to the substrate prevents in-plane shrinkage, thereby realizing anisotropic deswelling in an unprecedentedly simple manner. Quantitative stress–strain–reflection wavelength characterization reveals behavior in line with theoretical predictions: two linear regimes are identified for strains below and above the helix unwinding threshold, respectively. Up to a doubling of the sample length, the continuous color variation across the full visible spectrum repeatedly follows a volume conserving function of the strain, allowing the CLCE to be used as optical high-resolution strain sensor.

## 1. Introduction

Cholesteric liquid crystals (CLCs) spontaneously form a supramolecular helix with a pitch (period)  $p$  that can be in the range of a few hundred nanometers. This leads to Bragg reflection (structural color) of circularly polarized light with the same


handedness as the CLC helix, within a narrow band centered around the wavelength (as measured inside the CLC) equal to  $p$ .<sup>[1]</sup> The cholesteric helix pitch, and thus the reflection color, can be easily tuned by temperature,<sup>[2,3]</sup> electric field,<sup>[4,5]</sup> and light.<sup>[6–8]</sup> The responsiveness to different external stimuli is of great use for various applications, including sensors<sup>[9,10]</sup> and dynamic color tuning.<sup>[11–14]</sup> If a CLC is polymerized into an elastomer (CLCE), a very interesting material arises, combining the striking optical properties of a cholesteric phase with the viscoelasticity of a rubber.<sup>[15,16]</sup> Because of the coupling between the cholesteric helix and the elastomeric network, a mechanical deformation changes the reflection color and often also the polarization.<sup>[17]</sup> A striking demonstration was the continuous and reversible tuning toward shorter wavelengths of a CLCE film with vertical helix by stretching biaxially in the film plane, equivalent to a uniaxial compression along the helix.<sup>[18–20]</sup> This mechanochromic response was then used for tunable lasing controlled

by stretching the film.<sup>[18–22]</sup> This was also achieved using viscous liquid-state cholesterics, but the non-elastomeric nature of the helically modulated medium then required sandwiching between rubbery substrates.<sup>[23]</sup>

As color is one of the most natural indicators seen by the naked eye, the mechanochromic response of CLCEs is attractive far beyond lasing. CLCEs could enable tensile and/or pressure sensors capable of visualizing mechanical strain in many objects of significant societal, technical, and/or commercial importance, such as clothing, packaging, pressurized vessels or in various architectural features.<sup>[13]</sup> Picot et al. demonstrated such a strain sensor operating in real-time, albeit with an optical response range no greater than  $\approx 40$  nm (from orange to green)<sup>[24]</sup>; this limitation can be traced back to the bilayer nature of the sensor, the cholesteric being supported on a nylon substrate that restrains the overall elasticity. Of particular interest with a CLCE strain sensor that needs no support is—beyond the broad-range response given by a fully elastic material—that the response is local, that is, the color change in a certain point of the CLCE reflects the strain at that very location, enabling

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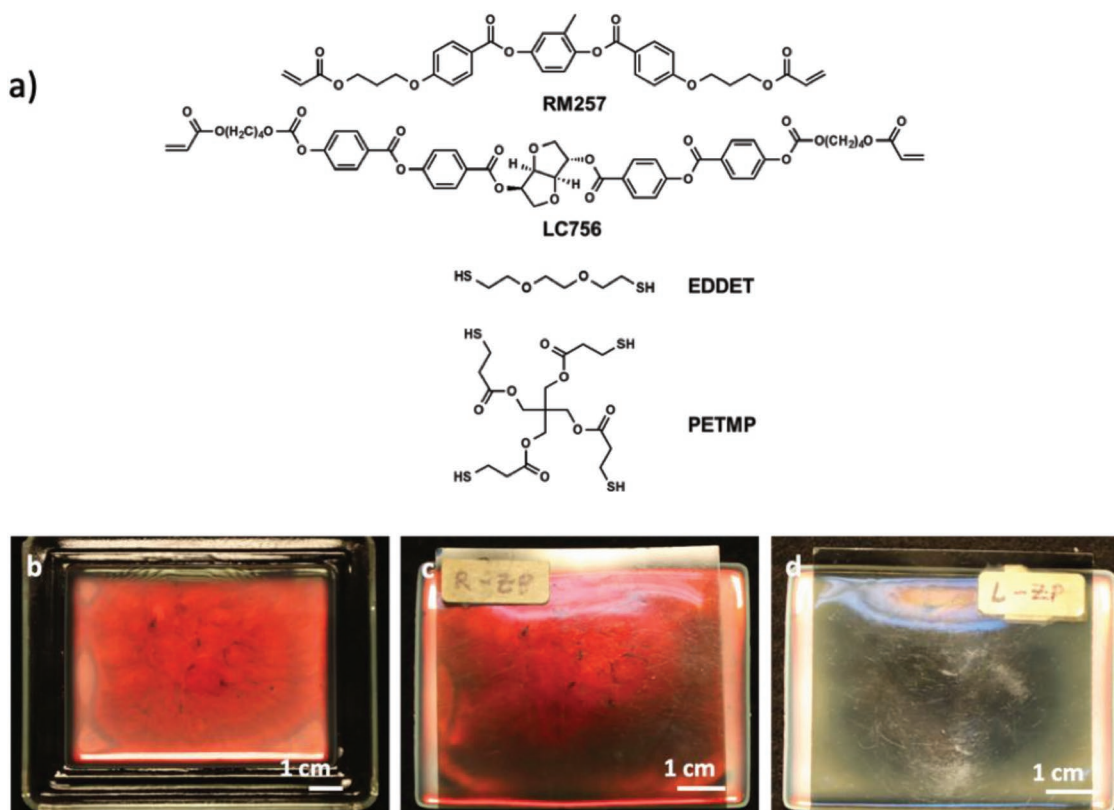
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pressure/tensile sensing with unparalleled spatial resolution. Moreover, CLCE-based sensors are autonomous, they require no electricity to operate, they can be designed to work over a range of temperatures including room temperature, and their response can be read out by the naked human eye or by standard video cameras. They are thus not subject to the conundrum of moving toward ubiquitous deployment of sensors for big data applications, including applications in wearable technology and robotics.

