Electronic Supporting Information (ESI)

A self-sustained soft actuator able to rock and roll

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Experimental details:

The liquid crystal polymer network is produced by the photo-copolymerization of two liquid crystalline monomers, a monoacrylate, (RM 23; 40,5mol%, Merck) and a diacrylate, (RM82; 56,5mol%, Merck), and initiated by a photoinitiator (Irgacure 819, 1mol%, Ciba). Light responsivity was achieved with the addition of a commercially available azo-benzene chromophore with a fast *cis-trans* isomerization, DR1A, (2 mol%, Sigma Aldrich). Prior to polymerization, the monomers were dissolved in DCM to obtain a homogeneous mixture, subsequently the solvent was evaporated.

Polymerization was carried out in a custom-made glass cell. Prior to cell fabrication the independent glass slides are thoroughly cleaned and subjected to a UV/Ozone treatment (UV Products PR-100) for the duration of 20 minutes to activate the glass surface for further coating. To provide surface alignment for the LC mesogens, the glass slides are coated with polyimide (PI) alignment layers; for a splay alignment one slide contained planar and the other homeotropic alignment layers, Optimer AL 1051 (JSR Micro) and 5661 polyimide (Sunever), respectively. In planar alignment molecules are aligned parallel to the surface and in homeotropic alignment the molecules are aligned perpendicular to the surface. For uniaxial planar alignment, the glass coated with planar alignment are gently rubbed with velvet cloth. The glass cells were prepared by gluing together two glass slides (one with homeotropic coating, the other with planar coating) with glue containing 20 μm diameter glass bead spacers, to achieve controlled cell thickness. The cells were filled with the LC mixture through capillary action. Cell filling was carried out at 95 °C, at which the LC mixture is isotropic. Subsequently, the filled cell was cooled to 80 °C, at which temperature the LC mixture is nematic, see **Figure S1**. Photo-polymerization of the reactive mesogens was done at this temperature with an Exfo Omnicure S2000 lamp (3-5 $mWcm^{-2}$) for 15 minutes; subsequent thermal treatment at 120 °C for 10 minutes released thermal stresses arising from polymer shrinkage during polymerization. After polymerization, the cell was opened, and the films are peeled from the glass with razor blades and cut into long strips, in which the alignment direction of the planar side is parallel to the long side of the film. See **Figure S6** for a simplified schematic of the fabrication process.

Description of the rolling motion:

To demonstrate the rolling motion, during the rocking motion, a triangular LCN is exposed to a collimated (455 nm) light at an oblique angle. This illumination induces the film to begin a directed rolling motion, Figure 4 in manuscript. The directed rolling motion results in the displacement of the film over the heated surface. Illumination directed from the side and top (Figure 4A ii and iv) shows rolling motility of the system. Once the rolling motion is initiated by side illumination (Figure 4A ii), the system comes to a rested state with its planar side facing the hot plate, Figure 4A iii. The film is then brought to an out-of-equilibrium position (Figure 4A iv and v) through top illumination, causing subsequent rolling of the film.

The sequence of illumination steps is repeated to realize rolling motility, **Movie S6**. The light source used for these experiments is a light emitting diode (LED) source (Thorlabs M455L3-C2), focused using a collimator (SM2F32-A) set to around 300 mW/cm^2 through the controller by Thorlabs DC4104.

Supporting Figures:

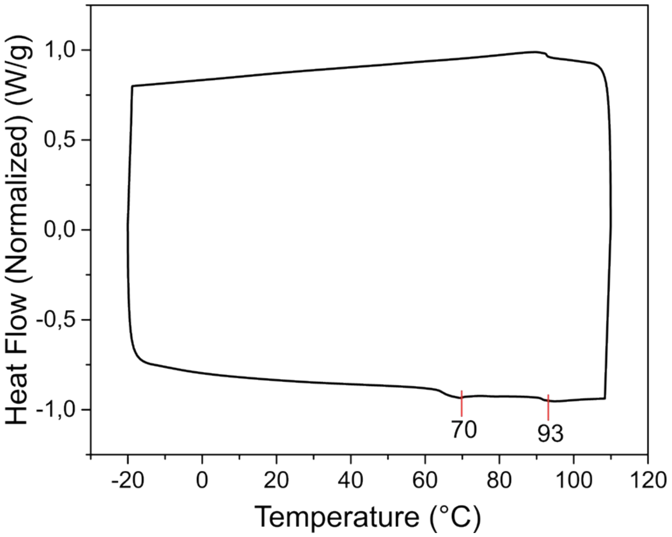

Figure S1. Differential scanning calorimetry scan (second run) of the LC mixture used in this study. We observe a nematic phase between 70 and 93 °C; above 93 °C the mixture is isotropic.

