Electronic supporting information

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Experimental procedures

Preparation of LC mixture: Two different reactive mesogens are combined in a glass vial in a 1-to-1 weight ratio, 4-methoxyphenyl-4-(6-acryloyloxyhexyloxy)benzoate (**1**) and 1,4-di(4-(6-acryloyloxyhexyloxy)benzoyloxy)-2-methylbenzene (**2**) (both obtained from Merck KGaA), making up 98 wt% in the total mix. 1 wt% of reactive photoresponsive dye *N*-ethyl-*N*-(2-hydroxyethyl)-4-(4-nitrophenylazo)aniline is added (**3**, obtained from Sigma-Aldrich). Finally, 1 wt% of bis(2,4,6-trimethylbenzoyl)phenyl phosphine oxide is added to allow for photoinitiated free radical polymerization (**4**, obtained from Sigma-Aldrich). To the solid mix, dichloromethane (DCM, obtained from Biosolve) is added to allow for proper mixing for about 30 minutes. The vial is then moved to a hot plate at 80 °C where it remains overnight, and after this, a few hours in a vacuum oven at room temperature. At this point, near-complete removal of the solvent is assumed.

Fashioning of wedge-shaped LC alignment cells: 3×3 cm² borosilicate glass slides are sonicated (Branson 2510) for 20 minutes in a mixture of 1-to-1 ethanol-isopropanol (volume-based). Subsequently, they are blown dry under an N_2 flow and placed in a UV/O_3 oven (UV Products PR-100) for 20 minutes to activate the glass's surface. After this step, a polyimide (PI) precursor is spincoated (5 s at 800 rpm followed by 45 s at 5000 rpm); depending on the desired boundary condition, the polyimide used is Optmer AL1051 (JSR Corp.) for planar alignment, or Sunever 5661 (Nissan Chemical Corp.) for homeotropic alignment. Two curing steps follow: ±10 minutes at 90 °C, after which the slides are baked 90 minutes at 180 °C.

Uniaxial planar alignment is induced by rubbing PI-coated slides on a velvet cloth. Wedge-shaped, splay-aligned cells are made by gluing planar- and homeotropic-aligned glass slides together with coated sides facing each other. The wedge shape of the cell is made by gluing one side with UV-curable glue containing 30-70 μm diameter beads, and the other side with the same glue containing beads of a smaller diameter, or no beads at all. Quality of the wedge-shape is judged by looking for a lamellar-shaped optical interference fringe pattern when the cell is viewed under a bright light source.

Fabrication of the LCNs: Wedge-shaped cells were filled with the liquid crystal mixture at 90 °C through capillary action. After cooling down to 55 °C (to the nematic phase of the LC mix), a UV flood exposure is initiated and maintained at 3-5 mW cm⁻² for 900 seconds (EXFO Omnicure S2000) to facilitate free radical polymerization. A post-curing step of 20-30 minutes on a 120 °C hot plate is done to ensure maximum conversion and remove shrinkage-related stresses after polymerization. Cells are opened, and shapes are cut from the square polymer films using a razor blade.

Materials characterization: Differential scanning calorimetry was performed with a TA Instruments DSC Q1000. Fourier-transform Infrared (FT-IR) spectroscopy was performed with a Varian 670-IR in attenuated total reflectance (ATR) mode.

Analysis of thermomechanical behaviour: Cut films are placed in an oven with a window in the door to allow filming of the samples as the environment heats up. The temperature is recorded using a digital sensor (Sensirion SHTC1), photographs are taken with a digital camera (Nikon D7200, Olympus OM-D E-M10 Mk III) in manual mode. ImageJ is used to quantise the bending by fitting a circle to the curvature at 1/5th the film's length from the fixation point.[2] Mechanical properties of the materials are determined with a TA Instruments DMA Q800. Temperature-dependent storage and loss moduli (*E'* and *E''*) and dissipation factor tan(δ) are measured at 1 Hz oscillation frequency, 10 μm strain amplitude and a temperature ramp of 3 °C min[-1]. Coefficients of thermal expansion (α) are approximated through DMA in constant stress mode as a function of temperature with a preload on the film of 5 mN, in a similar procedure as Verpaalen *et al.*[1]

Analysis of photoactivated behaviour: The azobenzene-doped films are held tight with self-locking tweezers and placed at 10 cm from two 455 nm LED light sources (Thorlabs M455L3) placed to the left and underneath of the sample. The film is placed such that upon actuation, it will bend into the light as to prevent self-shading. To actuate, the LED controller is turned on (Thorlabs DC4104) and set to 700 mA, corresponding to about 310 mW cm[-2] per LED, after focussing the light beam. LED collimators (Thorlabs SM2 P50A) are adjusted to focus the LED light on the sample films. After actuation, photographs and videos are recorded using an Olympus OM-D E-M10 Mk III digital camera.

