

Electronic Supplementary Information for *Soft Matter* manuscript:

Shear-induced polydomain structures in lyotropic chromonic liquid crystal disodium cromoglycate

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Optical analysis of the domain textures

A polarized beam with a given polarization state acquires a new polarization state when it propagates through one or more polarizing elements. The outgoing beam defined by the Stokes-vector \mathbf{S} can be related to the incident beam \mathbf{S}_0 through a 4×4 transformation matrix \mathbf{M} known as the Mueller matrix¹.

$$\mathbf{S} = \mathbf{M} \cdot \mathbf{S}_0 . \quad (1.1)$$

The amplitude and the phase of the incident beam can be changed using optical elements such as polarizer and wave plate, respectively. The Mueller matrix for a linear polarizer with the transmission axis that makes an angle χ with the vorticity y -axis in Fig. 2a

$$\mathbf{M}_{lp}(\chi) = \frac{1}{2} \begin{pmatrix} 1 & \cos 2\chi & \sin 2\chi & 0 \\ \cos 2\chi & \cos^2 2\chi & \cos 2\chi \sin 2\chi & 0 \\ \sin 2\chi & \cos 2\chi \sin 2\chi & \sin^2 2\chi & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} . \quad (1.2)$$

The Mueller matrix for a non-absorbing slab of a thickness h and birefringence Δn , with its slow axis making an angle χ with the vorticity direction is

$$\mathbf{M}_{wp}(\chi) = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos^2 2\chi + \sin^2 2\chi \cos \delta & \cos 2\chi \sin 2\chi (1 - \cos \delta) & \sin 2\chi \sin \delta \\ 0 & \cos 2\chi \sin 2\chi (1 - \cos \delta) & \cos^2 2\chi \cos \delta + \sin^2 2\chi & -\cos 2\chi \sin \delta \\ 0 & -\sin 2\chi \sin \delta & \cos 2\chi \sin \delta & \cos \delta \end{pmatrix} , \quad (1.3)$$

where $\delta = 2\pi h \Delta n / \lambda$ is the phase shift and λ is the wavelength of light in vacuum.

In the first optical setup used to identify domains with the director tilted away from the vorticity axis (also called a horizontal direction) in Region II, the beam propagates through the following elements:

- A linear polarizer with the transmission axis along the vorticity directions, Eq. (1.2) with $\chi = 0$;
- A slab of thickness 10 μm of DSCG 14wt% with $\Delta n = -0.016$, with the Mueller matrix defined by Eq. (1.3);

- c) A full-wave plate (FWP) of wavelength $\lambda_g = 530$ nm with its fast-axis at 45° with the horizontal direction, with the Mueller matrix deduced from Eq. (1.3) as

$$\mathbf{M}_{\lambda_g}(\pi/4) = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos(2\pi\lambda_g/\lambda) & 0 & \sin(2\pi\lambda_g/\lambda) \\ 0 & 0 & 1 & 0 \\ 0 & -\sin(2\pi\lambda_g/\lambda) & 0 & \cos(2\pi\lambda_g/\lambda) \end{pmatrix};$$

- d) A linear polarizer with the transmission direction along the flow, Eq. (1.2) with $\chi = \pi/2$.

We consider an incident beam of a unit intensity,

$$\mathbf{S}_0 = \begin{pmatrix} 1 \\ 1 \\ 0 \\ 0 \end{pmatrix}.$$

The Stokes vector of the beam transmitted through the system can be calculated as

$$\mathbf{S} = \mathbf{M}_{LP}(\pi/2) \cdot \mathbf{M}_{\lambda}(\pi/4) \cdot \mathbf{M}_{LC}(\chi) \cdot \mathbf{M}_{LP}(0) \cdot \mathbf{S}_0, \quad (1.4)$$

which yields

$$\mathbf{S} = \frac{1}{2} \begin{pmatrix} 1 - \cos \frac{2\pi\lambda_g}{\lambda} \left(\cos^2 2\chi + \cos \frac{2\pi h \Delta n}{\lambda} \sin^2 2\chi \right) + \sin 2\chi \sin \frac{2\pi\lambda_g}{\lambda} \sin \frac{2\pi h \Delta n}{\lambda} \\ -1 + \cos \frac{2\pi\lambda_g}{\lambda} \left(\cos^2 2\chi + \cos \frac{2\pi h \Delta n}{\lambda} \sin^2 2\chi \right) + \sin 2\chi \sin \frac{2\pi\lambda_g}{\lambda} \sin \frac{2\pi h \Delta n}{\lambda} \\ 0 \\ 0 \end{pmatrix}.$$

