Flow of a nematogen past a cylindrical micro-pillar†

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We study the flow of a nematic liquid crystal past a micron-sized cylindrical pillar within a microfluidic confinement of a rectangular cross-section. The liquid crystal molecules are anchored perpendicularly (homeotropic anchoring) to the surface of the pillar and the channel walls. Flow past the cylindrical obstacle generated topological defect structures whose nature, dimensions and morphology varied with the flow velocity and channel dimensions. On increasing the flow speed, we observed sequential evolution of a semi-integer loop, which transformed into an integer hedgehog defect, and finally equilibrated to an extended defect wall. On stopping the flow, the topological defect states reversed its sequence of appearance. Additionally, we introduce dual-focus fluorescence correlation spectroscopy as a general velocimetry technique for microfluidics of liquid crystal systems – with or without topological defect structures.

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1 Introduction

The advent of microfluidics¹ has opened up possibilities to study flows within micron-sized confinements and past minute obstacles. In contrast to the flows at macro scales, flow within microchannels is fundamentally different due to the drastic reduction of the inertial effects, resulting in very low Reynolds numbers.² In general, the flow of Newtonian fluids in a micro-channel is laminar. However, studies have shown that such a hydrodynamic regime may not be strict when a non-Newtonian fluid *e.g.* polymer solution³ is flowed, or in the presence of appropriate morphological patterning of the microchannel.⁴ Recent investigations by the authors have further revealed that the use of an anisotropic fluid (liquid crystal) as the flowing matrix can induce effects distinct from those observed for an isotropic fluid.^{5,6} The average orientation of the liquid crystal molecules, denoted by a vector $\vec{n}$ and referred to as the director,⁷ plays a decisive role in determining the rheological properties of the anisotropic fluid.⁸ Additionally, the boundary conditions existing on the surfaces that confine the liquid crystal have a crucial contribution in determining the static equilibrium of the director.⁹ The dynamics of liquid crystal flows, which are

inherently complex due to the coupling between flow and the orientational order,^{10,11} lead to a variety of interesting phenomena like generation of a transverse pressure gradient in Poiseuille flow,¹² anomalous colloidal rheology,¹³ and the backflow mechanism.¹⁴ A recent work by Lavrentovich *et al.*¹⁵ has elucidated the director distortion, and the consequent nematic flow within a rectangular micro-capillary due to the thermal expansion of a nematogen. Interestingly, the defect dynamics, for instance, in a homeotropic round capillary, induced by the thermo-mechanical stresses,¹⁵ show certain qualitative similarities with those discussed in the present article.

In this present work we have experimentally studied the flow of a room-temperature nematogen (liquid crystal in the nematic phase) past a cylindrical micro-pillar cast in a microchannel. The coupling between flow and nematic ordering induces director reorientation in the vicinity of the obstacle which is distinct from that farther away from the micro-pillar. Consequently, a sequence of flow-induced topological defects¹⁶ evolve at the pillar. The nature, dimension, and director morphology of the generated topological defects varied with the flow speed and the channel dimensions. In contrast to our observations, numerical studies previously carried out^{17,18} do not report the evolution of topological defects from the obstacles. However, the effect of flow on topological defects around colloids has been reported by Araki and Tanaka.¹⁹ More recently, flow of a nematic liquid crystal (NLC) through a homeotropic porous network has shown reconfigurable deformation of defects around the solid portions of the network.²⁰ The Stokes drag of spherical particles with accompanying topological defects at low Ericksen numbers, Er ,²¹ was investigated by Stark and Ventzki.²² Subsequently, the time evolution of flow-induced

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† Electronic supplementary information (ESI) available: ESI-1–7. (i) Defect loop instability within a 40 μm deep channel. (ii) Wall defect structure jetting out of the micro-pillar. (iii) Steady state condition of the wall defect. (iv) Defect wall dynamics and formation of the umbilic defect. (v) Formation of a radial-hyperbolic integer defect pair on termination of the flow. (vi) Defect wall dynamics on reversal of the flow direction. (vii) Bloch wall to Néel wall transformation. See DOI: 10.1039/c2sm27337c

orientational profiles of defects around colloids,²³ and around drops and bubbles rising through a nematic bulk,^{24,25} was investigated. Numerically or otherwise, flow of a nematogen past a cylindrical obstacle and the effect of high Er on the resulting defect morphology have not been reported yet.

In addition to studying the flow-induced topological defects and their transformation, we introduce dual-focus fluorescence correlation spectroscopy²⁶ as a technique for measuring the velocity field within flowing nematogens. Significantly, this technique is suitable for measuring velocities even in a co-existing system of high (nematogen) and low (topological defects) order phases. Conventional particle tracking methods are unsuitable due to the elastic interaction between the dispersed tracers and topological defects.

2 Experimental

2.1 Materials

We have used 4'-pentyl-4-biphenylcarbonitrile, commonly known as 5CB (Synthon Chemicals), as the flowing nematogen for all our experiments. The nematogen was used without any additional purification. 5CB is a single component liquid crystal (LC) which is in the nematic phase at room temperature. The nematic liquid crystal, 5CB, undergoes a transition to the isotropic phase at $\approx 33^\circ\text{C}$ and to the crystalline state at $\approx 18^\circ\text{C}$. Pure 5CB was doped with Nile Red dye (Sigma Aldrich) for fluorescence confocal polarization microscopy (FCPM). At appropriate concentrations, the dye molecules align along the long molecular axis of 5CB.²⁷ The dye concentration was maintained at $\approx 0.01\%$ to obtain an optimal fluorescence signal. The maximum emission of the Nile red corresponds to $\lambda_{\text{em}} = 640\text{ nm}$, when excited by light of wavelength $\lambda_{\text{exc}} = 553\text{ nm}$. Nanomolar concentration ($\approx 10\text{ nM}$) of ATTO Oxa12 (ATTO-TEC GmbH, Germany) in 5CB was used for the purpose of flow velocity (v) measurement. Approximately 1 mg of dye was dissolved in few μL of 5CB and the solution was diluted in three steps to achieve the desired concentration. The dyes were excited by a laser at $\lambda_{\text{exc}} = 640\text{ nm}$, and the maximum emission was recorded at $\lambda_{\text{em}} = 684\text{ nm}$. Prior to the experiments, we have treated the microchannels with an aqueous solution of octadecyldimethyl (3-trimethoxysilyl propyl) ammoniumchloride (60% in methanol, ABCR GmbH), also known as DMOAP (conc. 0.1% by weight) to induce homeotropic anchoring of the LC molecules.