So far, however, no commercial application of CLCEs has been attempted, a situation we may largely trace back to the difficulties of the methods reported so far for producing high-quality CLCEs with convenient elastic modulus as well as area and thickness. Especially when large strains are expected in the absence of excessive forces, as for instance in wearable technology, the starting CLCE must be soft in order not to restrain mobility, and have a sufficient thickness in order not to rupture during use. The critical requirement to achieve uniformly vertical orientation of the CLCE helix was first solved by the Finkelmann group with the “anisotropic deswelling” method.<sup>[25]</sup> Their solution involved continuous centrifugation at high temperature for 10 h, a hydrosilation reaction taking place during the first half, followed by solvent evaporation during the second half. While the method yields impressive results and subsequent work reduced the centrifugation time to between 5 and 8 h,<sup>[26,27]</sup> it is cumbersome, and scale-up appears challenging. More recent approaches utilized electric/magnetic fields or surface

alignment from sacrificial or integrated substrates,<sup>[20,24,28,29]</sup> but scalability and robustness are still critical issues. The procedures involve, for example, mixing of reactive monomers at high temperature,<sup>[30]</sup> breaking open sandwich cells and peeling off CLCE films from the substrates without rupture,<sup>[20,29]</sup> soaking in solvents to remove unreacted materials,<sup>[20,30]</sup> and sometimes custom synthesized chemicals are required.<sup>[29,31]</sup> To fully exploit the technological potential of CLCEs, a significantly simplified and robust procedure to make unsupported highly elastic CLCEs is needed, allowing a fast and reversible color shift across the full visible spectrum in response to strain.

Here, we report an elegantly simple, highly reproducible, and easily scalable method to prepare uniformly colored large-area CLCE films that show robust, rapid, and reversible mechanochromic response to stretching and compression under loads typical of those delivered by a human, using only off-the-shelf commercial chemicals. Our method is based on the two-stage thiol-acrylate Michael addition and photopolymerization reaction reported by Yakacki et al.<sup>[32]</sup> for making non-chiral nematic main-chain LCEs, the move to CLCEs being achieved by the introduction of the diacrylate chiral mesogen LC756 (Figure 1a; see Section 4 and Supporting Information for more details). While we also apply anisotropic deswelling to control the helix orientation, our modification allows us to carry out the procedure without centrifugation and near room temperature, simply casting the CLCE precursor solution onto a substrate, where it reacts to yield a CLCE film with uniform selective



**Figure 1.** a) Chemical structure of monomers used to prepare CLCE films. b) Photograph of a red-reflecting CLCE film just after first-stage Michael addition click reaction and solvent evaporation; c) freestanding CLCE film (7 cm × 5 cm × 0.1 cm) after photopolymerization viewed through right-handed (R-ZP) and d) left-handed (L-ZP) circular polarizer.

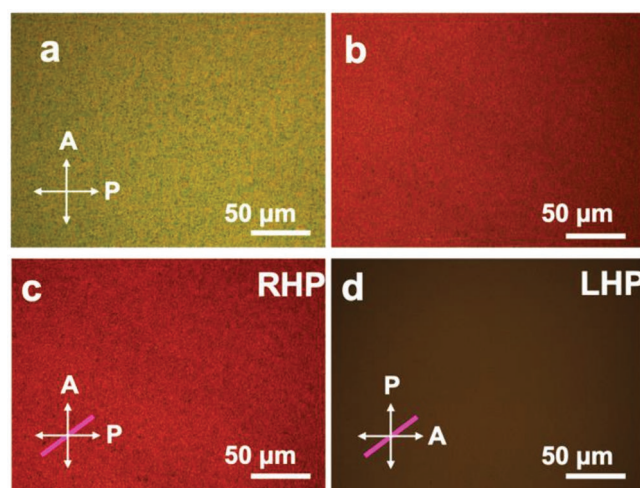
reflection color in the visible spectrum. The resultant film shows a dramatic blue shift while stretching the film uni- or biaxially orthogonal to the helical axis, and it immediately relaxes when the strain is removed, fully recovering the original state, even if the film was kept under strain for more than a day. We believe this easily translatable and simple mix-and-pour approach to fabricate high-quality autonomous tensile/pressure sensors will make a significant impact on the potential for commercial applications of CLCEs.