Thickness and molecular alignment characterization: Film thickness measurements were done with a surface profilometer (Bruker Dektak XT) or an interferometer (Fogale Nanotech Zoomsurf 3D). For scanning electron micrographs, LCNs were first fractured under cryogenic conditions (in liquid N_2), followed by an Au sputter coating treatment. Pictures were subsequently recorded using a FEI Quanta 3D SEM, and analysed with ImageJ.[2]

Finite element modelling of the wedge-shaped strips: A coupled thermomechanical model is developed to simulate the behaviour of tapered thin films in a commercial finite element software ABAQUS. The model incorporates geometrically nonlinear deformation to simulate the system with large rotation but with small strains. The constitutive equations to implement the thermomechanical model are presented in the following.

The thermal strain developed due to the temperature change can be formulated as,

$$\varepsilon_{ij}^{t} = \alpha_{ij}\Delta T$$

where, ε_{ij}^{t} is the thermal strain tensor, α_{ij} is the thermal expansion coefficient tensor and ΔT is the temperature change. The total strain is estimated as the sum of the elastic and thermal strains as,

$$\varepsilon_{ij} = \varepsilon_{ij}^{e} + \varepsilon_{ij}^{t}$$

where, ε_{ij} is the total strain and ε_{ij}^{e} is the elastic strain. Now, assuming a linear stress-strain relationship, the stress can be estimated as,

$$\sigma_{ij} = C_{ijkl}\varepsilon_{kl}^{e}$$

where, σ_{ij} is the stress tensor and C_{ijkl} is the elasticity tensor. The film is modeled as linearly elastic and transversely isotropic material with elastic moduli (E' and E'') measured as a function of temperature for both directions (see Figure S4). The value of the Poisson's ratio is

taken as $\nu_{12} = \nu_{23} = \nu_{13} = 0.3$. The values of the thermal expansion coefficient tensor (α_{ij}) are taken from thermal strain measurements as function of temperature (see Figure S6) to incorporate its anisotropic nature. The film is geometrically tapered along the length direction as shown in Scheme S1.

Composite shell elements (S4R) with discrete layers are used to model the variation of alignment along the thickness. The film is divided into three discrete layers of different thickness. The films with a total thickness in the range of 30 μm have 25 % planar, 25% "45°" and 50% homeotropic orientations (see Figure S5). Hence, the film is modelled with the respective alignment distribution. Scheme S1 shows the geometric conditions, boundary conditions and the variation of alignments through the thickness.
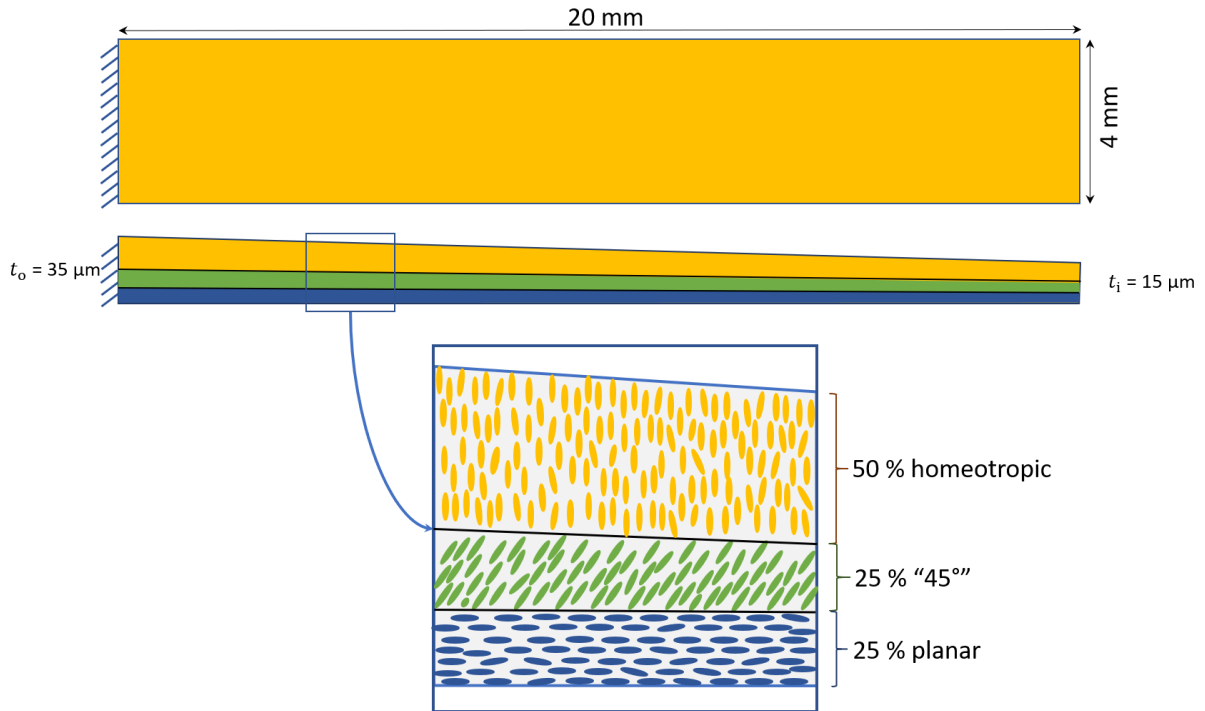
Scheme S1. A schematic showing the geometric conditions and film alignment distribution along the thickness used in the finite element model.