2.2 Microfluidic setup

The microfluidic devices were fabricated using channel reliefs cast on polydimethylsiloxane (PDMS, Sylgard 184, Dow Corning) and surface bonded to glass following exposure to oxygen plasma. The cylindrical pillar was formed as a part of PDMS cast prepared using soft lithography.²⁸ A schematic representation of such a microfluidic chip is shown in Fig. 1a. In the present work we consider simple linear channels with a rectangular cross-section and a cylindrical pillar placed at the middle of the channel. The depth of the channel, d , was varied between $8\text{ }\mu\text{m}$ and $50\text{ }\mu\text{m}$, and pillar diameter $2r$ was chosen between $10\text{ }\mu\text{m}$ and $60\text{ }\mu\text{m}$ (Fig. 1b). All our experiments were conducted within w

Fig. 1 Microfluidic confinement. (a) Microchannel with a micron-sized cylindrical pillar fabricated using surface bonding of PDMS and glass components. The schematic shows a linear channel of a rectangular cross-section with the integrated micro pillar. x , y , and z coordinates indicate the directions of nematogen flow, channel width, and channel depth respectively. The blue arrow heads show the flow path. (b) Schematic representation of the plan view and the cross-sectional view (xz plane position shown in the plan view) of a microchannel possessing homeotropic surface anchoring. r , w , and d denote the pillar radius, channel width and depth, respectively.

$\approx 100\text{ }\mu\text{m}$ wide and sufficiently long (20 mm) channels. In Fig. 1a, x , y , and z denote the flow direction, channel width and channel depth, respectively. Cylindrical holes of $750\text{ }\mu\text{m}$ diameter at either ends of the channel served as the housing for the inlet and outlet tubes (Teflon). The tubes ($300\text{ }\mu\text{m}$ inner diameter and $760\text{ }\mu\text{m}$ outer diameter) were inserted within the holes punched in the PDMS mold. The smaller diameter of the PDMS housing as compared to the external diameter of the tube ensured good mechanical fitting. Additionally, uncured PDMS was applied at the point of fitting. After curing of the PDMS, any possible leakage at the inlet and outlet ports was ruled out. For filling the channel and subsequent flow experiments, a gear pump (neMESYS, Cetoni) with flow rate precision of nL h^{-1} ($\pm 0.02\%$ of the corresponding flow rates) was used to drive a gas-tight microliter syringe (1001LT, Hamilton Bonaduz), connected to the inlet tube. The outlet tubing was dipped in a sink of 5CB. In agreement with the observed dependence of the swelling ratio on the substance polarity ($\approx 4.8\text{ D}$ for 5CB),²⁹ no perceptible swelling of PDMS by 5CB was observed during the course of our experiments.

2.3 Surface anchoring and boundary conditions

Microfluidic devices fabricated using soft lithography are composed of two chemically distinct surfaces, namely PDMS and glass. Typically the LC molecules orient perpendicular to the PDMS surface, whereas on glass, the molecules anchor parallel to the surface with azimuthal degeneracy.³⁰ Homogeneous homeotropic surface anchoring was achieved by functionalization of the microchannel and pillar surfaces with an aqueous solution of DMOAP.³¹ The channel was first filled with the DMOAP solution and then rinsed with deionized water (after $\approx 10\text{ min}$). The surface treatment was thereafter thermally stabilized by heating the channel at 80°C for 15 min and at 110°C for 1 h . This yielded a homogeneous homeotropic anchoring on all the surfaces, shown schematically in Fig. 1b. For all our experiments, the

channels were filled with 5CB in the isotropic phase and allowed to cool down to the nematic phase.

2.4 Microscopy methods

The orientation of the LC molecules within the microchannel was analyzed using polarizing optical microscopy (POM) and fluorescence confocal polarization microscopy (FCPM). For POM experiments, the sample was placed between crossed-polarizers and observed in white light on a Nikon Eclipse LV100 polarizing microscope. Imaging was carried out with a $20\times$ air objective, $NA = 0.45$. Additionally, we observed the sample in conoscopic mode by introducing a Bertrand lens between the analyzer and the ocular. The FCPM technique³² was employed to resolve the LC orientation spatially. A $20\times$ air objective, $NA = 0.5$ on a Leica TCS-SP2 confocal microscope was employed for this purpose. Linearly polarized laser light ($\lambda_{\text{exc}} = 543 \text{ nm}$) at a power of $50 \mu\text{W}$ excited the fluorescent Nile Red in the nematic matrix. The excitation laser was used at low powers to avoid any photo-induced reorientation of the LC molecules and photobleaching of the dye molecules. The fluorescence signal obtained from the FCPM measurements varied with the relative orientation of LC molecules (which corresponded to that of the dye molecules) and the polarization direction of the exciting laser. The maximum fluorescence intensity was obtained for mutually parallel orientation. The signal intensity reduced continuously as the relative angle increased, yielding the minimum signal at mutually orthogonal orientation. These correspond to 1 and 0 respectively on the intensity scale bar as shown in Fig. 3a. Signals were obtained from two different reference planes: xy -scanning with stacking in the z direction, and yz -scanning with stacking in x , by varying the direction of the laser beam polarization continuously from 0 to $\pi/2$. The director field within the channel was finally reconstructed by a combination of the obtained POM and FCPM data.