## 2. Results and Discussion

To prepare a CLCE that reflects red (green) color in the relaxed state, we let the chiral monomer LC756 make up 4.2 wt% (5.0 wt%) of the total diacrylate monomer mass, the remainder being the non-chiral RM257, which made up 100% of the diacrylate mass in the original Saed et al. formulation. Dipropylamine is added to initiate the first-stage click reaction between diacrylates and thiols, and Irgacure 651 serves as a photoinitiator for triggering a second-stage radical-induced photocrosslinking to make the CLCE structure permanent. In order not to initiate premature acrylate–acrylate crosslinking, all preparations are conducted in a yellow light environment. Directly after mixing, the CLCE precursor solution is poured onto a substrate and allowed to react at room temperature (25 °C) in a fume hood, without covering the sample. The reaction takes about 5 h but we keep the sample in the fume hood for 24 h in order to fully evaporate the toluene; active solvent extraction would allow this time to be significantly reduced. After confirming (by brief exposure to white light) that the desired reflection color has been achieved (Figure 1b), the film is irradiated with UV light to trigger the final crosslinking. The produced film reflects right-handed circularly polarized (RHP) light as confirmed by viewing it through right- and left-handed circular polarizers (LHPs) (see Figure 1c,d). From this result, we conclude that the CLCE helix is right-handed.

We carry out polarized optical microscopy (POM) to further elucidate the optical characteristics of the CLCE film (Figure 2). In accordance with the macroscopic behavior, the film shows a red color in reflection mode and appears dark when viewed through a left-handed polarizer. Microscopically, the film displays a slightly grainy texture, although on the macroscopic scale it looks highly uniform. Importantly, the optical quality of the CLCE film is consistent over large areas (we have produced films up to 35 cm<sup>2</sup>, but there is no reason to expect this to be an upper limit), and insensitive to the film thickness (our films range from  $\approx 100\ \mu\text{m}$  to 2 mm thickness; see Supporting information).

Figure 3 summarizes the behavior of an initially red-reflecting CLCE film to uniaxial stretching orthogonal to the helix axis at room temperature (see also Movies S1–S4, Supporting Information). The reflected color changes continuously and without delay from red to green and then to blue (Figure 3a–c). This process is fully reversible, with no visible lag between mechanical relaxation and color recovery, and the film returns to its original shape with red reflection when the film is allowed to relax. To quantify the color–strain response, we conduct uniaxial strain experiments with the sample kept