Validation of the finite element model: The finite element model is validated by comparing the results for planar/homeotropic bilayers against the analytical solution derived by Timoshenko[3] for a bilayer film. For a bilayer film with total thickness as H and equal layer thicknesses, the radius of curvature R can be estimated as,

$$R = H \frac{m + m^{-1} + 14}{24(\alpha_1 - \alpha_2)\Delta T}.$$

Here, $m = \frac{E_1(1 - \nu_1)}{E_2(1 - \nu_2)}$, E_i, ν_i and α_i $(i = 1, 2)$ are the elastic modulus, Poisson's ratio and the coefficient of thermal expansion, respectively for the layer i and ΔT is the change in temperature. Finite element simulations are performed with different values of m and different element sizes for a bilayer tapered film of length 20 mm, width 5 mm and thickness as 50 μm at the fixed edge and 20 μm at the free edge. Comparison of the radius of curvatures

estimated from the finite element simulations with that of the analytical solution shows good correspondence. An element size of 0.1 mm is found to achieve mesh convergence and hence an element size of 0.1 mm is used in all the simulations.

ESI Bibliography

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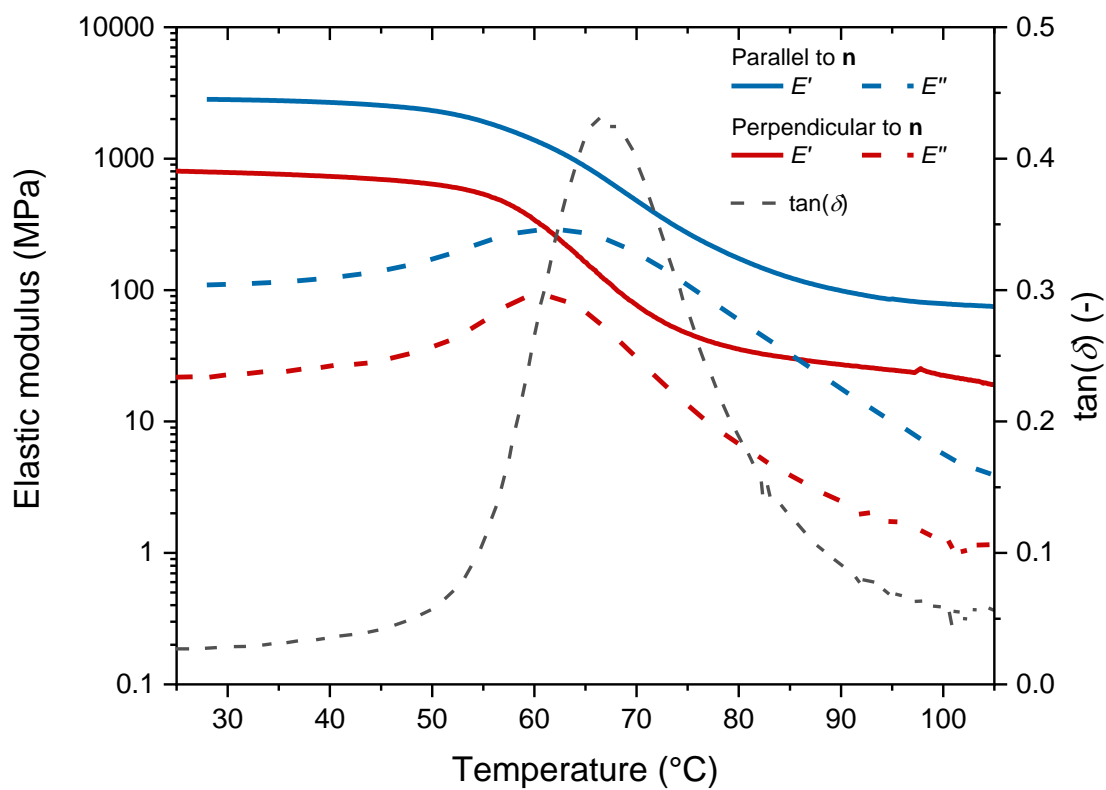3   S. Timoshenko, *J. Opt. Soc. Am.*, 1925, **11**, 233.