The intensity of the outgoing beam is defined by the first component of the Stokes vector

$$I(\chi, \lambda) = \frac{1}{2} \left[1 - \cos \frac{2\pi\lambda_g}{\lambda} \left(\cos^2 2\chi + \cos \frac{2\pi h \Delta n}{\lambda} \sin^2 2\chi \right) + \sin 2\chi \sin \frac{2\pi\lambda_g}{\lambda} \sin \frac{2\pi h \Delta n}{\lambda} \right]. \quad (1.5)$$

Eq. (1.5) is used to calculate the intensities of light transmitted through the FWP with $\lambda_g = 530$ nm, and the LC cell with $h = 10\mu\text{m}$ and $\Delta n = -0.016$, at different wavelengths, $\lambda = (470, 580, \text{ and } 530)$ nm, corresponding to blue, yellow, and green colors, respectively, as a function of the director orientation \hat{n} specified by the angle χ , Fig.S1a.

To compare the theoretical calculations to the experimental data, we measured the transmitted light intensity through the same optical system, in which a uniformly aligned planar cell of 14wt% DSCG, $h = 10\mu\text{m}$ and $\Delta n = -0.016$, was rotated, thus continuously changing the angle χ , Fig.S1b. The intensity is measured using "Plot Profile" function in Fiji/ImageJ², and the gray values of all the pixels in the selection divided by the number of pixels. The minimum intensity is observed for $\chi = 0$, in the region that appears magenta in color in observations with FWP and correspond to the director along the vorticity direction. The transmitted light intensity at the blue and yellow wavelengths reaches maxima at -45° and 45° , respectively. The theory predicts a stronger blue maximum than the yellow one, Fig.S1a, while the experiment shows the opposite relationship. The reason is that the tungsten-halogen lamp used in the microscope Olympus BX40 produces a much stronger light intensity in the yellow region as compared to the blue region.

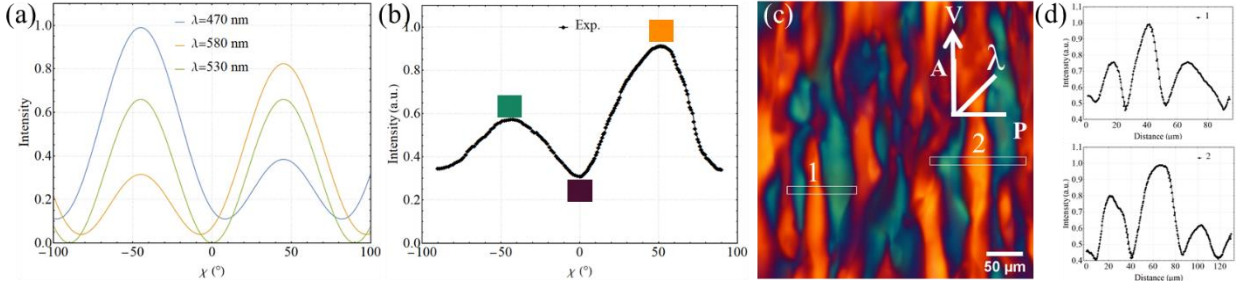


Fig.S1. The method used to define the size of domains a_x and a_y in which the director deviates from the vorticity axis in Region II textures. (a) The intensity of light transmitted through the LC cell, FWP and two crossed linear polarizers, at different wavelengths $\lambda=470, 580,$ and 530 nm calculated using Eq.(1.5); (b) the intensity of light transmitted through the same optical elements, in which the LC cell is a uniformly aligned planar cell that rotates around the axis that coincides with the propagating beam; (c) POM texture of $10 \mu\text{m}$ slab of the nematic DSCG sheared at $\dot{\gamma} \approx 3 \text{ s}^{-1}$ recorded with the optical setup shown on the photograph, (d) Intensity profile for the two horizontal regions marked 1 and 2 in panel (c).

The analysis above shows the blue and yellow patches of the POM textures correspond to the director tilts away from the vorticity axis. The domains boundaries, a_x and a_y can be determined by finding the regions where the light intensities corresponding to the blue and yellow colors are at their minima. Using the “Brightness/Contrast” tool in Fiji/ImageJ², we adjusted the “minimum” and the “maximum” to be equal to the smallest and the largest pixel values in the image’s histogram; almost all of the analyzed images lies in the range of $(0.35 - 1)$, where 0.35 is the minimum pixel value and 1 is the largest pixel value. Then using the “Color Threshold” and by choosing the RGB color space and setting the value of the Red channel to 255, and Green and Blue channels are adjusted manually to select the edges of the domains corresponding to the minimum intensities. The minimum intensities at the edges of each of the yellow and the blue domains in Fig. S1b are used to select these domains from the background to find a_x and a_y as shown in Fig. S2.



