Small Surface Pretilt Strikingly Affects the Director Profile during Poiseuille Flow of a Nematic Liquid Crystal

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(Received 1 February 2010; revised manuscript received 8 April 2010; published 14 June 2010)

Conventional nematic liquid crystal cells are fabricated with small surface pretilt of the director induced by rubbed polymer alignment. Depending on the orientation of the bounding surfaces, this may lead to two slightly different untwisted director configurations, splay and parallel. This small difference leads to remarkably different director profiles during pressure-driven flow, observed here using optical conoscopy. Data show excellent agreement with numerical modeling from Leslie-Ericksen-Parodi theory.

DOI: 10.1103/PhysRevLett.104.248301 PACS numbers: 47.57.Lj

Complex fluids play an important role in many technological applications. These can range from low molar mass liquid crystals in liquid crystal displays to liquid crystal polymer (LCP) injection moulding and spinning of textile fibers [1]. Quality, order, and defects in finished products fabricated from methods such as LCP injection moulding can often be very sensitive to the molecular orientation and flow channel dimensions prescribed [2]. To this end, a full understanding of the pressure-driven flow characteristics of nematic liquid crystals in micron-sized channels is of some significance.

The dynamic behavior of nematic liquid crystals under varying flow conditions has been explored by many. Dynamic reorientation of the director (a unit vector, $\hat{\boldsymbol{n}}$, defining the principal axis of orientation) under the application of an electric field has been shown to affect the response time of displays [3,4], as well as the *flow induced* transition from the V state (director aligned homeotropically at the cell center) to the H state (director aligned planar homogeneously at the cell center) above a critical velocity in a homeotropically aligned cell [5]. Pieranski and Guyon [6] have examined the hydrodynamic instability (suggested to be a hydrodynamic analogue of the Freedericksz transition) for an initial azimuthal alignment normal to the flow direction, where a threshold velocity occurs. Further work has been undertaken on nematic flow alignment in samples with colloidal dispersions and the topological defects associated with them, as are commonly found in foods, paint, and drugs [7]. The work presented here follows from a previous study examining director orientation under pressure-driven flow for a sample aligned at an initial azimuthal angle $\phi_0 = 45°$ to the direction of flow [8].

Van Horn *et al.* [9,10] have shown for the sheared flow of N-(4-methoxybenzylidene)-4-butylaniline (MBBA) that surface pretilt may play an important role in the average director distortion, depending on the magnitude of the pretilt and the initial director alignment (rubbing direction) relative to the flow. Here, we present a pressure-driven or Poiseuille flow study, examining the properties of liquid crystal cells filled with 5CB (4-Cyano-4′-pentylbiphenyl) having high (almost normal to the flow) initial azimuthal alignment angle (ϕ_0) and small surface pretilt (θ_0) (from buffed polyimide [11]) in both the splayed and parallel aligned states (see Fig. 1).

In general, liquid crystal flow experiments have involved shearing one boundary plate of an aligned cell relative to the other at a constant velocity, producing a linear velocity distribution across the depth of the cell [12–14]. Under pressure-driven flow, a distinctly different, often symmetric about the cell midplane, velocity distribution is observed [15] (see Fig. 2). Similar work on the large variation in director orientation for different (planar and homeotropic) anchoring conditions has also been modeled [16,17] in some depth.

In the absence of disclinations, the dynamics of nematic fluid flow may be described by the continuum theory proposed by Leslie [18], Ericksen [19], and Parodi [20]. Modeling flow along channels is properly a three-dimensional problem, but provided that the channel is far longer and broader than it is deep, much of its behavior can be understood in terms of a one-dimensional model. Far from the inlet pipe, the flow is assumed to be uniform along the x axis, parallel to the channel, and to vary quadratically

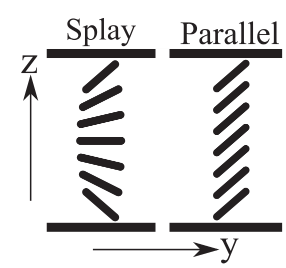

FIG. 1. A schematic diagram representing the difference between the initial splayed and parallel director alignments in a nematic flow cell. For this figure, the flow direction can be visualized as either into or out of the page, noting that experiments were carried out at $\phi_0 \approx 87°$, i.e., initial azimuthal alignment distorted 3° into the page.