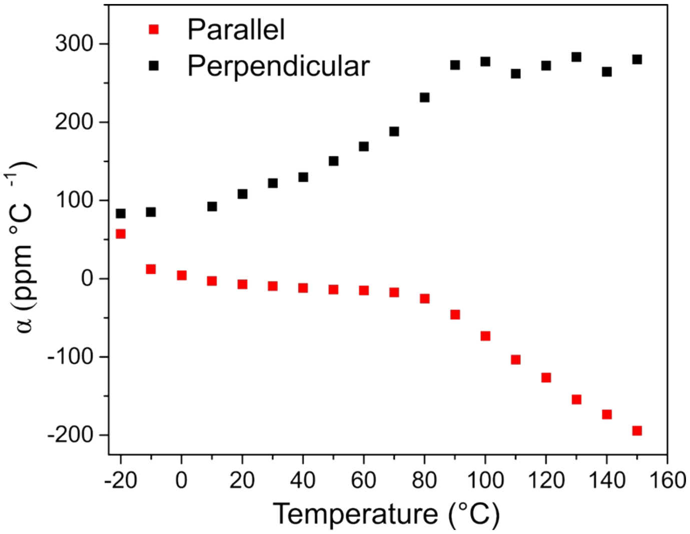

Figure S2. Coefficients of thermal expansion, perpendicular (black squares) and parallel (red squares) to the molecular orientation with reference temperature of -20°C.

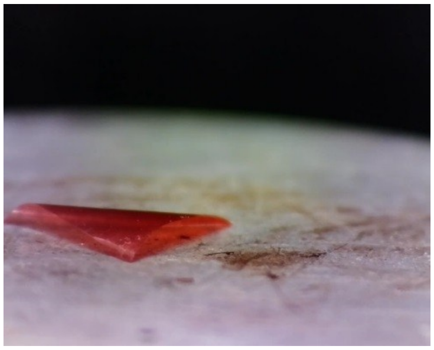

Figure S3. Uncurled shape of triangular film when placed with its planar side onto the heated surface. This position is stable and does not initiate further motion.