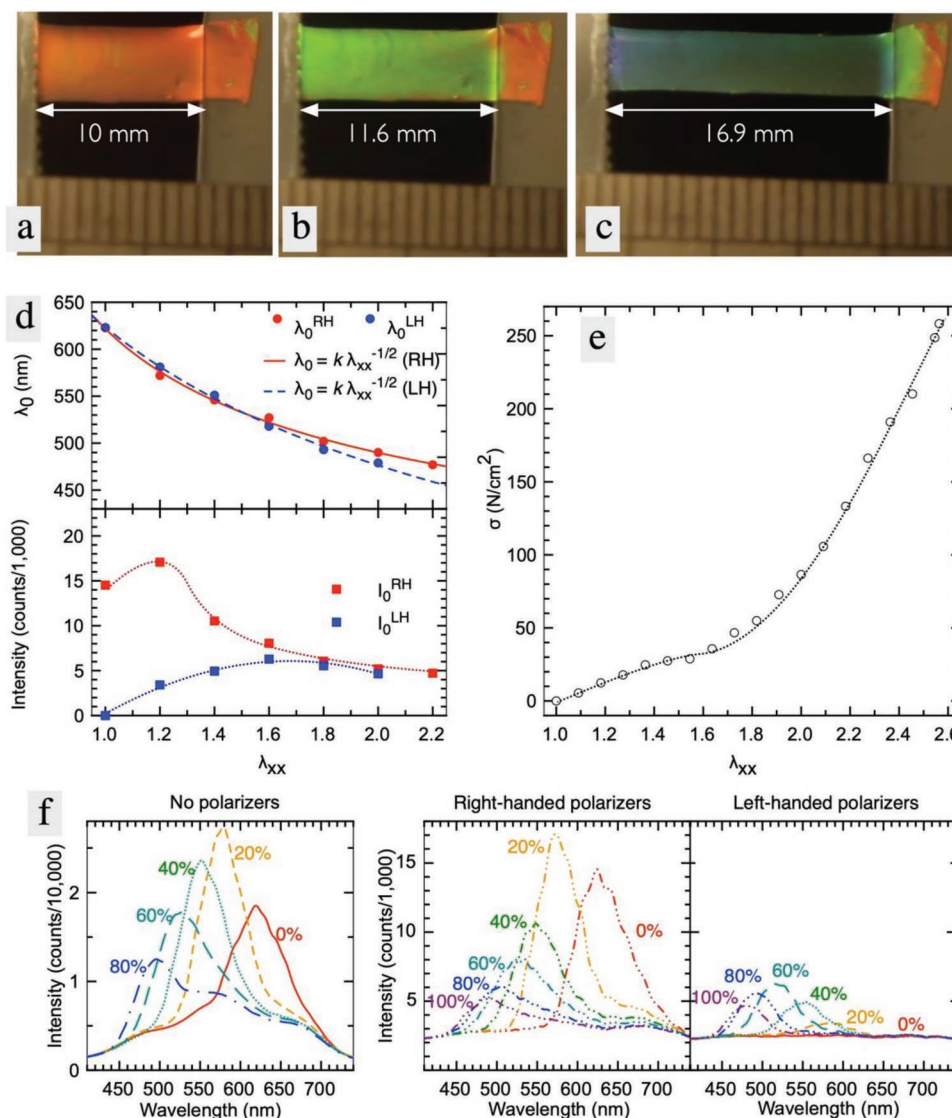


**Figure 2.** POM images of CLCE a) in transmission mode with crossed linear polarizers, b) reflection mode without analyzer and polarizer, c) reflection mode with right-handed circular polarizer (RHP), and d) reflection mode with left-handed circular polarizer (LHP). The purple line indicates the optic axis orientation of the lambda plate, inserted between the polarizer and analyzer; note that the latter two have their orientations exchanged between (c) and (d).

in the polarizing microscope, to which a spectrophotometer is attached, yielding the change in reflection peak wavelength  $\lambda_0$  and the maximum peak intensity  $I_0$  as a function of relative length change  $\lambda_{xx} = l/l_0$ , where  $l$  is the dynamic length and  $l_0$  is the initial length. These measurements (Figure 3d; raw reflection spectra in Figure 3f) are carried out without polarizers and with right-handed (RH) and left-handed (LH) circular polarizer and analyzer, respectively. Similar to earlier observations and theoretical predictions,<sup>[27]</sup> the film starts reflecting also in the left-handed channel when uniaxial stretching is applied (Figure 3d and Movie S2, Supporting Information). The reflection color change on subsequent stretching is similar for both RH and LH polarizations while subjected to uniaxial or near biaxial stretching (Movies S5 and S6, Supporting Information). Since loss of circular polarization of the reflected light is not expected for the uniaxial helix compression that corresponds to true biaxial stretching,<sup>[27]</sup> the fact that LH reflection appears in both cases suggests that the optical response of the CLCE is sensitive enough to detect the deviation from ideal biaxial stretching in this manually conducted experiment.

The peak reflection wavelength in either polarization channel can be fitted very well with a  $\lambda_0 \propto 1/\sqrt{\lambda_{xx}}$  function, as expected from volume conservation:<sup>[17,33]</sup> if the length increases by  $\lambda_{xx}$ , the width and thickness must each, in the first approximation, decrease by  $1/\sqrt{\lambda_{xx}}$ , and since the pitch scales with the thickness in a CLCE (see below), so must  $\lambda_0$ . Using a force gauge while stretching the film uniaxially while observing the color by a video camera (Movie 3, Supporting Information), we establish the stress–strain relationship up to the point of rupture at a relative length change  $\lambda_{xx} \approx 2.6$  (corresponding to an engineering strain of  $\varepsilon = \Delta l/l_0 = 1.6$ ). Interestingly, the range over which we are able to establish the stress–strain relationship is large enough to recognize the theoretically predicted behavior<sup>[17]</sup> with two regimes of linear behavior, below and above helix





**Figure 3.** Photographs through a right-handed circular polarizer of reversible mechanochromic tuning of selective reflection band by uniaxial stretching (a–c) (still images from Movie 1, Supporting Information). The reflection band peak  $\lambda_0$  (d: upper graph; lines are best fits to the data) and the maximum reflection intensity (d: lower graph; dotted lines are guides to the eye) are plotted against the relative length change  $\lambda_{xx} = l/l_0$  for right-handed (RH) and left-handed (LH) circular polarizer and analyzer, respectively. The stress  $\sigma$  is plotted as a function of  $\lambda_{xx}$  in (e) (dotted line is a guide to the eye). f) The raw reflection spectra corresponding to (d).