Fig. S2. Identification of domain structure and determination of a_x and a_y in Region II. (a) POM texture with FWP and crossed polarizers; (b) blue and yellow domains selected using Fiji2/ImageJ color threshold², (c) binary image of part (b), (d) outlines of the domains, and ellipses fitting the domains in panel (d), obtained by using Analyze Particles in Fiji2/imageJ². Projections of the two axes of the ellipses onto the directions of flow and vorticity define a_x and a_y , respectively.

The second optical setup is used to find the regions in which the director is along the vorticity direction. In this setting, the beam passes through

- A linear polarizer, Eq. (1.2) with $\chi = \pi/4$;
- A DSCG slab of thickness $10 \mu\text{m}$ with $\Delta n = -0.016$, with the Mueller matrix defined by Eq. (1.3);
- A full-wave plate (FWP) of wavelength $\lambda_g = 530 \text{ nm}$ with its fast-axis along the flow direction, with the Mueller matrix deduced from Eq. (1.3) as

$$\mathbf{M}_{\lambda_g}(\pi/2) = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & \cos(2\pi\lambda_g/\lambda) & \sin(2\pi\lambda_g/\lambda) \\ 0 & 0 & -\sin(2\pi\lambda_g/\lambda) & \cos(2\pi\lambda_g/\lambda) \end{pmatrix};$$

- A linear polarizer, Eq. (1.2) with $\chi = -\pi/4$.

The Stokes-vector for the outgoing beam can be calculated as

$$\mathbf{S} = \mathbf{M}_{LP}(-\pi/4) \cdot \mathbf{M}_{\lambda_g}(\pi/2) \cdot \mathbf{M}_{LC}(\chi) \cdot \mathbf{M}_{LP}(\pi/4) \cdot \mathbf{S}_0. \quad (1.6)$$

Using Mathematica to simply the above expression we obtained

$$\mathbf{S} = \frac{1}{2} \begin{pmatrix} 1 - \cos \frac{2\pi\lambda_g}{\lambda} \left(\cos^2 2\chi \cos \frac{2\pi h \Delta n}{\lambda} + \sin^2 2\chi \right) - \cos 2\chi \sin \frac{2\pi\lambda_g}{\lambda} \sin \frac{2\pi h \Delta n}{\lambda} \\ 0 \\ -1 + \cos \frac{2\pi\lambda_g}{\lambda} \left(\cos^2 2\chi \cos \frac{2\pi h \Delta n}{\lambda} + \sin^2 2\chi \right) + \sin 2\chi \cos 2\chi \sin \frac{2\pi\lambda_g}{\lambda} \sin \frac{2\pi h \Delta n}{\lambda} \\ 0 \end{pmatrix}.$$

Thus, the intensity of the outgoing beam reads,

$$I(\chi, \lambda) = \frac{1}{2} \left[1 - \cos \frac{2\pi\lambda_g}{\lambda} \left(\cos^2 2\chi \cos \frac{2\pi h \Delta n}{\lambda} + \sin^2 2\chi \right) - \cos 2\chi \sin \frac{2\pi\lambda_g}{\lambda} \sin \frac{2\pi h \Delta n}{\lambda} \right]. \quad (1.7)$$

The calculated transmitted intensities at $\lambda = (470, 580, \text{ and } 530) \text{ nm}$ as a function of LC orientation χ show that the transmitted blue light intensity is maximum when $\hat{\mathbf{n}}$ is parallel to the vorticity direction, $\chi = 0$, Fig. S3a. The blue color intensity decays to its minimum at $\chi = \pm 50^\circ \pm 5^\circ$. In this optical setting, the LC texture appears yellow when $\hat{\mathbf{n}}$ is parallel to the flow direction, $\chi = 90^\circ$, Fig. S3a. The minimum intensities at the edges of blue domains in Fig.S3b are used to select the domains with the director aligned predominantly along the vorticity axis and to measure α_w as shown in Fig.S4. Since the blue color of the texture fades only at $\chi = \pm 50^\circ \pm 5^\circ$, the measured value of α_w corresponds to the entire range of angles that the director makes with the vorticity axis, $-50^\circ \pm 5^\circ \leq \chi \leq 50^\circ \pm 5^\circ$.

