Orientational transition in nematic liquid crystals under oscillatory Poiseuille flow

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Abstract

We investigate the orientational behaviour of a homeotropically aligned nematic liquid crystal subjected to an oscillatory plane Poiseuille flow produced by an alternating pressure gradient. For small pressure amplitudes the director oscillates within the flow plane around the initial homeotropic position, whereas for higher amplitudes a spatially homogeneous transition to out-of-plane director motion was observed for the first time. The orientational transition was found to be supercritical and the measured frequency dependence of the critical pressure amplitude in the range between 2 and 20 Hz was in quantitative agreement with a recent theory.

1 Introduction

In nematic liquid crystals (NLCs) complex behaviour arises from the strong coupling between velocity \( \mathbf{v} \) and director \( \mathbf{n} \). For a steady flow along the \( x \) axis with a velocity field \( v_x(z) \), \( v_y = v_z = 0 \) (rectilinear plane flow, typically of the Couette or Poiseuille type) the director will, in the absence of other torques, align in the flow plane \((x-z)\) plane) at the angle \( \theta_{fl} = \pm \arctg(\sqrt{\alpha_3/\alpha_2}) \) with the \( x \) axis if \( \alpha_3/\alpha_2 > 0 \) (the \( \pm \) sign corresponds to positive/negative shear rate \( \partial v_x/\partial z \)) [1, 2]. In common nematics \( \alpha_3/\alpha_2 \) is small but positive \((\approx 0.01)\), however, in some materials (in particular near a nematic-smectic transition) one has \( \alpha_3/\alpha_2 < 0 \) and...
instead of flow alignment there is more complicated tumbling motion [3-7]. Note that \(\alpha_2\) is always negative, so the sign of \(\alpha_3/\alpha_2\) is determined by that of \(\alpha_3\).

When the velocity field oscillates periodically and symmetrically around zero [time average \(\langle v_x(z,t) \rangle = 0\)] the situation becomes more complicated. Here we are concerned with oscillatory Poiseuille flow where the velocity field \(v_x(z,t)\) is induced by applying an alternating pressure difference \(\Delta P \cos(\omega t)\) between the ends of a plane cell filled by a NLC (bounding plates parallel to the \(x - y\) plane). The planarly aligned case with the director oriented perpendicular to the flow plane, i.e., in the \(y\) direction, has been studied intensively in experiments by Pieranski and Guyon [8, 9] and theoretically by Dubois-Violette and Manneville [10, 11]. For very low frequencies a spatially homogeneous instability at some critical pressure amplitude has been found whereas for higher frequencies a transition to periodic rolls oriented along the flow direction was observed [8, 9].

When the director is prealigned within the flow plane the behaviour under oscillatory Poiseuille flow can be quite complex and has not yet been clarified completely. In the high-Ericksen number limit where elasticities (and therefore also boundaries) are neglected the time-averaged torque is zero and one has reversible behaviour (no attractors). Thus one expects the existence of a two-parameter family of solutions for the director oscillations depending on the initial orientation. When the effect of elasticities, which represent singular perturbations, is included and the shear rate is spatially nonhomogeneous (as is the case for Poiseuille, but not for Couette flow), there arises an averaged torque, which produces attractors [12]. It is then found that, without surface anchoring, the flow-alignment solution is always unstable and one is left with one major attractor, which for \(\alpha_3/\alpha_2 < 0\) is the orientation perpendicular to the flow plane, or, for \(\alpha_3/\alpha_2 > 0\), oscillations around an orientation close to it (in the \(y - z\) plane). In this case also the perpendicular orientation becomes unstable which is consistent with the results of [8, 9, 10, 11]. In both cases, the oscillations in the flow plane around the homeotropic direction are unstable. Including boundary effects one in fact expects a threshold for an out-of-pane transition [12].

In this letter we give the first experimental confirmation of this effect. We also present new, rigorous numerical calculations of the threshold as a function of frequency and compare with the experimental results.

## 2 Experimental setup

Experiments were carried out with the nematic MBBA \([\text{N-(4-methoxybenzylidene)-4-butylaniline}]\), a flow aligning material \((\alpha_3, \alpha_2 < 0)\). The nematic was confined between two 3 mm thick glass plates of lateral dimensions \(L_x = 15\) mm \(\times\) \(L_y = 20\) mm. The thickness \(d\) of the nematic layer was controlled by two polyester-foil spacers placed along the shorter
sides at \( y = \pm L_y/2 \) (nominal thickness 23 \( \mu \)m). They also served to close the flow cell along these sides. The other sides were open. The nematic was held inside the cell by capillary forces. To induce homeotropic alignment of the director at the glass plates the inner surfaces were treated with the surfactant DMOAP [dimethyloctadecyl-(3-trimethoxysilyl)-propylammonium chloride].