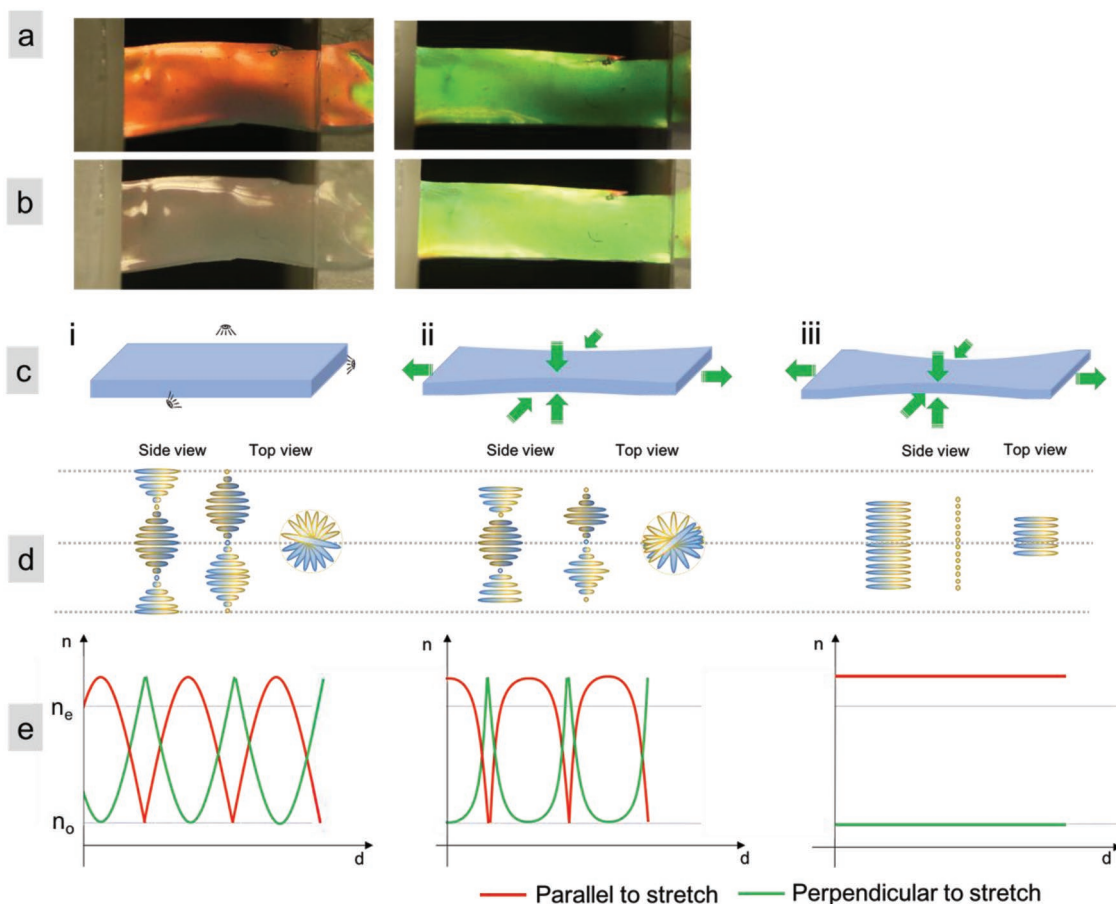
unwinding, separated by a strongly non-linear behavior while helix unwinding is taking place.

We also perform local uniaxial compression experiments against a glass plate, pressing with a glass rod, to compare the color shift with those seen under extensive strain. The change in color is illustrated in Movie 7, Supporting Information. Where it is compressed, the reflection turns from red to blue and returns to the original state swiftly upon release of the pressure. Movie 8, Supporting Information, further confirms that the color shift of the CLCE is local. For this experiment, half of the CLCE film is placed between two glass plates and a uniaxial stretch is applied to the freely suspended part. The free part of the film shifts the selective reflective band in response to the stretching, while, pleasingly, the film section not exposed to stretching retains its color, meaning that the CLCE response

is local. The local response gives the CLCE strong potential for pressure/tensile sensing with unparalleled spatial resolution.

To determine whether uniaxial stretching might deform the CLCE helix more seriously over time, to the extent that the periodicity slowly changes or is even lost, we maintained a film in the stretched state for an extended time (Figure S2, Supporting Information). Interestingly, the CLCE retains its mechanically induced blue color under constant stretching for 24 h. Upon relaxation, the original green color immediately reforms.