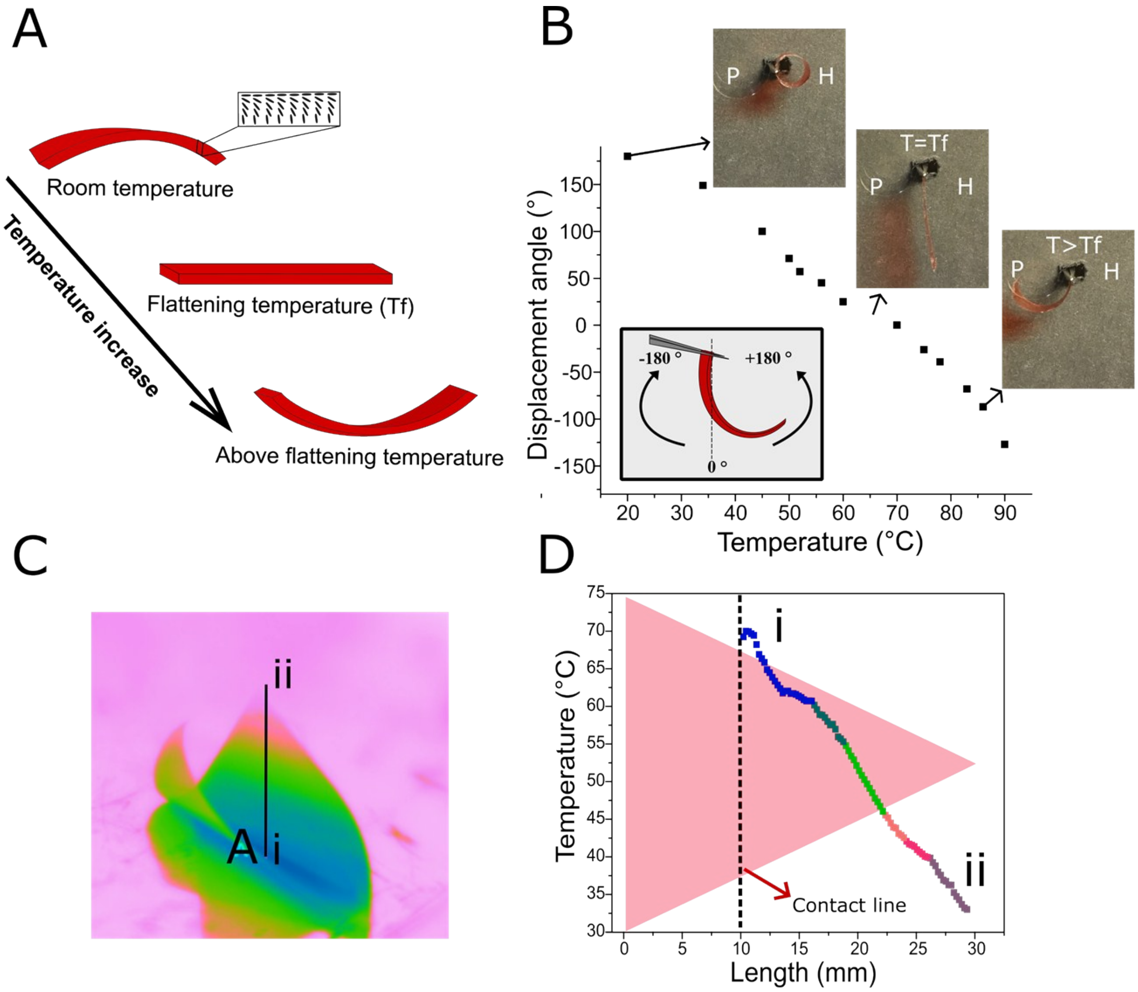

Figure S4. A. Schematic drawing of the temperature triggered deformation of a splay aligned LCN strip with three main deformation shapes: below, at, and above Tf. **B.** Deformation plot for the LC film (1.6 cm in length) in response to temperature variation in an oven. 'P' and 'H' refer to the planar and homeotropic sides of the splay film, respectively. **C.** Thermal image of the triangular film in contact with a heated surface above Tf, showing a steep thermal gradient along the film's length. **D.** Plot showing the thermal gradient along the length of the triangular film (Line A in Figure S4C, the colors of the data corresponding to the colors in the thermal photograph).

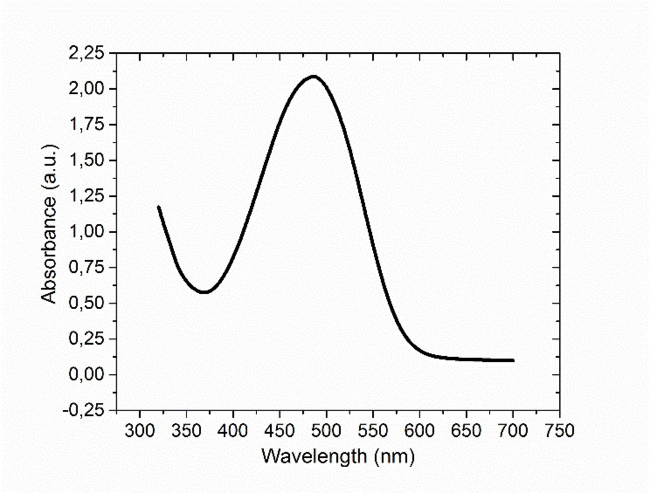

Figure S5. Absorbance spectrum for splay aligned film containing azobenzene chromophore.

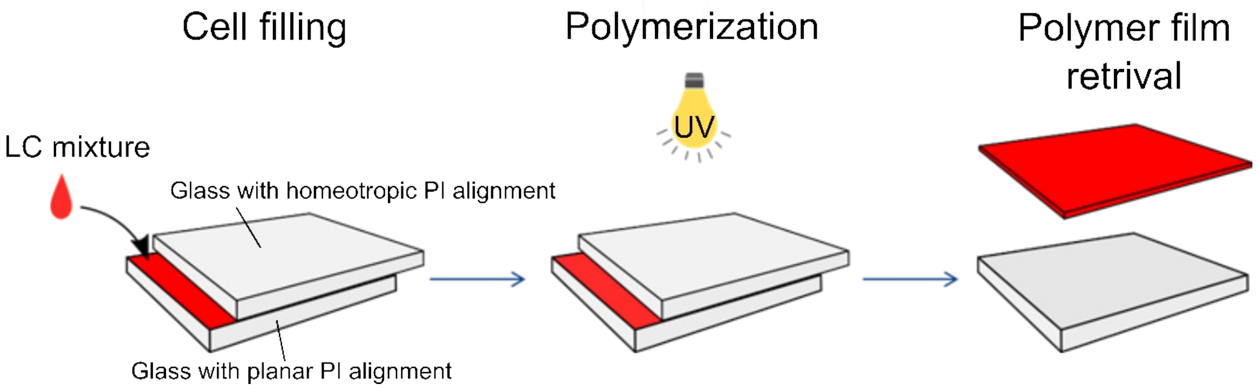

Figure S6. Schematic figure depicting a simplified demonstration of the polymer film fabrication procedure.