The setup surrounding the cell was designed to apply an oscillatory air pressure difference \( P_{1,2} = \Delta P_{1,2} \cos(\omega t + \varphi_{1,2}) \) to the two open sides of the cell (fig. 1). The cell was mounted inside a brass block of outer dimensions \( 4 \times 5 \times 10 \) cm. The open sides of the cell were in contact with channels in the brass block which widened towards the cell in order to gain the width \( L_y \). As a result the pressure was distributed rather evenly over the entire width of the cell. The channels connected, via cavities of adjustable volume, to two vertical cylinders accommodating pistons driven sinusoidally in opposing sense by an electromotor. The displacement was \( v_d = 5 \) cm\(^3\) and the frequency \( f = \omega/(2\pi) \) ranged from 1 to 20 Hz. The volume of the cavities could be adjusted by screws in order to regulate the “passive” volume \( V_i \) of air between the lower position of the pistons and the cell. This allowed to vary the amplitude \( \Delta P \) of the applied pressure in a range from 1 to 20 kPa. Pressure sensors were connected to the open sides of the cell in order to measure the pressure head with respect to the surrounding air.

The response of the nematic was detected optically by exploiting its birefringence. We used a light source with maximum intensity at wavelength 610 nm (cold source with filter) and crossed polars (polariser \( \| \hat{e}_x \), analyser \( \| \hat{e}_y \)). The transmitted light intensity was measured by a semiconductor detector, either integrating over an area of 28.3 mm\(^2\) or with a spatial resolution of 2 \( \mu \)m (long-range microscope). The photo detector in the microscope could be replaced by a CCD camera. Interfaces for the signal processing were either an A/D board or a frame grabber with a resolution of 512 \( \times \) 512 pixels and 256 grey scales. The temperature of the brass block was kept at 25.0 \( \pm \) 0.1°C by a water thermostat.

Due to the large aspect ratio of the flow cell \( (L_y/d \approx 10^3) \) one may expect rectilinear Poiseuille flow \( (v_y = v_z = 0) \) except in thin boundary layers near the edges. In order to test the flow field and to calibrate the pressure sensors the motion of Al\(_2\)O\(_3\) tracers (diameter 5 \( \mu \)m) was recorded with the CCD camera and analysed. No \( y \) component of the flow was detected.

An alternating pressure applied to a fluid layer decays by compressibility effects over a length \( \delta = d\sqrt{2\rho c^2/(\pi^2\omega \eta)} \) [14, 15]. Here \( \rho \) is the mass density and \( c \) is the speed of sound. The viscosity for the homeotropic orientation is given by \( \eta = (-\alpha_2 + \alpha_4 + \alpha_5)/2 \). Using MBBA material parameters \( (\rho = 10^3 \text{ kg/m}^3, \ c = 1.5 \cdot 10^3 \text{ m/s} \) [16], \( \eta = 135.4 \cdot 10^{-3} \text{ N\cdot s/m}^2 \)), cell thickness \( d = 23 \) \( \mu \)m, and the frequency \( f = 20 \) Hz one obtains \( \delta \approx 12 \) cm which is considerably larger than the cell length \( L_x = 15 \) mm. Moreover, since in our setup the
pressure difference is applied symmetrically at both sides of the capillary the flow is expected to be practically homogeneous along the $x$ direction. This was also verified by tracing the Al$_2$O$_3$ particles.

3 Experimental results and comparison with theory

Let us define the director components as $\mathbf{n} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$ with the polar angle $\theta$ measured with respect to the $z$ axis and azimuthal angle $\phi$ measured from the $x$ axis. For amplitudes $\Delta P$ below any instability the director is expected to oscillate within the flow plane, i.e. $\phi = 0$, and in our geometry of crossed polars the intensity should be minimal (and constant in time). As a result of an out-of-plane transition the intensity should increase except when $\phi = \pi/2$ and $\phi = \pi$. In fig. 2 we show examples of the signals of the two pressure sensors together with the signals of the transmitted light intensity measured by the photo diode as functions of time for an amplitude below and above threshold. We have $\varphi_2 = \varphi_1 + \pi$ and $\Delta P_1 = \Delta P_2 = \Delta P/2$.