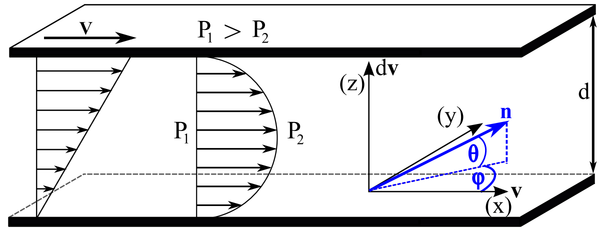

FIG. 2 (color online). A schematic diagram representing the difference in velocity profiles in a cell under sheared flow (on the left) and under pressure-driven flow on the right, where **v** is the velocity of the sheared plate, d is the cell thickness, and P_1 [pressure in a given (y-z) plane] is greater than P_2. A standard coordinate system is also shown, with director orientation defined by two polar angles (θ, ϕ).

in y, across the width of the channel, with a maximum at the center. The resulting model has four variables: the Euler angles θ and ϕ which give the director,

$$\hat{\boldsymbol{n}} = (\cos\theta\cos\phi, \cos\theta\sin\phi, \sin\theta), \tag{1}$$

and the x and y components of the velocity [21]. By prescribing the pressure gradient on the right-hand side of the Ericksen-Leslie equations, steady-state director profiles can be related to the flux of material at the inlet. A more detailed description of the one-dimensional model and the computer program written to solve it numerically is given by Cornford [22].

Because of the birefringence exhibited by an ordered nematic liquid crystal, conoscopy enables determination of both the average azimuthal director angle and the mean absolute tilt angle exhibited by a sample. Azimuthal distortion is revealed by symmetry of the conoscopic figure without accounting for the effect of tilt distortion on total birefringence. The conoscopic interference figure gives an average but smaller value of the magnitude of director distortion at the midplane of the sample, as described by Boudreau *et al.* [13]. The azimuthal angle of the measured conoscopic figure rotation is compared with figures computed from the modeled director profiles by Berreman's 4×4 matrix method [23,24].

Liquid crystal flow cells were fabricated using two glass ($n = 1.52$) microscope slides, bonded together by a thermally annealing plastic (Parafilm) to define channel walls. Typical flow channel dimensions are 4 cm long, 3 mm wide, and 70 μm thick. A homogeneous surface treatment (AL 1254) was spin coated to a depth of ≈ 100 nm on both glass plates and rubbed, using a velvet cloth, at an angle $\phi_0 \approx 87°$ to the x axis to promote bulk alignment through the cell [25] and a surface pretilt of $\theta_0 \approx 2°$. The two plates were constructed in either antiparallel alignment (to produce the parallel director profile) or parallel alignment (to produce the splayed director profile (see Fig. 1).

The flow cells are then connected to a Perkin Elmer syringe drive via 0.8 mm diameter stainless steel tubing (at the cell) and polypropylene hose, with the cell ends sealed using UV-curing glue. Cells were filled with 5CB via the stainless steel tubing with a 250 μL syringe and left to reach a uniformly aligned, nonflowing homogeneous state.

A conoscope was constructed, based on the design used by Parry-Jones *et al.* [26]. The samples were then placed at the convergence point of the incoming beam and the conoscope aligned to produce an interference figure focused at the CCD. The volumetric flow rate of the syringe drive was increased in steps of 2 μL/h over the range of 0 to ≈ 50 μL/h. Images of the interference figure were captured after each increment, allowing time for the system to reach a stable state. In order to accurately measure the angle of conoscopic interference figure rotation, a computer code was written to track the mirror plane of the interference figure. Minimizing the difference between the two halves of the figure for a straight line at a given angle to the figure's x axis allows for accurate determination of the rotation of the figure's mirror plane as a function of volumetric flow rate.

Figures 3 and 4 show modeled and observed conoscopic figure rotation for a cell aligned at $\phi_0 \approx 87°$ to the direction of flow with surface pretilt, in both the parallel and the splayed state. It is immediately apparent that there is a vast difference in the response of the two cells.

Most strikingly, Fig. 3 shows the average azimuthal distortion for a cell aligned at $\phi_0 \approx 87°$ to the direction of flow with the surface pretilt in the parallel state. It is seen from the modeling (Fig. 5) that the director rotates azimuthally in opposite directions in the top and bottom halves of the cell, with one half rotating towards $\phi = 0°$ and the other rotating towards $\phi = 180°$. The modeled tilt re-