Finite element model to simulate the rocking behavior in triangular films:

A dynamic coupled thermo-mechanical model to simulate the rocking behavior observed in LC thin films is developed in a commercial finite element software ABAQUS. The equations modeling the mechanical behavior of the material under a temperature stimulus are the following.

The thermal strain developed in the film due to change in temperature is formulated as,

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

where, ε_{ij}^{t} is the thermal strain tensor, α_{ij} is a tensor representing the coefficient of thermal expansion and ΔT represents the change in temperature. The total strain (ε_{ij}) developed in the film will comprise of the thermal strain and the elastic strain (ε_{ij}^{e}),

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

The material is assumed to be linearly elastic with the stress-strain relationship,

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

where, C_{ijkl} is the elastic stiffness matrix. The orientation dependence of the elastic modulus is incorporated by modeling the film as a transversely isotropic material with elastic moduli estimated as a function of temperature along perpendicular direction. [15] The Poisson's ratio is taken as $\upsilon_{12} = \upsilon_{23} = \upsilon_{13} = 0.3$. The coefficient of thermal expansion ($\alpha_{\parallel}$ and $\alpha_{\perp}$) is measured as a function of temperature, Figure S1. The same is used in the model to account for the anisotropic thermal response of the film.

The thin film is modelled as a tri-layer composite with 50% homeotropic, 25% tilted at 45° orientation and 25% planar as shown in **Figure S7**.[15] The triangular film has an edge length of 35 mm and a thickness of 20 µm.

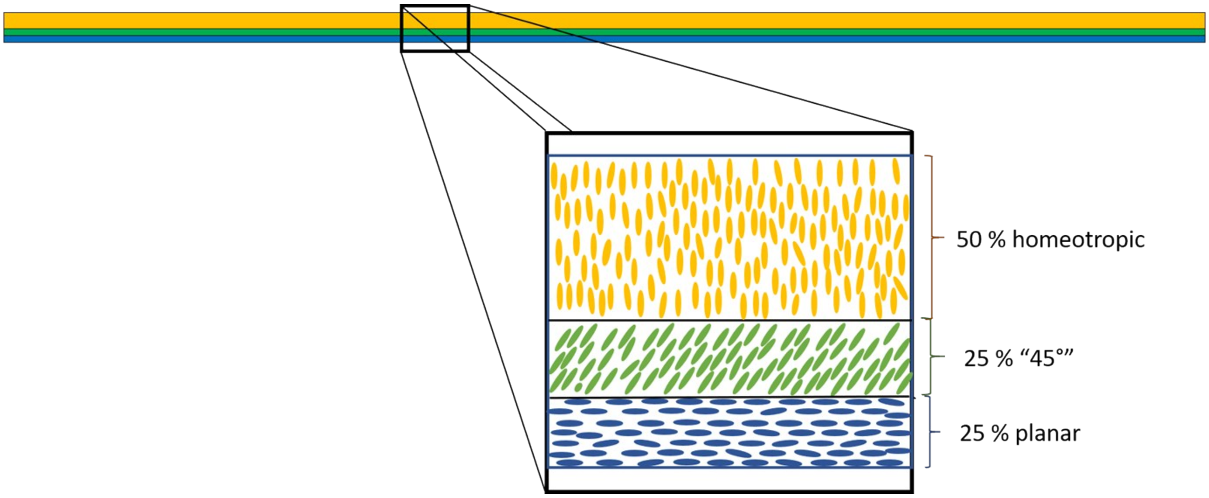

Figure S7. Schematic showing the alignment distribution through the thickness of the film used in the finite element model.

Apart from the film, a fixed discrete rigid body is created to act as a ground for the film, depicting the hot plate in the experiments. A frictional surface contact is imposed between the film and the rigid surface. A downward body force is applied on the film to simulate the gravity effect and a mass proportional damping factor is included to bound the kinetic energy of the film (the internal energy is ensured to be much higher than the dissipated energy through damping) and also to provide some numerical damping to the system.

The thin film is discretized using approximately 5000 composite shell elements (S3 and S4R). A linearly decreasing initial temperature profile based on the distance of each node from the contact line is specified. The temperature of each element is raised to the rocking temperature profile from the initial temperature. Hereafter, the rocking temperature distribution in the film is entirely dependent on the position of the contact line and as the film rocks, the temperature distribution in the film is changed accordingly. The initial and rocking temperature distributions can be seen in Figure S4C. The FE simulations are carried out using Abaqus Standard Implicit Dynamic solution process.