2.5 Flow velocity measurement

Dual-focus fluorescence correlation spectroscopy (2fFCS) was employed for measuring the velocity profile of the flowing LC. This technique is particularly relevant to measuring flow profiles of LC flows coexisting with topological defects. The use of particle tracking velocimetry for such coexisting systems is unsuitable due to the interaction of the tracer particles with topological defects. For 2fFCS, we mixed the LC with a lipophilic dye ATTO Oxa12 at nanomolar concentration. The dye-doped LC was flowed within a microfluidic chip bonded to a microscopy cover slip. Measurements were performed on a standard inverted confocal microscope for time resolved fluorescence measurements (MicroTime 200 with dual-focus option, PicoQuant GmbH, Germany), schematically depicted in Fig. 2a.²⁶

Light from two identical pulsed diode lasers ($\lambda_{\text{em}} = 640 \text{ nm}$, pulse duration 50 ps fwhm) with orthogonal polarization was combined with a polarizing beam splitter and coupled into a polarization-maintaining fiber. Both lasers were pulsed alternating with a repetition rate of 20 MHz , with an average laser power adjusted to $20 \mu\text{W}$ each. The light from the fiber is collimated and reflected by a dichroic mirror into the microscope objective (UPLSAPO $60\times$ W, 1.2 N.A. , Olympus Deutschland GmbH,

Fig. 2 Flow velocity (v) measurement using 2fFCS. (a) Schematic of the 2fFCS setup. The light of two perpendicularly polarized laser beams is combined by a polarizing beam splitter, passed through an optical fiber, and reflected towards the objective by a dichroic mirror. Before entering the objective, the light is passed through a Normarski prism which deflects the light depending on its polarization. After focusing the beams through the objective, two overlapping foci are generated. The resulting fluorescence light is collected by the same objective and then focused through a confocal aperture and detected by two single photon avalanche diodes. (b) Typical 2fFCS correlation curves for Atto Oxa12 in NLC 5CB flowing through a rectangular microfluidic channel with a volume flow rate of $0.05 \mu\text{L min}^{-1}$. The four curves comprise two autocorrelations (one from each focus; blue and red curve) and the forward and reverse cross-correlations (green and yellow curve). Solid lines are fits to the data (squares). The data shown here correspond to measurements in the *parallel* orientation, and therefore show a maximum in the *forward* cross-correlation. The resulting flow rate as determined by 2fFCS measurement was $124.1 \mu\text{m s}^{-1}$.

Germany). The fluorescence signal is collected by the same objective and focused on a confocal pinhole (diameter $150 \mu\text{m}$). The signal is further split by a polarizing beam splitter onto two single-photon avalanche diodes (SPCM-AQR-13, PerkinElmer Optoelectronics, Germany). Prior to each flow-profile acquisition, a cross-sectional image of the channel was acquired by moving the channel with the attached piezo electrical stage (P-562.3CD, Physik Instrumente, Germany). Flow profiles were acquired at discrete points along a line at half the height of the channel. Typical acquisition time was two minutes per point. Data analysis and fitting were subsequently performed using custom Matlab routines (Fig. 2b). The velocity of the flowing nematogen was measured across the channel width at a distance of $100 \mu\text{m}$ from the pillar on both upstream and downstream directions.

3 Results and discussion

3.1 Director morphology in the absence of flow

The equilibrium nematic director in the absence of flow was studied for a range of microchannels having different

combinations of aspect ratio (w/d) and pillar dimension (r). Fig. 3a shows micrographs of a $(r, d, w) = (20, 40, 100) \mu\text{m}$ channel filled with nematic 5CB, and observed under white light and confocal microscopes. The white light micrograph (top) clearly shows the channel boundary and the cylindrical micro-pillar. When crossed-polarizers were introduced, distinct dark and bright patterns were observed in the vicinity of the confining walls and the pillar surface. The sample stage was rotated gradually by $\pi/4$, till one of the polarizers was parallel to the channel length. At this relative orientation between the channel and polarizers, the contrast between the dark and bright stripes reduced to a minimum. However, across the central dark region, no variation of the transmitted intensity was observed. The director (hence the optical axis) orientation in the proximity of the surfaces is thus distinct from that within the rest of the nematic bulk. Close to the pillar and confining surfaces, the director experiences a gradual elastic deformation to accommodate the existing boundary conditions. Effectively,

bending of the director near the surfaces alters the relative orientation of the optical axis and light polarization, and generates ordinary and extraordinary components from the transmitted white light. These components interfere and consequently produce the dark and bright interference pattern that aligned parallel to the surfaces possessing homeotropic anchoring. The number of fringes appearing in the pattern increased with the channel depth.²¹ Farther away from the pillar and confining surfaces, the LC director equilibrates without any elastic deformation. Owing to the surface-induced homeotropic alignment, the director is orthogonal to the light polarization irrespective of the rotation of the sample stage. The whole-field director configuration was further confirmed from the FCPM measurements (xy scan, laser polarized along y). As seen in the FCPM micrograph (Fig. 3a), a strong fluorescence signal is observed at the PDMS boundaries, whereas a weak signal is obtained from the rest of the channel.