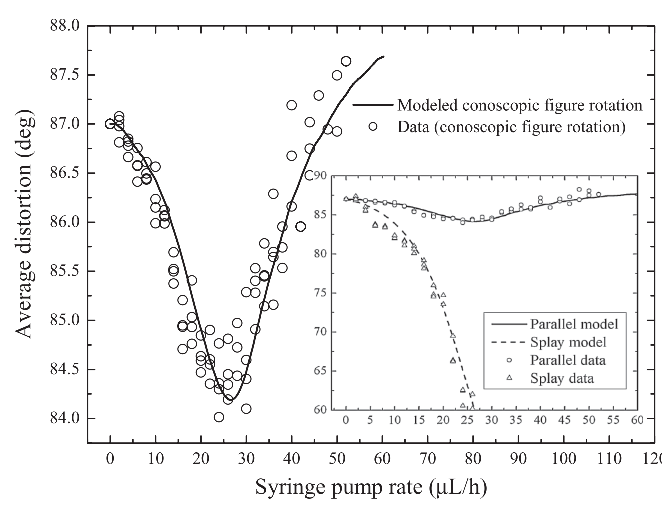

FIG. 3. Measured (symbols) and modeled (solid line) angle of conoscopic interference figure rotation for 5CB aligned with $\theta_0 \approx 2°$ (parallel) and $\phi_0 \approx 87°$ to the direction of pressure-driven flow. A striking optical response is seen to occur, with the conoscopic figure distorting through an azimuthal minimum due to asymmetric distortions about $z = d/2$. The inset highlights the difference in average azimuthal distortion between the parallel and splayed pretilt conditions.

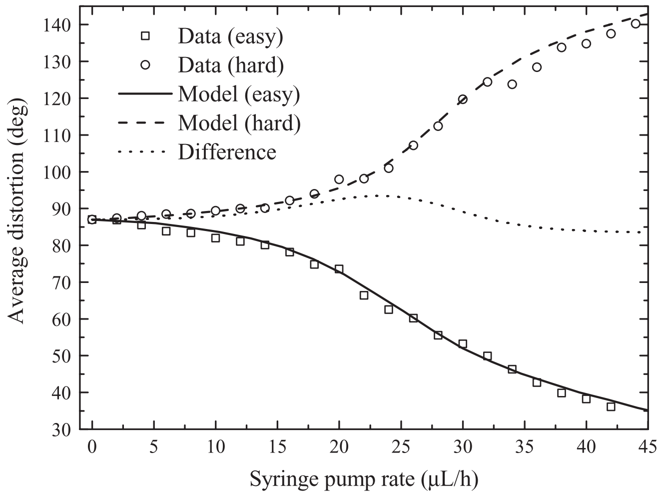

FIG. 4. Measured (symbols) and modeled (lines) angle of conoscopic interference figure rotation for 5CB aligned with $\theta_0 \approx 2°$ (splayed) and $\phi_0 \approx 87°$, for flow in the easy direction (solid line) and the hard direction (dashed line). The modulus of the difference is also plotted (dotted line) showing the small discrepancy in the bulk azimuthal distortion at low flow rates for opposite flow.

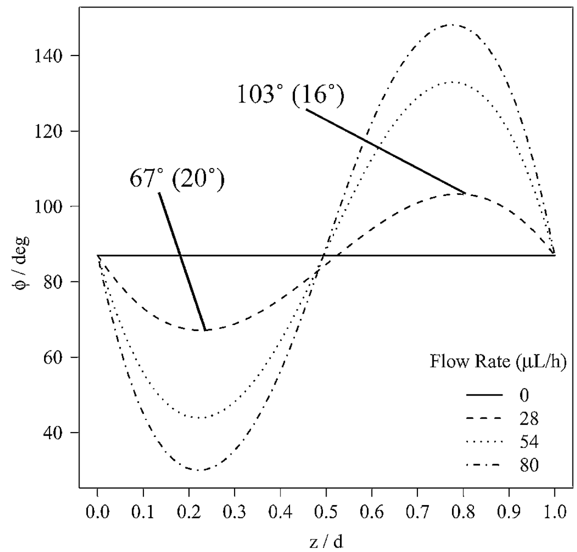

FIG. 5. Modeled azimuthal director distortion as a function of cell depth and flow velocity for a cell aligned at $\phi_0 = 87°$ in a parallel surface pretilt geometry. The asymmetry is shown for the lowest flow rate with an azimuthal distortion to 67° (a shift of 20° from $\phi_0 = 87°$) in the lower half of the cell and a maximum distortion to an azimuthal angle of 103° (a shift of 16° from $\phi_0 = 87°$) in the upper half.

sponse of the director is also shown [Fig. 6(a)]. Here we see that the response appears to be asymmetric at low flow rates, reaching a stable symmetric state at higher flow rates, but at a finite value ($\approx 10°$) away from the starting tilt distortion. This increase in the average tilt angle of the director is observed experimentally as a lateral translation of the conoscopic figure under flow.