Experimental results
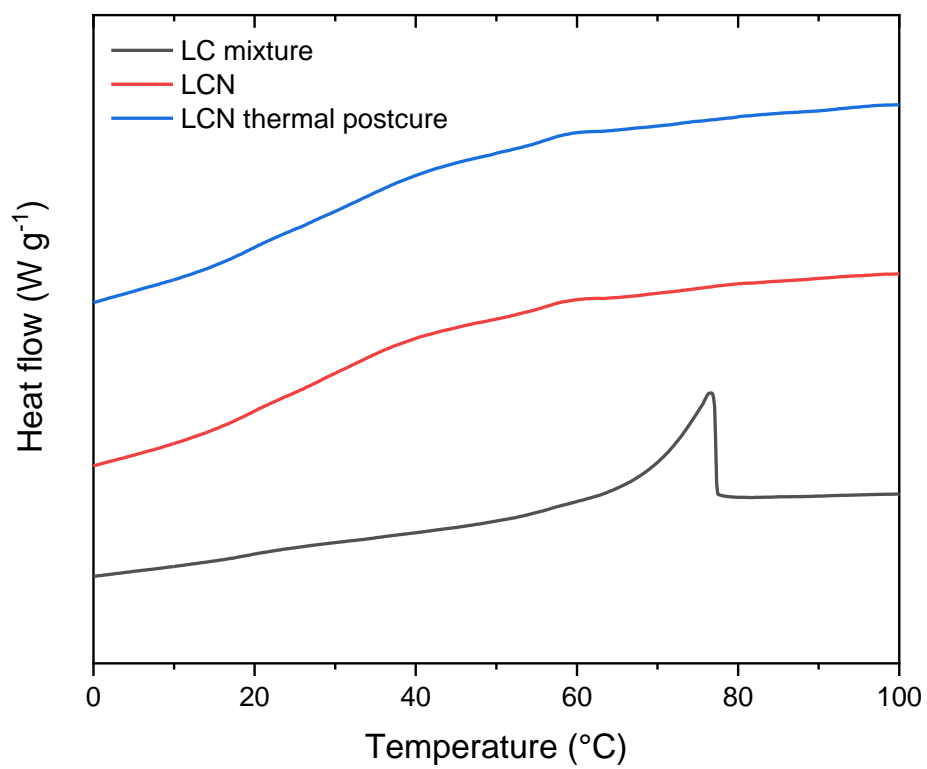
Figure S2. Differential scanning calorimetry of the LC mixture's phase behaviour.
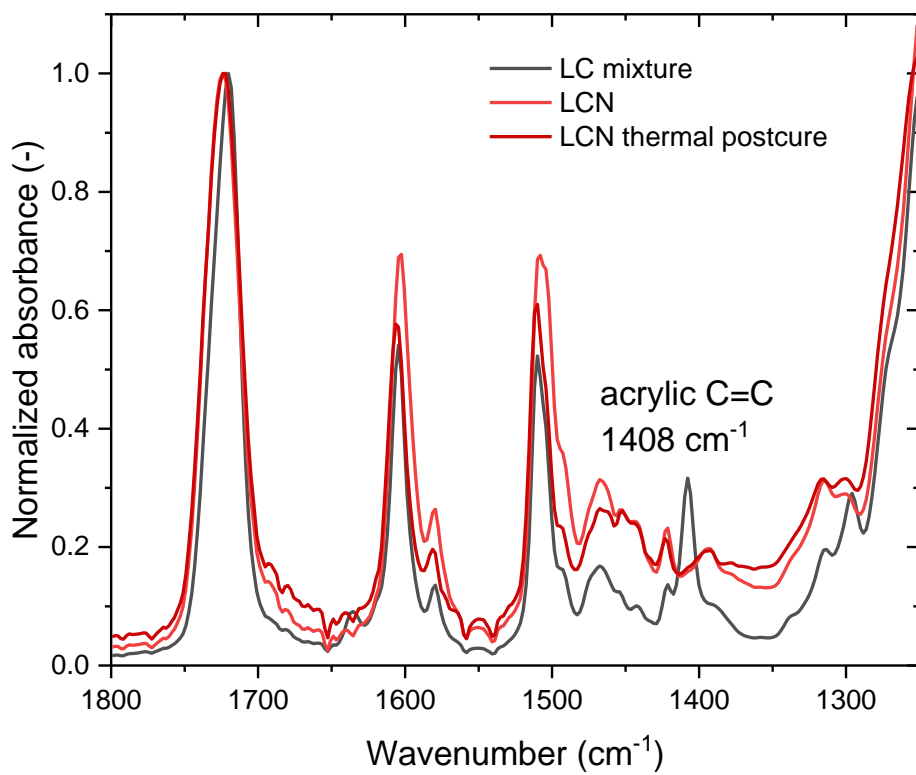
Figure S3. FT-IR spectra of the unpolymerized mixture (black), polymerized film (red) and thermally annealed film (dark red).