Following Terentjev and co-workers,<sup>[27]</sup> we give the following plausible reason for the change in color and polarization of the reflected light upon uniaxial stretching, schematically illustrated in the lower half of Figure 4. In the cholesteric phase, the local director is perpendicular to the helix axis, which we define along  $\hat{z}$ . The director rotates as we move along  $\hat{z}$ , as described by the



**Figure 4.** Deformation process of the helical structure under uniaxial stretching. CLCE film viewed through a) right-handed polarizer (RHP) and b) left handed polarizer (LHP). c) Schematic illustration of the macroscopic deformation of the film; i) relaxed film; ii) stretch prior to helix unwinding; iii) strong stretch with fully unwound structure (never obtained in our experiments). d) Side view (orthogonal to [left] and along stretching [right], respectively) and top view of the helical structure deformation. e) The refractive index projection on the direction parallel and perpendicular to the stretch along the film thickness direction  $z$ .

linear director angle function  $\varphi(z) = (2\pi/p)z + \varphi_0$ . Here,  $\varphi_0$  is the director angle at a certain starting point  $z = 0$ ,  $p$  is the pitch, and  $z$  is the distance along the helix, as shown in Figure 4d. By applying uniaxial strain in the plane perpendicular to  $\hat{z}$ , the director angle loses its linear dependence on  $z$ , giving rise to a distorted helix in which some sections exhibit a compressed twist while others approach an unwound state. The helical structure is deformed, but the structure is still periodic, now with a shorter pitch due to the helix compression that accompanies the stretching as a result of volume conservation, and the refractive index projection parallel and perpendicular to the stretch is shown below. For the undeformed ground state, we have:

$$n_{\parallel} = n_e \cos\left(\frac{z}{p} + \varphi_0\right) + n_o \sin\left(\frac{z}{p} + \varphi_0\right) \quad (1)$$

$$n_{\perp} = \sqrt{(n_e^2 + n_o^2)} \sin\left(\frac{z}{p} + \varphi_1\right) \quad (2)$$

$$\varphi_1 = \varphi_0 + \tan^{-1} \frac{n_e}{n_o} \quad (3)$$

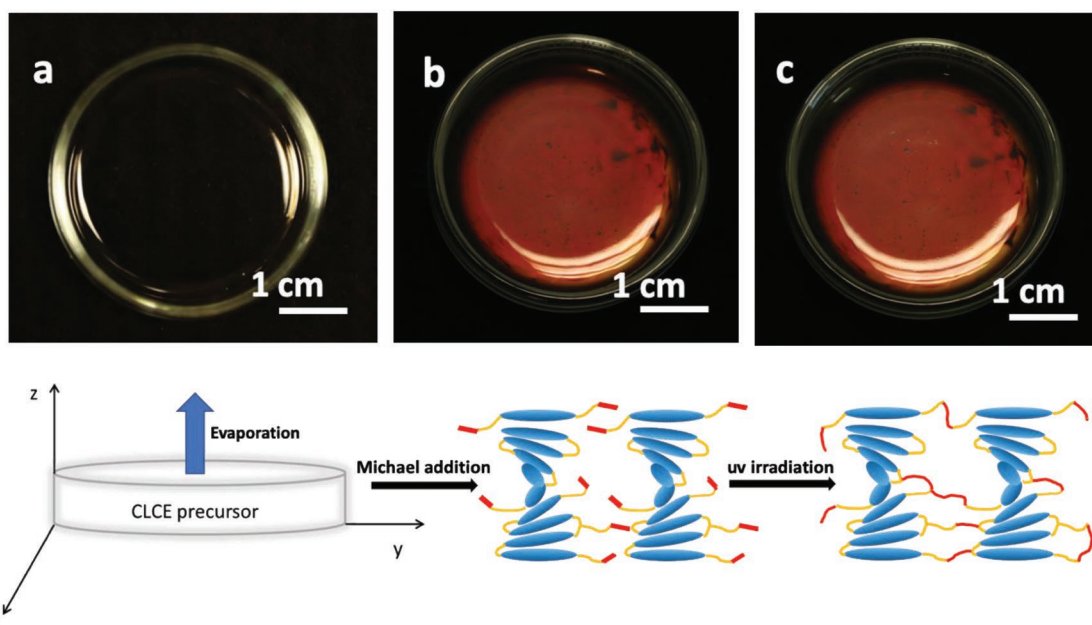
$$n_{\perp} = n_o \cos\left(\frac{z}{p} + \varphi_0\right) + n_e \sin\left(\frac{z}{p} + \varphi_0\right) \quad (4)$$

$$n_{\perp} = \sqrt{(n_e^2 + n_o^2)} \sin\left(\frac{z}{p} + \varphi_2\right) \quad (5)$$

$$\varphi_2 = \varphi_0 + \tan^{-1} \frac{n_o}{n_e} \quad (6)$$

Figure 4e sketches how the refractive indices are modulated under various states of strain. The film thus still yields Bragg reflection, at a color corresponding to shorter wavelength, but the polarization is no longer circular. At yet higher stretching, the helix starts unwinding, eventually yielding a uniform director structure if the film would support it, as illustrated on the far right in Figure 4d.