The lateral spread of the interference pattern at the surfaces varied with the channel depth. As is shown in Fig. 3b (top), no significant interference is observed within a channel of depth, $d \approx 10 \mu\text{m}$. The reduction in the number of the interference fringes within shallow channels additionally leads to a sharp appearance of the channel and pillar boundaries. In contrast, the interference pattern is significantly prominent in the $(20, 40, 50) \mu\text{m}$ channel (Fig. 3b, bottom). Due to the relatively small vertical separation (depth, d) in shallow channels, the confining surfaces (top and bottom) inhibit elastic deformations close to the channel walls. However, as the channel depth increases, stabilization of the director due to the interaction between the top and bottom surfaces weakens. Additionally, as the channel width decreases, the anchoring contribution from the side walls competes against the vertical director stabilization, and leads to a gradual elastic deformation. Thus, within a channel with $w/d = 1$, the molecules have equal tendency to orient orthogonal to either of the xy or zx planes. Effectively this results in an integer singularity at the center of the channel, which however is seldom observed experimentally. Instead, the director conforms to an energetically favourable configuration by escaping to the third dimension, as is also observed in capillaries.³³ By varying the aspect ratio ($w/d > 1$) of the channel, we were essentially able to tune the homeotropic coverage within the microchannels.

Fig. 3c shows POM and FCPM images obtained in a $(30, 40, 100) \mu\text{m}$ channel. FCPM intensity distribution from the channel cross-section along sections 1 and 2 (Fig. 3d) clearly resolves the homeotropic orientation of the LC molecules on the channel walls and the pillar surface. Interestingly, the lateral spread of the bend deformation at the side walls, seen as the bright fluorescence signal, is wider in section 1 as compared to that in section 2. This difference occurs due to larger elastic deformations effected by the micro-pillar. For a given aspect ratio, the elastic interaction is more significant in the presence of a larger pillar than a smaller one. For example, a $40 \mu\text{m}$ diameter pillar (Fig. 3a) reduced the lateral spread of the bend deformation at the walls by $\approx 50\%$, whereas, the presence of a $60 \mu\text{m}$ pillar reduced the spread further to $\approx 75\%$ (Fig. 3c). The overall

Fig. 3 Equilibrium director orientation without flow. Micrographs (xy plane) showing channels filled with nematic 5CB. Polarized micrographs are indicated by a pair of crossed arrows (polarizers at $\pi/4$ to the channel length). (a) From top: white light, polarized optical, and fluorescence confocal polarized micrographs (laser polarization at $\pi/2$ to the channel length, indicated by the double-headed arrow) of a $(r, d, w) = (20, 40, 100) \mu\text{m}$ channel. 0 and 1 on the FCPM intensity scale bar correspond to director orientations parallel and perpendicular to the laser polarization direction. (b) From top: POM images of a $(r, d, w) = (30, 10, 100)$ and $(r, d, w) = (20, 40, 50) \mu\text{m}$ microchannel respectively. Note the reduction of the number of interference fringes in the shallow $(r, d, w) = (30, 10, 100) \mu\text{m}$ channel. (c) POM and FCPM images of a $(r, d, w) = (30, 40, 100)$ microchannel. (d) FCPM imaging of the yz cross-sections along the lines 1 and 2 in (c). The polarization of the excitation laser is shown with the double-headed arrow. (e) Reconstruction of the director field from the POM and FCPM images. Hatched lines depict the glass surface. Cross-sectional view (yz) through the pillar (shown in grey colour) shows two possible director orientations around it: singular $+1/2$ defect loop (top) or a non-singular director configuration (bottom). Both configurations possess symmetry about the cylinder z axis. (f) Schematic nail representation of the director field on the xy plane. The nail head presentation indicates that the director tilted partially into the image plane.

confinement-induced behavior, discussed so far, is generally observed also in the presence of multiple pillars within a homeotropic matrix.

The director field around the micro-pillar, as shown in Fig. 3e and f, was reconstructed from the analyzed POM and FCPM data. At the surfaces of the confining walls and the micro-pillar, LC molecules are homeotropically anchored. Far from the surfaces, the director is normal to the xy plane (out of the POM micrograph plane), represented by black dots in Fig. 3f. In the vicinity of the side walls and the pillar, the director undergoes a transition from the in-plane to the out-of-plane orientation through a gradual bend deformation (indicated by the nail heads in Fig. 3f). The director orientation around the micro-pillar preserves a rotational symmetry about its axis (z). The existing boundary conditions however lead to two possible director orientations about the yz plane (Fig. 3e): director configuration with a singular $+1/2$ defect loop (sharp pillar base), and a continuous non-singular director field (rounded pillar base). Although symmetric projections of the above two cases are shown in Fig. 3e, asymmetric director configurations are also likely.^{34,35} Typically in our experiments we have observed the non-singular configuration long time after equilibration from the isotropic to the nematic phase. The singular structure generally appeared at slightly elevated temperatures, under external perturbations, or due to pinning of the defect loop at surface irregularities.

3.2 Director morphology in the presence of flow

The surface-induced equilibrium of the director configuration (Section 3.1) is altered as flow of the nematogen is initiated within the microchannel. The director undergoes reorientation along the flow direction,²¹ which increases non-linearly with v . In Poiseuille flow of a homeotropically aligned sample, two different transition layers of director deformation develop.^{5,7} Due to the difference of the local shear rates is close to the surface and at the channel mid-plane ($z = d/2$), the thickness over which the nematic director reorients away from the homeotropic alignment is different. With increasing the flow speeds, the transition thickness reduces, leading to alignment of the nematic director along the flow. The director orientation within the flowing matrix progressively attains the saturation Leslie angle,¹¹ $\theta_L = \tan^{-1} \sqrt{\alpha_3/\alpha_2}$ ($\approx 22^\circ$ for nematic 5CB). Here α_2 and α_3 are the Leslie viscosities satisfying the Parodi relation $\alpha_3/\alpha_2 > 0$, and θ_L is measured relative to the flow direction about the xy plane.³⁶ Owing to the small dimensions of the channel, the Reynolds number, Re , is generally low ($\approx 10^{-3}$), ensuring a Stokes regime hydrodynamically. However, it is the competing effect between the viscous and elastic parameters of the LC that determines the flow-induced director orientation. The non-dimensional Ericksen number, $Er = \mu v d^*/K$, captures the relative influence of viscosity (μ) and nematic elasticity (K) within the flowing matrix. Here v and d^* , respectively, denote the flow velocity and the relevant length scale of the confinement. Ideally at $Er = 1$, the viscous and elastic effects balance each other. However, depending upon the boundary conditions,

numerical³⁷ and experimental³⁸ investigations have shown that the flow-director equilibria could be achieved at $Er > 1$ as well.