The signal of the light intensity in fig. 2b exhibits two rather sharp double peaks over the oscillatory flow period. We interpret the minimum as the (rapid) passage of the director through $\phi = \pi/2$. The long low-intensity interval between the peaks then results from $\phi$ approaching 0 and $\pi$ while the director reverses its motion (trajectories of the director motion are shown in ref.[12]). Slightly above the out-of-plane instability threshold one obtains for the azimuthal angle $\phi$ the following expression [12]

$$\tan \phi = \chi/[8a(z/d) \sin \omega t],$$

where $\chi (\ll 1)$ is the out-of-plane angle between the director and flow plane ($x-z$ plane) when the director passes through $\phi = \pi/2$ and $a = A/d$ is the dimensionless flow displacement amplitude at midplane. Clearly $|\phi|$ is maximal at the cell boundaries $z = \pm d/2$ and the leading contribution to the transmitted light intensity comes from there (for $\chi \ll 1$). This gives an approximate time dependence of the transmitted light intensity

$$I_{Tr} \sim \sin^2[2\phi(z = d/2)] \sim \left[ \frac{\chi^4a \sin \omega t}{\chi^2 + (4a \sin \omega t)^2} \right]^2.$$  

Equation (2) describes the observed signal of the light intensity (fig. 2b) and agrees with the results of full simulations of the optical response based on the numerical solution of Maxwell’s equations with the director distribution calculated from the nematodynamic equations (see below). In principle such type of signal could also be caused by an elliptical motion of the director without breaking the left-right symmetry. We have excluded this possibility by measurements with the analyser rotating with the driving frequency.
Since the out-of-plane transition involves the breaking of a two-fold symmetry one expects the appearance of patches of the two states. Indeed, under the microscope we always observed domain boundaries indicating patches of $\sim 0.1 - 1 \text{mm}^2$ area (fig. 3). After some initial coarsening they did not change much over times of several minutes, except for oscillatory motion of the boundaries with the flow field (see the bottom part of fig. 3).

In order to obtain quantitative results for the threshold we determined the average intensity of the transmitted light $\langle I_{Tr}(t) \rangle$. In fig. 4a $\langle I_{Tr}(t) \rangle$ is shown versus the pressure amplitude for a frequency $f = 10 \text{Hz}$. By integrating eq.(2) one finds $\langle I_{Tr}(t) \rangle \sim \chi$. Since one expects $\chi \sim \sqrt{\Delta P - \Delta P_c}$ we have fitted the measured values of $\langle I_{Tr} \rangle$ accordingly. The best fit curve $\langle I_{Tr} \rangle = 16.4\sqrt{\Delta P - 7.3}$ is shown in fig. 4a (solid). Clearly the out-of-plane motion sets in at a well-defined threshold $\Delta P_c$ via a continuous (forward) bifurcation (no hysteresis), as predicted by theory [12].

Direct numerical simulations of the standard set of nematodynamic equations [17] where the director $\hat{n}$ and velocity $v$ are functions only of the distance $z$ from the boundaries and time $t$ (see [13] for details) were performed using finite differences for the spatial derivatives and the predictor-corrector scheme for the time discretisation. The critical amplitude of the pressure gradient $\Delta P_c(f)/L_x$ was calculated for the frequency range $0.5 \text{Hz} \leq f \leq 20 \text{Hz}$, layer thickness $d = 23 \mu\text{m}$ and MBBA material parameters at $25^\circ\text{C}$ [18, 19] (elastic constants in units of $10^{-12}\text{N}$: $K_{11} = 6.66$, $K_{22} = 4.2$, and $K_{33} = 8.61$; viscosity coefficients in units of $10^{-3}\text{N}\cdot\text{s/m}^2$: $\alpha_1 = -18.1$, $\alpha_2 = -110.4$, $\alpha_3 = -1.1$, $\alpha_4 = 82.6$, $\alpha_5 = 77.9$, $\alpha_6 = -33.6$; mass density $\rho = 10^3\text{kg/m}^3$). The results of the numerical computations together with the measured values of $\Delta P_c(f)$ are shown in fig. 4b. The agreement of the measured values with the calculated ones is quite good (no adjustable parameters). Discrepancies near the ends of the frequency interval can be explained by deviations from the sinusoidal driving in these regions. One notices that for not too low frequencies ($\omega \tau_d \gg 1$, where $\tau_d$ is the director relaxation time) $\Delta P_c$ rises linearly with $\omega$. This corresponds to the critical Poiseuille flow amplitude $a_c$ being independent of $\omega$, which can be understood from simple scaling arguments [12]. Quantitative discrepancies with our earlier theoretical work results from the fact that there the effect of the director distribution on the flow field, which leads to deviations from the parabolic profile, was not included.