Why does our CLCE develop with strong uniform color and vertically oriented helix throughout the sample, although we do not apply centrifugation, alignment layer, or an external magnetic or electric field for aligning the sample? We propose

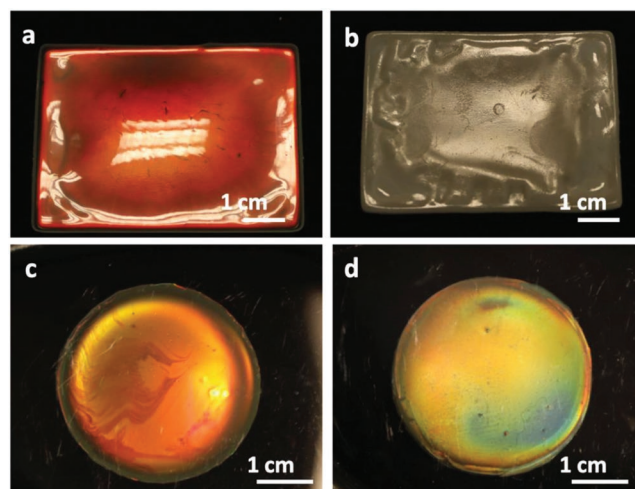


**Figure 5.** Photographs of preparation of CLCE film and corresponding schematic drawing of cholesteric network formation with the helical axis along the direction of evaporation, that is, along the  $z$ -axis. a) CLCE precursor solution in a glass petri dish; b) the appearance of CLCE reflection color after first stage thermal cure and evaporation of the solvent; c) CLCE after photopolymerization. The initial cholesteric liquid crystalline network formed by Michael addition reaction cannot shrink in the sample plane because it is already cross-linked and pinned to the bottom of the petri dish. Finally (right side), the system is photopolymerized to arrest the CLCE state.

that we do in fact benefit from anisotropic deswelling, but it is realized in a very much simplified manner compared to the original Finkelmann approach,<sup>[25]</sup> as illustrated schematically in **Figure 5**. A key element is, we believe, the gel formation during the initial click polymerization of acrylates and thiols in the precursor mixture:<sup>[32]</sup> it appears that the gel forming when using this chemistry adheres so strongly to the substrate that it cannot shrink in the lateral dimension as the solvent evaporates, as evidenced from the unchanged surface area of the sample during drying (see **Figure 5b**). This restricts the deswelling to take place only in the vertical ( $\hat{z}$ ) direction as the solvent evaporates, causing an in-plane orientation of the liquid crystal director and thus a vertical helix axis, that is, along  $\hat{z}$ . With our simple mix-and-pour method, we thus achieve the same beneficial anisotropic deswelling method for aligning the CLCE helix as reported by Finkelmann and co-workers, with no need for centrifugation.

To the best of our knowledge, no prior publications on CLCEs formed using the anisotropic deswelling method explicitly discuss potential differences between the bottom and top surfaces of the produced films. **Figure 6a,b** shows both sides of one of our CLCE films that has been made by anisotropic deswelling from the glass substrate of a petri dish. The bottom of the film, that is, the CLCE side that was in contact with the substrate during the reaction exhibits no selective reflection. It scatters light strongly, yielding a colorless appearance. We believe that this is due to the obstructed solvent evaporation through the glass petri dish, preventing unidirectional compression on this side of the CLCE film as it forms. To confirm this, we perform an additional experiment in which the precursor mixture is poured into a mold made of a metal ring covered with a thin

film of cured polydimethylsiloxane (PDMS) on one side, while the other side is kept open to have natural evaporation. The PDMS layer is swellable by the solvent used for the precursor mixture, thus enabling removal of solvent through the bottom as well as toward the top. Indeed, a CLCE prepared in this mold



**Figure 6.** Photographs of a CLCE film (0.8 mm thickness) made by evaporation from a glass petri dish; the red reflection is strongly seen from the top (a) but from the bottom (b), the film appears a dull grey-white due to strong scattering from a poorly aligned structure. In contrast, a CLCE film (1 mm thick) prepared in a mold made of a metal ring and a PDMS base shows strong reflection color both from the top side (c), exposed to air during solvent evaporation, and from the bottom side (d), exposed to the PDMS base.



shows color on both sides, (see Figure 6c,d) a very beneficial advantage. This result further confirms that the evaporation along a single dimension of solvent from the CLCE precursor solution during the formation of the cholesteric phase is crucial to get a well-aligned cholesteric network in the CLCE.

To quantify the effect, we prepared two new films on PDMS and on glass, respectively, placing them over a graphical background to compare the transparency as seen by the eye, and also measuring the normalized transmission with a spectrophotometer attached to a microscope (see Figure S3, Supporting Information). Although the PDMS-made film is three times thicker (2 mm) than the glass-made film (0.6 mm), the transparency is significantly higher, ranging from 70% to 90% outside the cholesteric reflection band, compared to 20–45% for the glass-made CLCE. While both sides of the PDMS-made CLCE shows the red color of the relaxed cholesteric, the glass-made film is much whiter and less colorful when viewed from the side that was in contact with the glass. Interestingly, the spectrophotometer investigation reveals that the reflection band occurs at somewhat shorter wavelength for the PDMS-made film, possibly indicating that the more efficient anisotropic deswelling leads to a somewhat compressed helix in the relaxed state, compared to the film made on glass.