Figure S4. Dynamic mechanical analysis (DMA) measurement performed on films of 20 μm thickness with planar alignment. Plotted as function of temperature: E' and E'' for parallel (E_{11}, blue) and perpendicular ($E_{22,33}$, red) to **n**, as well as dissipation factor tan(δ).
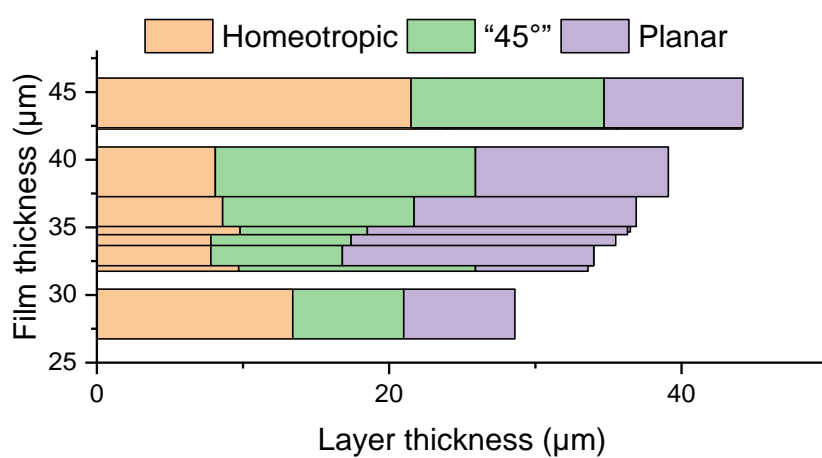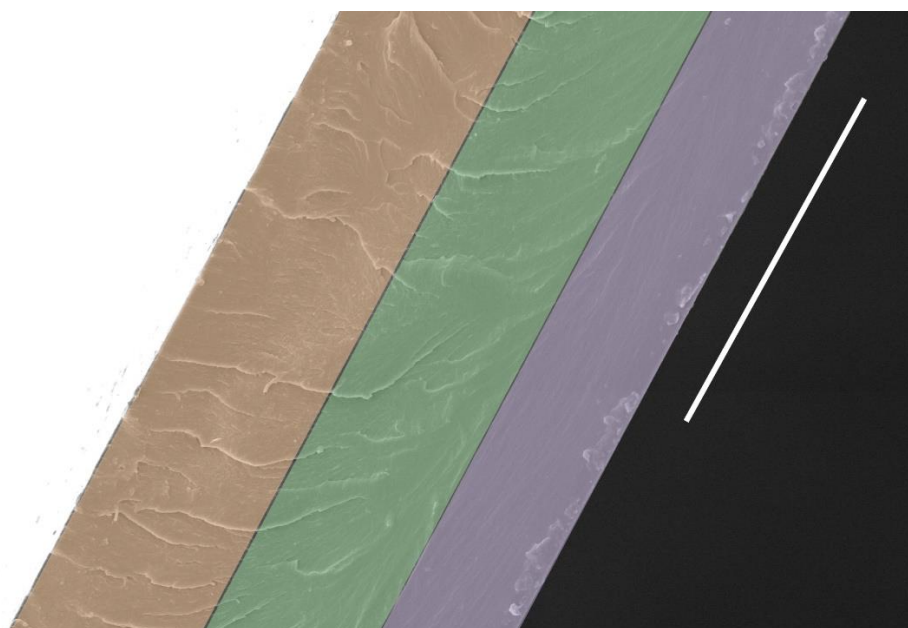
Figure S5. Scanning electron micrograph marking the different regions in the splay LCN film, as well as their penetration depths as function of the total film thickness.