Because of the surface anchoring and the small initial azimuthal rotation away from 90° ($\phi_0 \approx 87°$), the bulk azimuthal distortion is asymmetric about $z = d/2$, resulting in a shallow curve with a minimum to be observed in the mean rotation of the conoscopic figure of approximately 3° at a pump rate of approximately 27 μL/h. The director can be thought of as azimuthally distorting in one (negative) direction in more than one half of the cell, while the remainder of the cell distorts azimuthally in the opposite (positive) direction by a lesser amount. This difference creates a small but observable overall azimuthal distortion in the negative direction (the initial dip in conoscopic figure rotation seen in Fig. 3). At increased flow rates this effect is reversed, with more than half of the cell azimuthally distorting in the positive direction and the remainder of the cell distorting in the negative direction, overall increasing the average azimuthal distortion and creating the turning point and increase in the angle of conoscopic figure rotation seen in Fig. 3. The inset of Fig. 3 shows how dramatically the overall director profile, and consequently the optical response of the cell, is changed by the presence of a small surface pretilt aligned in the parallel state compared with pretilt in the splay state (described below).

Figure 4 shows the conoscopic figure distortion for two flow directions (parallel to x) for the splayed cell, where the first direction is defined to be "easy" with the splay surface anchoring aligned such that boundary (and bulk) azimuthal distortion is already twisted into the direction of flow, and the second "hard" direction where the average azimuthal distortion is initially aligned twisted against the direction of flow. Both flow directions exhibit similar responses (with the exception that the overall average director rotation is in the opposite direction) with a small difference that arises from the initial azimuthal director alignment ($\phi_0 \approx 87°$) relative to flow. In the case of the easy flow direction the bulk azimuthal angle is already aligned in the direction of flow, due to the combination

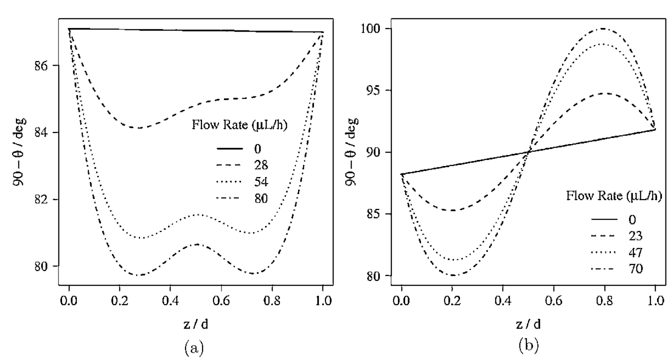

FIG. 6. Modeled director tilt profiles for both the (a) parallel and (b) splayed starting director profiles. Both graphs show how the director tilt angle evolves as the flow rate is increased as a function of cell depth.

of surface pretilt anchoring and rubbing direction. In the case of the hard flow direction, the same condition creates initial bulk azimuthal alignment opposite to the flow direction. Figure 6(b) shows the modeled tilt profile for the splayed cell flowing in the easy direction. As can be seen, tilt distortions about $z = d/2$ cancel exactly, creating no net tilt in the sample under flow, and hence no lateral translation of the conoscopic interference figure. The resulting distortion appears instead, as a rotation of the figure, which is in turn proportional to the average azimuthal director distortion in the cell.

In summary, we have demonstrated that for pressure-driven flow of the nematic liquid crystal 5CB, the presence of a small surface pretilt, which may lead to different symmetries of the static director profile, can greatly affect the director profile in the bulk of the sample when the initial azimuthal alignment is not normal to the flow direction. For only a few degrees surface pretilt in the splayed state, the magnitude of director distortion as a function of volumetric flow rate is seen to be similar for forward and backward flows (particularly at low flow rates), while the same degree of pretilt in the parallel state can produce entirely opposite azimuthal distortions of the director about $z = d/2$ and hence entirely different averaged optical properties. For the splayed state, we observe no net tilting of the director, yet a positive net azimuthal distortion, whereas for the parallel state, we observe a net tilt distortion but no net azimuthal distortion. This difference results in completely distinct optical conoscopic figures, whereby one (splayed state) shows a rotation and the other (parallel state) shows a lateral translation and small rotation minimum. The conoscopic data agree well with the theoretical figures simulated by a simplified, one-dimensional model solving the Ericksen-Leslie equations to produce director profiles under flow. These results highlight the extreme sensitivity of the flowing director orientation to only a few degrees of surface pretilt.

The authors would like to thank the EPSRC and HP for funding this work, along with Nick Cole for technical assistance.

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