For a given combination of channel dimensions and boundary conditions, Er depends only on the velocity v , and determines the flow-induced director orientation. The presence of an obstacle within the microchannel modifies the flow path, and consequently the director configuration. As summarized in Fig. 4, a sequence of topological defect structures, generated due to the flow-director interplay, are observed. In the absence of flow (Section 3.1), the free-energy barrier between the non-singular and singular director configurations is easily overcome under normal experimental conditions. Therefore, even at a small flow speed of $v \approx 8 \mu\text{m s}^{-1}$, a singular loop stabilizes at the xy mid-plane (Fig. 4b), without any significant modification of the initial equilibrium director configuration (Fig. 4a). As the speed increases, the director undergoes deformation, accompanied with extension of the loop defect (Fig. 4c). The extension continues until the extended loop experiences a structural instability at higher speeds (Fig. 4d). Development of the instability is marked by the increasing measure of flow-alignment of the nematic director. Ultimately, this results in the formation of a hyperbolic hedgehog defect (-1) in the pillar downstream (Fig. 4e) while the micron-sized pillar now mimics a virtual radial defect ($+1$). The hyperbolic hedgehog defect,

Fig. 4 Morphological evolution of the defect structures in the presence of flow. (a) Schematic representation of the semi-integer defect loop around the pillar in the absence of flow. (b) In the presence of a small flow or perturbation, the loop shifts up vertically to the channel mid-plane. (c) At higher speeds, flow-induced director distortion results in extension of the loop along the downstream direction. (d) Instability of the stretched loop sets in just prior to the flow-alignment of the LC director along the xy mid-plane ($z = d/2$). (e) The transition to flow-alignment at the mid-plane effects a topological transformation of the loop into a hyperbolic (-1)-radial ($+1$) hedgehog pair. The hyperbolic hedgehog defect (red dot) is downstream to the pillar, and a (virtual) radial defect at the pillar center forms a Néel wall type configuration in between them. (f) The Néel wall structure is stretched downstream on increasing the flow speed further, finally transforming into a diffused Bloch wall.

though observed to be metastable in deeper channels ($d > 20 \mu\text{m}$), is typically stable within shallow channels, and also on reversing the flow. Further increase of the flow speed results in a defect wall (Fig. 4f), which jets out and extends over hundreds of μm downstream.

3.2.1 Semi-integer defect loop. In the presence of flow, the defect loop experiences the viscous force and is pulled downstream. Tethered to the micro-pillar, the loop is progressively stretched on increasing the speed (and Er). The stretch length, l , was measured as the distance between the pillar center and the leading end of the defect (Fig. 5, inset).

As plotted in Fig. 5, l increased monotonically with the flow speed, linearly in the initial stages (upto $Er \approx 80$), after which a non-linear dependence on Er was observed. Interestingly, at comparable flow speeds, the defect loop within a shallow ($(r, d, w) = (30, 10, 100) \mu\text{m}$) channel clung too close to the pillar for optical detection (Fig. 6).

The magnitude of the director reorientation, determined by the interplay of viscous and elastic interactions, is different in the vicinity of the micro-pillar, than that away from it. At the micro-pillar the flow is divided into two symmetric halves on either side of the pillar, each of which travels with higher local speeds. Larger reorientation of the nematic director thus follows in the region between the channel wall and the micro-pillar, visualized as a distinct birefringent hue (red/yellow) in Fig. 6, $t = 0$ s. Similarly, the symmetrical division of the flow was directly observed when a flow-aligned front⁵ arrived at the pillar (Fig. 6, $t = 0.3$ s). Depending upon the local shear stress, director distortion around the pillar was minimum at the hydrodynamic stagnation point ($v = 0$, the arrow-head in Fig. 6) and maximum at angular separations of $\pm\pi/2$ relative to the stagnation point. The distortion however goes down once again on moving beyond $\pm\pi/2$. Consequently, the no-flow director symmetry about the pillar axis is disrupted in the presence of flow. With increasing the speed, a flow-induced symmetry about the azimuthal (xy) plane cutting the channel into two halves at

Fig. 5 Extension of the singular loop in a $(r, d, w) = (20, 40, 100) \mu\text{m}$ channel. The maximum length (measured between the pillar center and the leading end of the defect) shows a non-linear dependence with the Ericksen number, Er . Insets: micrographs show extension of the semi-integer defect loop with increasing the flow speed, observed between crossed-polarizers. Scale bar: $50 \mu\text{m}$.

Fig. 6 Time sequence of polarized micrographs represents the flow-alignment of the nematic director in a $(r, d, w) = (30, 10, 100) \mu\text{m}$ channel. The arrow-head indicates the hydrodynamic stagnation point ($v = 0$). At $t = 0$ s, a distinct birefringent domain (green in appearance) with a parabolic boundary is observed upstream of the micro-pillar. On the xy mid-plane ($z = d/2$), the nematic director is oriented along the flow behind the parabolic front, whereas, ahead of it, the director is vertical. As the front moves downstream ($0 < t < 0.3$ s) and encounters the cylindrical pillar ($t = 0.3$ s), the director around the pillar is oriented along the flow. The front however travels with reduced speed at the center ($y = w/2$), seen in the time sequence $t > 0.4$ s. Finally, it separates from the pillar ($t = 0.9$ s) and subsequently regains the parabolic profile further downstream. Scale bar: $50 \mu\text{m}$.