We also investigated the effect of heating the precursor solution during the reaction (see Figure S4, Supporting Information). Heating to about 47 °C already leads to significant variations in the color of the final film, and higher temperatures yield a strongly scattering white film, with color observable only at the sample edges. This gives evidence of the importance of the anisotropic deswelling process while the liquid crystalline order develops: if the solvent evaporates too rapidly, as is the case at high temperature, the uniform vertical helix alignment is not obtained.

Finally, we investigate if the bounding surfaces might have a role in aligning the helix, specifically comparing the behavior on hydrophilic and hydrophobic surfaces. We pour the CLCE precursor solution into a petri dish half coated with dimethyloctadecyl[3-(trimethoxysilyl)propyl]ammonium chloride (DMOAP) and the other half untreated, to ensure otherwise identical experimental conditions. The hydrophobic nature of the DMOAP treatment, as well as an air interface, might be expected to align the molecules closest to the interface normal to the film; hence, different CLCE alignments might be expected for the two substrate halves. However, we do not observe any significant difference between the two cases, both sides showing strong red reflection in the ground state (see Figure S5, Supporting Information.). This result demonstrates that surface treatment is not necessary to achieve the uniformly aligned helix in the CLCE. In fact, since the network starts developing in the solvent-enriched isotropic precursor solution, no effect of the interfaces on the alignment of the liquid crystal state should be expected.

### 3. Conclusion

In conclusion, we have presented a facile and highly reproducible procedure to fabricate CLCEs that we believe can easily be scaled up for industrial application. A simple evaporation of

solvent from the CLCE precursor solution results in a macroscopic uniformly colored CLCE, thanks to the adherence to the substrate of the gel that forms from the very beginning. Interestingly, the CLCE film shows a reversible and immediate blue shift in reflection color, covering the full visible range from red to blue, when subjected to uniaxial or biaxial strain orthogonal to the cholesteric helix axis. As the color shift reflects a compression of the cholesteric helix, the CLCE reflection blue-shifts also in response to pressure. The response to stretching as well as to pressure is local and quantitatively related to the mechanical strain in the film, meaning that we can trace a complex strain pattern within the CLCE just by evaluating its color across its surface. We believe that the scalable and straightforward method of preparation of CLCE and its rapid and reversible structural color change to any mechanical deformation, coupled with the general benefits of CLCEs such as autonomous operation over a broad temperature range including room temperature, independent of electrical power, open the door for a variety of applications in flexible optics, sensing, etc. For example, we are now exploring the incorporation of our CLCE into fibers that could be incorporated into smart textiles for strain/pressure sensing in a variety of contexts, from wearable technology to indicators that give early warning signs of imminent failures in bridges and other constructions.

### 4. Experimental Section

**CLCE Fabrication:** Molecular structures of monomers used to prepare CLCE films are shown in Figure 1a. The diacrylate mesogen, 1,4-bis-[4-(3-acryloyloxypropyloxy)benzoyloxy]-2-methylbenzene (RM257) (Wilshire Technologies), and the chiral dopant (3R,3aS,6aS)-hexahydrofuro[3,2-b]furan-3,6-diyl bis(4-((4-(3-acryloyloxy)butoxy)carbonyloxy) benzoyloxy) benzoate (LC756) (Synthon Chemicals) were heated to 80 °C for 5 min. and cooled to room temperature. Then, a tetra-functional thiol cross-linker pentaerythritol tetrakis(3-mercaptopropionate) (PETMP) (Sigma Aldrich), a dithiol monomer 2,2-(ethylenedioxy) diethanethiol (EDDET) (Sigma Aldrich), a photoinitiator 2,2-dimethoxy-2-phenylacetophenone, Irgacure 651 (Sigma Aldrich) were mixed with the solution. Lastly, dipropylamine diluted to 1:50 ratio with toluene was added as a catalyst for the first-stage Michael addition reaction. The solution was stirred for 5 min. and poured into a glass petri dish which was left open for 24 h. Subsequently, the CLCE film was irradiated with UV light at 365 nm with an intensity of 32 mW cm<sup>-2</sup> for 5 min at 25 °C. All chemicals were used as received.

**CLCE Characterization:** The macroscopic samples, in the stretched state and relaxed state, were filmed using a Canon EOS camera. For microscopic observation, the samples were imaged using an Olympus BX51 polarizing optical microscope. Stress-strain measurements were conducted by mounting films cut to well-defined dimensions in a microscopy sample translator (Olympus), filming or measuring the transmission or reflection spectrum as a function of the strain. The spectra were obtained with an Avantes AvaSpec-2048 spectrophotometer connected via optical fiber to the polarizing microscope. The stress was obtained by measuring the stretching force using a Mark-10 force gauge (Model M3-5).

### 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.

## Keywords

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