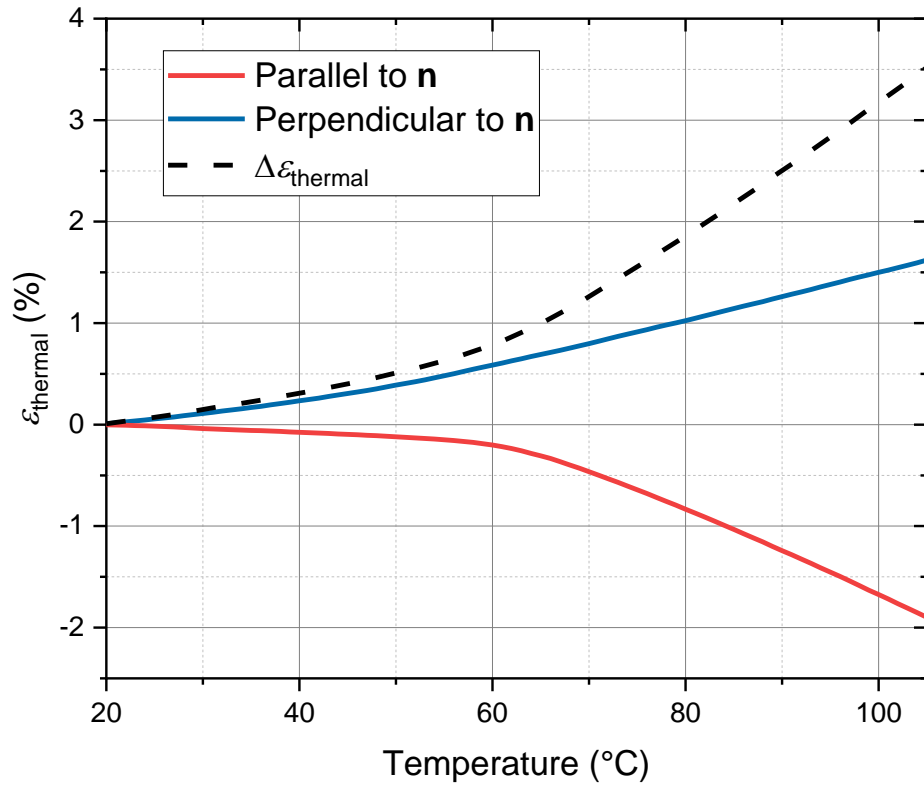
Figure S6. Thermal strain for directions **n** parallel to the strain analyser direction (ε_{11}), **n** perpendicular to the strain analyser ($\varepsilon_{22,33}$), and the difference ($\Delta\varepsilon$) as function of the temperature. The values are calculated with reference point $T_0 = 19$ °C.
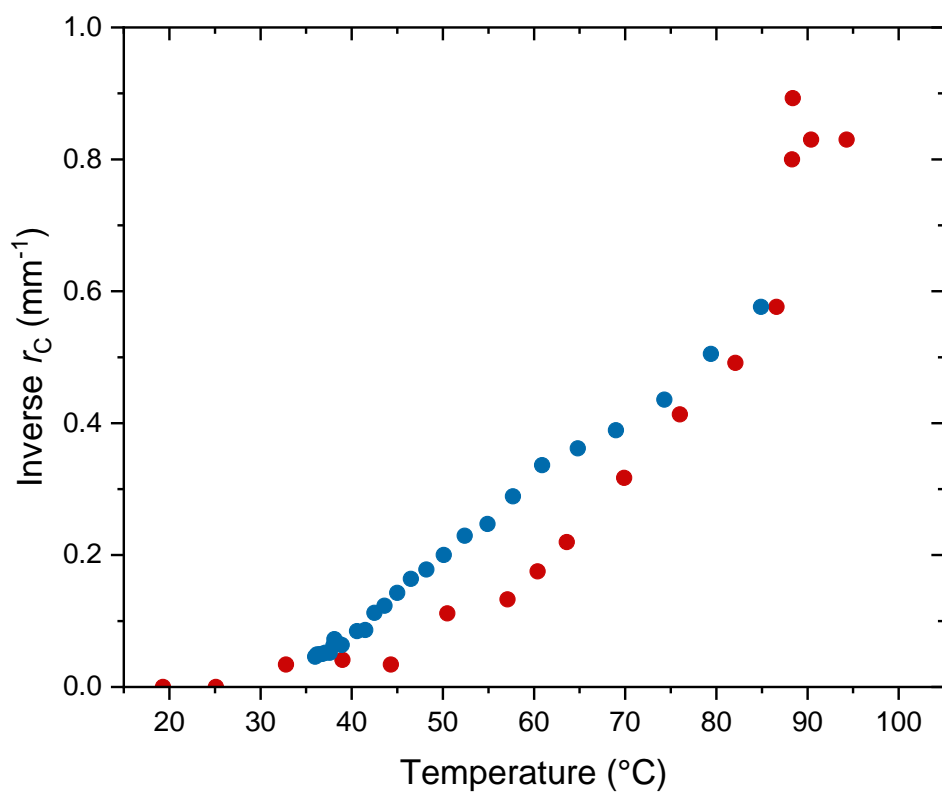