$d/2$ (Fig. 4c) begins to evolve. Experimentally, this was resolved by comparing the polarized micrographs at $t = 0$ and $t = 0.9$ s in Fig. 6. In the latter, the director field suggests the creation of a virtual +1 radial defect at the pillar center. Within deeper channels, this topological transformation is observed as an instability of the extended defect loop (see ESI-1†). Quantitative estimation of the director and its gradient around the pillar and the stretched loop should be possible through numerical studies.

Evolution of the +1 defect is topologically balanced by the spontaneous creation of a -1 hedgehog defect downstream of the pillar, shown in Fig. 7b. The hedgehog defect is stable particularly within shallow channels (Fig. 7d) due to the strong influence of anchoring induced by the surfaces. However, within a deeper channel, it is readily convected downstream and transformed into a defect wall (Fig. 4e and f). This phenomenon, which sets off at $v \approx 90 \mu\text{m s}^{-1}$ in a $40 \mu\text{m}$ deep channel (see ESI-2†), qualitatively resembled the viscous jetting in microflows.³⁹ Similar wall defect structures are generated also within a $10 \mu\text{m}$ (shallow) channel, however only at high flow speeds $v > 150 \mu\text{m s}^{-1}$ (Fig. 7c and e). Deeper channels were suitable for the morphological investigation of the defect wall (Section 3.2.2): In addition to a better optical access to the director configuration, the defect structure evolved spontaneously even at a low v .

3.2.2 Morphology of the wall defect. The flow-induced director morphology of the wall defect was resolved using a combination of POM and FCPM studies as shown in Fig. 8. A sequence of polarized micrographs show the development of the wall defect in Fig. 8a. A fully developed wall in steady-state condition is shown in ESI-3.† Using high magnification POM, we identified two distinct planes z_1 and z_2 , at which the wall was

Fig. 7 POM imaging of the flow-induced director field within a $(r, d, w) = (30, 10, 100)$ μm channel. (a) Flow alignment: increasing transmission of the polarized light (top to bottom) indicates progressive reorientation of the nematic director with flow. Note that the birefringence observed between the pillar and channel walls is distinct from the rest of the channel due to the high local shear rate. (b) Formation of a hyperbolic hedgehog defect downstream of the pillar (the second micrograph from top, indicated by the arrow-head). The hedgehog is stretched apart from the pillar as the flow speed increases, finally resulting in the formation of the defect wall (c). Scale bar: 50 μm . Magnified view of the hedgehog defect (d) and the defect wall (e).

Fig. 8 Evolution of the wall defect within a $(r, d, w) = (20, 40, 100)$ μm channel. (a) Polarized micrographs show the development of the wall defect. At $Er \approx 100$ a fully developed wall jets out of the pillar and extends over hundreds of micrometers downstream. Scale bar: 40 μm . (b) High magnification POM micrographs of the wall structure. The images were taken at two different focal planes z_1 and z_2 , separated vertically by ≈ 11 μm . (c) Corresponding white light (without polarizers) micrographs. Image taken at z_2 shows a bright core running through the length of the defect. Scale bar: 10 μm . (d) Polarized fluorescence micrographs of the defect structure at the point of origin and further downstream. Direction of the laser polarization was along the flow direction in both cases (double-headed arrow). Scale bar: 50 μm . (e) Cross-section (yz plane) FCPM image through the wall defect. The flow and laser polarization directions are normal to the image plane. Scale bar: 10 μm . (f) Schematic of the director field projection (xy plane) around the pillar, resolved using POM and FCPM imaging data.

focussed with maximum contrast (Fig. 8b). At the image plane z_1 two high contrast bright parallel lines, separated by approximately 8 μm , were observed. These lines meet each other at the anterior end of the defect through an angular curvature. On

varying the distance between the sample and the objective, the bright lines moved out of the objective focus continuously. With the reduction of the sharp contrast, the lines now appeared diffused. However, at z_2 , at a vertical separation of ≈ 12 μm from z_1 , a second focussed micrograph was obtained (Fig. 8b, bottom). The micrograph at z_2 revealed a texture distinct from the one obtained at z_1 . We observed two closely placed thin parallel lines which terminated also at the anterior end of the defect. These lines, located symmetrically within the ones detected at z_1 , enclosed a narrow dark region, of width less than 1 μm , between them. Further observations carried out in white light (without the polarizers) are shown in Fig. 8c. Imaging at the z_2 plane revealed a bright core running through the entire length of the defect which coincided with the closely placed lines observed between crossed-polarizers. The core, surrounded by darker diffused boundaries, disappeared gradually on focussing the sample back to the z_1 plane. Moreover, the above boundary coincided with the bright parallel lines observed between crossed-polarizers at z_1 . The white light micrographs thus suggest a lens-effect through the defect, which arises due to difference in the refractive index between the core and the outer parts of the defect.