4 Conclusion

For the first time the out-of-plane orientational transition in homeotropically oriented nematic liquid crystal under oscillatory Poiseuille flow was observed experimentally. The phenomenon is interesting because the underlying torque on the director resulting from spatially inhomogeneous driving in the presence of diffusive coupling, arises quite unexpectedly.
The analysis of the transmitted light intensity shows that the instability has a supercritical character without hysteresis. The frequency dependence of the critical pressure amplitude is in a good quantitative agreement with theoretical results without any adjustable parameters. Further efforts will be made to increase the range of frequency and pressure of the measurements. The theory predicts that at high driving amplitudes the director saturates at an orientation perpendicular to the flow plane ($\alpha_3 > 0$) or near to it ($\alpha_3 > 0$) unless a further transition to a spatially periodic state intervenes.

Let us finally mention that a more complicated scenario is expected for boundary anchoring corresponding to the flow-alignment angle or, alternatively, simple planar anchoring of the director with an additional magnetic field in the flow plane at an angle of 45° with respect to the x axis. Then a transition to a slow time-periodic director precession has been predicted theoretically. This director motion emerges with increasing flow amplitude through a homoclinic bifurcation and disappears through a Hopf bifurcation. It would be interesting to perform experiments to test this prediction.

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References

[1] Leslie F. M., *Arch. ration. Mech. Anal.*, 28 (1968) 265.
[2] Pikin S. A., *Zh. Eksp. Teor. Fiz.*, 65 (1973) 2495 [Sov. Phys. JETP, 38 (1974) 1246].
[3] Cladis P. E. and Torza S., *Phys. Rev. Lett.*, 35 (1975) 1283.
[4] Pieranski P. and Guyon E., *Commun. Phys.*, 1 (1976) 45.
[5] Pieranski P., Guyon E. and Pikin S. A., *J. Phys. (Paris) Colloq.*, 37 (1976) C1-3.
[6] Manneville P., *Mol. Cryst. Liq. Cryst.*, 70 (1981) 223.
[7] Zúñiga I. and Leslie F. M., *Europhys. Lett.*, 9 (1989) 689; *Liq. Cryst.*, 5 (1989) 725.
[8] Guyon E. and Pieranski P., *J. Phys. (Paris) Colloq.*, 36 (1975) C1-203.
[9] Jánossy I., Pieranski P. and Guyon E., *J. Phys. (Paris)*, 37 (1976) 1105.
[10] Manneville P. and Dubois-Violette E., *J. Phys. (Paris)*, 37 (1976) 1115.

[11] Manneville P., *J. Phys. (Paris)*, 40 (1979) 713.

[12] Krekhov A. P. and Kramer L., *J. Phys. II (Paris)*, 4 (1994) 677.

[13] Krekhov A. P. and Kramer L., *Phys. Rev. E*, 53 (1996) 4925.

[14] Landau L. D. and Lifshitz E. M., *Course of Theoretical Physics: Hydrodynamic*, Vol. 6 (Pergamon Press, New York) 1986.

[15] Blinov L. M., Davidyan S. A., Reshetov V. N., Subachyus D. B. and Yablonshi S. V., *Zh. Eksp. Teor. Fiz.*, 97 (1990) 1597 [*Sov. Phys. JETP*, 70 (1990) 902].

[16] Eden D., Garland C. W. and Williamson R. C., *J. Chem. Phys.*, 50 (1973) 1861.

[17] de Gennes P. G. and Prost J., *The Physics of Liquid Crystals*, (Clarendon, Oxford) 1993.

[18] de Jeu W. H., Claassen W. A. P. and Spruijt A. M. J., *Mol. Cryst. Liq. Cryst.*, 37 (1976) 269.

[19] Knappe H., Scheider F. and Sharma N. K., *J. Chem. Phys.*, 77 (1982) 3203.
Figure 1: Cross section of the experimental setup.
Figure 2: Signal of the pressure sensors (dashed and dot-dashed lines) and transmitted light intensity (solid line) for a subcritical pressure (a) and a supercritical pressure (b).
Figure 3: Snapshot of patches with left- and right-side out-of-plane director configurations (upper part) and $x - t$ plot showing time oscillations of domain boundaries (lower part) far from threshold. The crossed polars were rotated slightly away from the symmetric orientation to obtain different intensities in the two configurations. The dark vertical stripes pertain to dust particles on the glass plates.
Figure 4: a) Transmitted light intensity averaged over the flow period for increasing (⊙) and decreasing (△) pressure amplitude ($f = 10$ Hz). b) Frequency dependence of the critical pressure amplitude $\Delta P_c$ for out-of-plane transition: experimental (squares) and calculated (solid line).