Figure S7. Extended image analysis data of a heating and cooling run of the tapered LCN polymer film. Red data points are measured during heating, while blue points are measured during cooling.
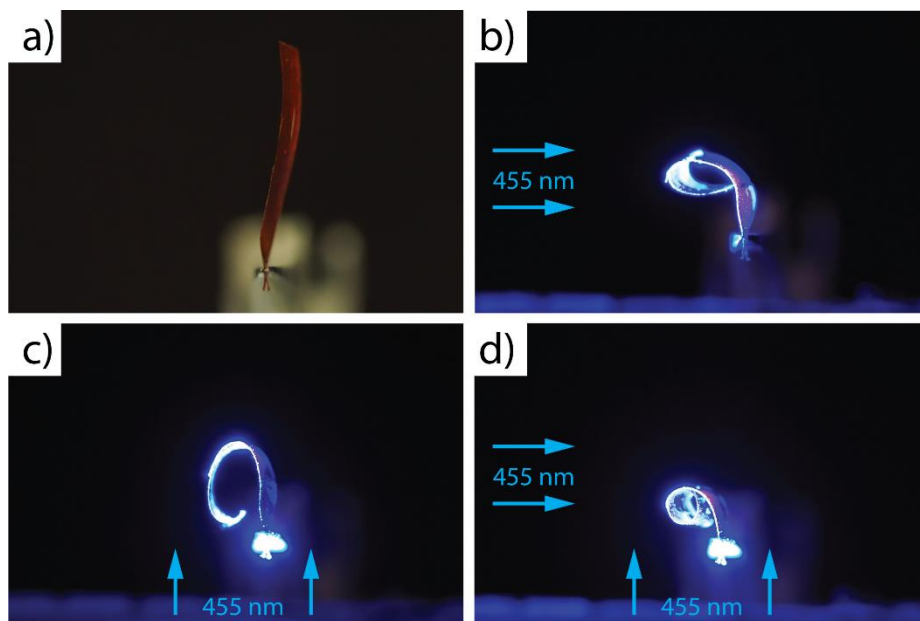
Figure S8. Extended data concerning photo-actuation of the tapered splay-aligned LCN film. a) No illumination, b) 455 nm illumination from one side (left) at ±310 mW cm⁻², c) 455 nm illumination from one side (underneath) at ±310 mW cm⁻², d) 455 nm illumination from both aforementioned sides at a combined ±620 mW cm⁻².