Fig. 8d and e show the xy and yz sections of the defect observed using FCPM technique. Corresponding to the bright lines (z_1 plane) in Fig. 8b, a parallel set of low-fluorescence-intensity lines, separated by ≈ 8 μm , were observed. However, a high fluorescence signal, comparable to that of the flowing nematic bulk, was obtained from the region confined between the low intensity lines. While the low fluorescence signal suggests that the LC molecules are orthogonal, the high signal indicates alignment parallel to the laser polarization direction. The cross-sectional micrograph of the defect (Fig. 8e) additionally shows the spread of the structure along the vertical direction and existence of a three-dimensional structure resembling a Bloch wall. The director at the center of the wall (Fig. 8b, bottom) undergoes an out-of-plane twist deformation that extends along the channel depth. Combining the POM and FCPM observations, the resulting projection of the nematic director is proposed in Fig. 8f. In the case of an obstacle possessing spherical symmetry, the structure is expected to possess a circular cross-section. The overall structure in the present case is analogous to that of a defect stretched from a homeotropic colloid or droplet.^{19,23,25} However, the absence of rotational symmetry along the flow imprints a vertical extension on the singular defect arising from the micro-pillar obstacle.

3.2.3 Dynamics of the wall defect. Movie ESI-4† shows the life span of a defect wall tethered to the micro-pillar. Typically lasting over a few tens of seconds, the defect life initiates with its creation, followed by stretching, and finally it terminates when the wall snaps off. Thereafter, the wall regains its length over time and the cycle is repeated. The snapped-off portion constitutes an umbilic defect^{40,41} which is convected downstream by the flow. The posterior and anterior ends of the annihilating structure are constituted of +1 radial and -1 hyperbolic hedgehog defects respectively, forming a classical umbilic defect. The umbilic possesses a rotational symmetry about the axis joining the defect pair. While travelling

downstream, the umbilic annihilates, as observed from the progressive reduction in its length with time. The annihilation dynamics and its dependence on the flow rates will be discussed elsewhere.

The time dependence of the length of the wall defect, l , was measured for three different flow speeds, as shown in Fig. 9. As shown in Fig. 9b, the length of the wall increased linearly during the early stages of the life-time: over a span of ≈ 3 s, the lengths increased by approximately $450\ \mu\text{m}$ (black squares), $600\ \mu\text{m}$ (red circles), and $1100\ \mu\text{m}$ (blue triangles). As time increased, the defect length gradually approached a saturation behavior, distinctly visible in the plot corresponding to $v \approx 200\ \mu\text{m s}^{-1}$ (Fig. 9b). At higher flow rates the wall extended beyond the POM field of view. During the final stages of life, the defect reached a maximum length at $t \approx 21$ s, after which the length started to reduce. This is shown in Fig. 9b (inset) at a flow speed of $v \approx 200\ \mu\text{m s}^{-1}$. The latter marked the defect snap-off and generation of the umbilic structure.

Fig. 10a plots the corresponding rate of extension of the wall $\dot{l}$ as a function of time. Irrespective of the flow speed, the defect accelerated right after its creation and attained a maximum speed. The time span of the initial acceleration however shortened with increasing the velocity of the nematic flow. After reaching the maximum, the wall retarded and travelled at lower speeds. $\dot{l}$ decreased monotonically over time and changed its sign ($+ \rightarrow -$) in the final stages of the defect life. As shown in Fig. 10a (inset), $\dot{l}$ reduced continuously over $16\ \text{s} < t < 24\ \text{s}$ and changed its sign at $t \approx 21$ s. The coordinates of the sign change correspond to the maximum on the plot shown in the inset of Fig. 8b. Thus, at its maximum extension, the wall defect halts momentarily just prior to the snap-off upstream in the channel,

Fig. 9 Length of the defect wall. (a) POM image of a fully developed wall stretched in the pillar downstream region (crossed polarizers with one polarizer along the flow direction). The defect length l is defined as the distance between the pillar center (red dots) and the leading end of the defect (red arrow head). Scale bar: $50\ \mu\text{m}$. (b) The plot shows the length of the defect as a function of time for three different flow rates of the nematogen. The inset shows the defect length during the late stages (shown here for $v = 200\ \mu\text{m s}^{-1}$). In this case it reaches a maximum of $l \approx 1300\ \mu\text{m}$, before snapping off. At this instance the leading end begins to move backwards.

Fig. 10 Wall velocity. (a) The plot shows the time dependence of the rate of change of the defect length, shown here for three different flow speeds. The inset shows the time dependence towards the later stages ($v = 200\ \mu\text{m s}^{-1}$). Observe the change of sign at $t \approx 21$ s. (b) Time sequence of polarized micrographs represents the process of snap-off of the defect wall. Each micrograph is separated by 1 s, the time increasing from top to bottom. The snap-off produces an integer defect pair, which annihilates while convecting downstream. The wall originating from the pillar regains its length over time.

indicated by the arrow-head in Fig. 10b (top micrograph). As seen in the sequence of micrographs, the defect wall connected to the pillar regains its length over time, while the umbilic annihilates traveling downstream.

3.2.4 Flow reversal: wall to hedgehog transformation. The above-discussed evolution of defect structures reverses their sequence when the flow rate is gradually reduced to zero. However, an abrupt termination (ESI-5†) or a gradual reversal (ESI-6†) of the flow results in transformation of the wall into a well-defined radial-hyperbolic hedgehog defect pair. This is particularly distinct in deeper channels, as presented by the micrograph sequence in Fig. 11a. The transformation leads to reduction of the wall length, and is accompanied with modification of the POM texture (see the last two micrographs in Fig. 11a). The anterior end of the wall transforms into a fully developed -1 hedgehog defect (ESI-7†). Fig. 11b and c show the final stages of the transformation with the pillar and channel boundaries visible (a low magnification objective was used in this case). The topological sign of the defect was estimated by rotating the polarizers and maintaining them always in the crossed position. The sense of rotation of the dark extinction brushes around the defect point was found to be opposite to that of the polarizers. Additionally, the brushes around the pillar rotated in the same sense as the polarizers, confirming the $+1$ topological nature of the pillar center. With the formation of the hedgehog defect, the two distinct focal planes, discussed in Section 3.2.2, were no more observed. Correspondingly, in white light, the bright defect core observed previously was substituted by a point defect, which appeared

Fig. 11 Flow reversal. (a) POM image sequence showing transformation of the wall into a hyperbolic hedgehog defect on gradual reversal of the flow direction. Note the accompanying textural change, especially in the last two micrographs. Micrographs in (b) and (c) show the final stages, imaged using a lower magnification objective (larger field of view). Scale bar: 50 μm . (d) The schematic representation corresponding to a Néel wall type director configuration.

dark due to scattering of light. The textural changes show that an out-of-plane director orientation within the Bloch wall transformed into an in-plane (xy) bend deformation, reminiscent of a Néel wall type configuration. The corresponding director field defining the Néel wall configuration is schematically shown in Fig. 11d.