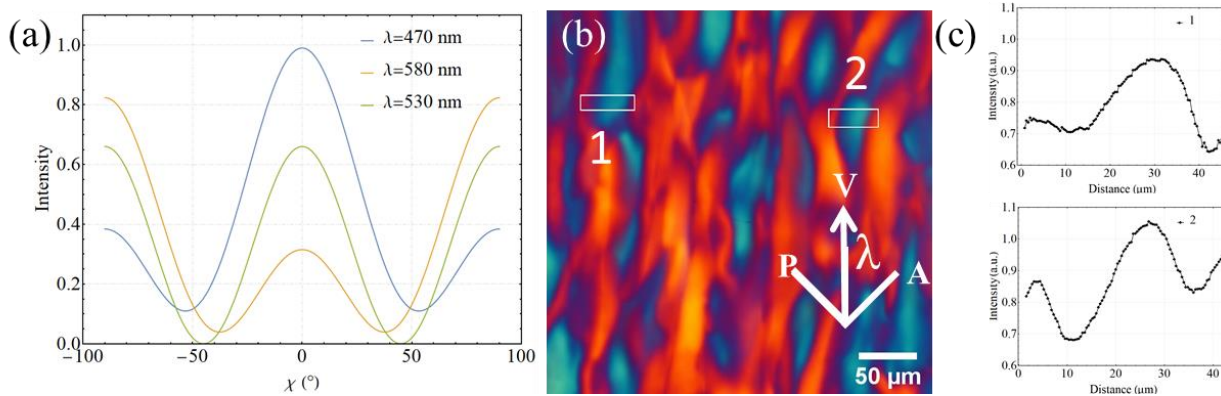


Fig.S3 The method used to define the width a_w of the domain walls in which the director is predominantly along the vorticity axis. (a) The intensity of light transmitted through the LC cell, FWP and two crossed linear polarizers, at different wavelengths $\lambda= 470, 580,$ and 530 nm calculated using Eq.(1.7); (b) a typical POM texture with the superimposed geometry of the optical elements; (c) Intensity profiles for the along the horizontal regions marked 1 and 2 in panel (b).

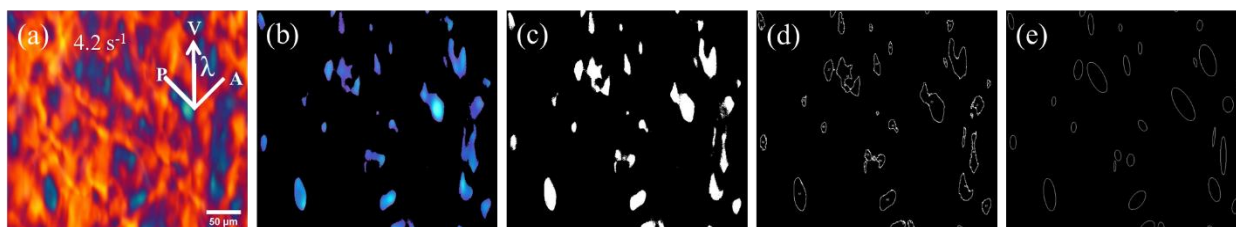


Fig.S4 The method used to determine a_w . (a) A POM image with polarizers and FWP; (b) blue domains selected using color Threshold, (c) binary image of part (b), (d) outlines of the domains, and (e) ellipses fitting the domains in panel (d) obtained using Analyze Particles in Fiji2/imageJ².

WAXS analysis

The wide-angle x-ray scattering (WAXS) peaks arise from the characteristic separation between the DSCG molecules within the aggregates. The staking distance w of the DSCG molecules within aggregates is measured as $w=2\pi/Q$, where Q is the wavevector corresponding to the maximum intensity³, Fig.S5c. The stacking distance $w = 0.34$ nm does not change as the shear rate changes. The correlation length $\xi_{||}$ along the aggregates, calculated from the full width at half maximum FWHM of the WAXS peaks³, is also independent on the shear rate in the range explored, Fig. S5d.

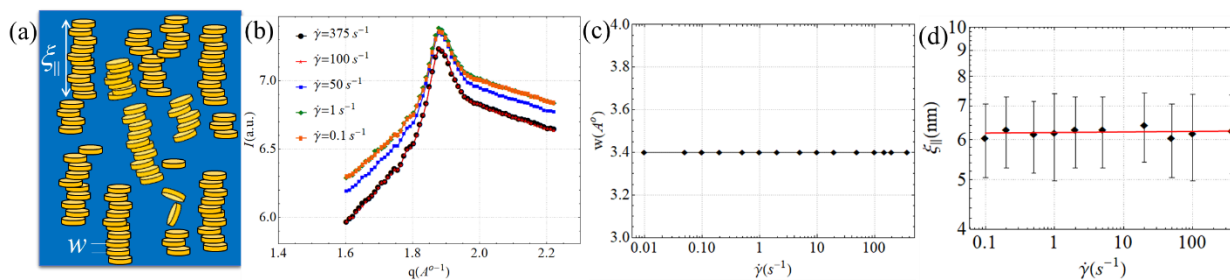


Fig. S5 (a) Molecular structure of DSCG aggregates; (b) WAXS peaks at different shear rates; (c) stacking distance as a function of shear rate; (d) correlation length parallel to the aggregates long axis $\xi_{||}$ as a function of shear rate.

References

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