4 Flow velocity distribution

LCs within micro-confinements can be typically described as a system of coexisting low and high molecular order species. The low ordering corresponds to different kinds of singular topological defects present within the LC matrix of a relatively higher molecular ordering.¹⁶ The presence of inclusions, *e.g.* colloids or droplets, within the nematic matrix leads to accompanying defects owing to localized frustration of the nematic order.⁴² Consequently, the conventional technique of particle tracking turns out unsuitable for the velocity measurement within such coexisting systems. Not only do the tracer particles dispersed within the LC spontaneously form self-assembled structures,⁴³ they also experience elasticity-mediated interactions with the defects induced by flow.⁶ Consequently, the tracer trajectory in the vicinity of a defect structure does not depict an accurate flow field, and hence is not suitable for velocimetry within coexisting phases of different ordering.

We address this experimental challenge by introducing the technique of dual-focus fluorescence correlation spectroscopy – 2fFCS (Section 2.5) to measure flow fields even in the vicinity of the defects. The velocity distribution at $z \approx d/2$, was evaluated across the two lateral sections 1 and 2, shown in Fig. 12a. Relative to the pillar, these were located 100 μm behind (pillar upstream) and ahead (pillar downstream) of the pillar, respectively. Fig. 12b shows the velocity distribution (v/v_{max}) through sections 1 (red dots) and 2 (black squares). The velocities have

Fig. 12 Velocity distribution across the wall defect. (a) POM image showing the pillar upstream (section 1) and pillar downstream (section 2) regions. Scale bar: 40 μm . (b) Normalized velocity distributions (red dots: pillar upstream, black squares: pillar downstream) measured at half depth of the channel using dual-focus FCS are plotted as a function of channel width. The position of the defect wall is shown by the normalized intensity distribution across section 2 (blue triangles).

been normalized with respect to the maximum velocity, $v_{\text{max}} = 456 \mu\text{m s}^{-1}$, obtained experimentally from the measurements. Additionally, a plot of the normalized intensity distribution (through section 2) through the defect wall indicates the position of the latter (Fig. 12b, blue triangles). A whole-field velocimetry using 2fFCS is expected to be a practical tool for resolving the local rheological parameters in the vicinity of topological defects in flow. The velocity field measured across the defect line in Fig. 12 (black squares) indicates possible enhancement of the flow speed at the center of the defect wall. Further investigations are underway to resolve the three dimensional velocity field in the vicinity of the defect line.

5 Conclusions

In this paper we have reported on the flow of a nematogen past a micro-sized cylindrical obstacle. The resulting morphology and dynamics of the defects were analyzed through experiments carried out within channels of different aspect ratios using POM and FCPM techniques. Though the static director distribution was determined by the obstacle geometry, the flow-induced morphology did not exhibit any significant dependence on the geometry of the pillar. Fabrication of the microfluidic confinements typically yielded PDMS structures with rounded edges, which resulted in continuous director field. However, around the pillar, singular configurations were observed at slightly perturbed systems or due to surface irregularities. The reported observations can be generalized for different combinations of r , d , and w . Though, within shallow channels ($d < 10 \mu\text{m}$), the formation of the singular loop is hindered, the hyperbolic hedgehog defect evolved after flow-alignment is observed to be stable. In contrast, within a deeper channel ($d > 20 \mu\text{m}$), a well-defined singular loop is observed, though the hedgehog

structure is typically unstable. Thus, a higher flow speed was necessary to induce the hedgehog-to-wall transformation within shallow channels. Nevertheless, the defect wall was observed within the entire range of channel depth ($7 < d < 50 \mu\text{m}$) considered in this work. Furthermore, this behavior is supported by the nematic elasticity relationship $K_2 < 1/2(K_1 + K_3)$,⁴⁴ where $K_1 = 6.2 \text{ pN}$ (splay), $K_2 = 3.9 \text{ pN}$ (twist), and $K_3 = 8.2 \text{ pN}$ (bend) denote elastic constants for nematic 5CB. Conversely, the stability of the director transformation on flow reversal also exhibited qualitative dependence on the channel depth, d . Within channels $d < 10 \mu\text{m}$, reduction of the flow speed readily converted the wall defect to a hedgehog. On the other hand, even small perturbations created within the flow were sufficient to transform the hedgehog configuration to the Bloch wall type. The Néel wall configuration was better stabilized within a channel with lower d . Homeotropic anchoring on the top and bottom surfaces of a shallow channel stabilized the hedgehog defect point of the Néel wall structure. Since the director readily reorients by flow in a deeper channel, the equilibrium was readily shifted towards the formation of the wall structure. It is thus expected that the flow-director interactions observed within the shallow channels can be analogously simulated even within deeper channels by enhancing the boundary effects, e.g. by increasing the strength of homeotropic anchoring with appropriate surface treatment. Furthermore, 2fFCS is demonstrated as a potential technique for velocity measurements in coexisting systems of ordered and disordered phases.

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