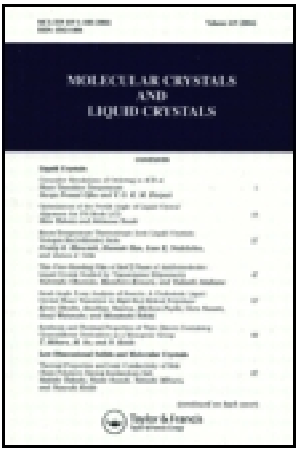

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Observations of Non-Steady Poiseuille Flow of the Liquid Crystal 8CB

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We report observations of non-steady Poiseuille flow of the liquid crystal 8CB in a radial Hele-Shaw cell. Air was injected into the cell via a syringe with a mass attached to the plunger. In both the nematic and smectic phases, when viewing the cell between crossed polarizers, persistent periodic variations of the intensity of transmitted light were observed. These observations are consistent with abrupt changes in the viscosity of the liquid crystal due to changes in director configuration brought about by changes in flow velocity.

In liquid crystals undergoing shear, the orientation of molecules is coupled to the macroscopic velocity gradients. The nematic director $\hat{n}$, which denotes the direction of the average local molecular alignment, experiences viscous torques due to the velocity gradients; and a local rotation of $\hat{n}$ can also give rise to flow. A continuum model describing the hydrodynamics of nematic liquid crystals has been given by Ericksen and Leslie.[1] Subsequently, the flow alignment of nematics has been studied in considerable detail.[2-3] In general, the director configuration depends on both viscous and elastic torques, and the response of a sample is determined by the elastic constants and the Leslie viscosity coefficients.

In materials with Leslie coefficients $\alpha_3 > 0$ and $\alpha_2 < 0$, discontinuous director field transitions called "tumbling" can occur. In this case for simple shear flow, Carlsson[4] calculated that the apparent viscosity of the flowing liquid crystal should change abruptly as the flow velocity or the shear force is monotonically increased. Experimentally, Cladis and Torza[5] found discontinuous director changes in Couette flow when the flow velocity was increased continuously. In the case of materials where $\alpha_3 < 0$, shear flow was expected to give rise to stable director alignment.[4] It has been shown[6] recently that even if $\alpha_3 < 0$, for sufficiently large shear, a large number of distinct equilibrium director configurations exist, and it has been proposed that propagating excitations that have been observed in shear flow are fronts between states with such configurations.[6]

In this paper we report our observations of non-steady flow of the liquid crystal 8CB (4,4′-*n*-octylcyanobiphenyl) in a radial Hele-Shaw cell. The cell consisted of

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the liquid crystal sample between two glass plates with dimensions 15 cm × 15 cm × 3 mm separated by 15 μm teflon spacers. The sample temperature was controlled to within ±0.5°C by illuminating the sample from above with IR light. Air was injected into the center of the cell via a syringe with cross-sectional area 1.6 cm.^{2} The barrel of the syringe was vertical, and a mass was attached to the plunger. The cell, between crossed polarizers, was illuminated with white light from below. When the plunger was released, the air injected into the cell displaced the liquid crystal, and the patterns due to the advancing air–liquid crystal interface and due to the flow alignment of the liquid crystal were recorded using a video camera and recorder. By focusing the image of the entire monitor screen onto a photodetector using a lens, we measured the total transmitted intensity over the entire cell as a function of time.

The air–liquid crystal interface is unstable, and, instead of a circular bubble, smoothly evolving dense-branching or dendritic morphologies are observed.[7] Away from the interface, because of its large viscosity, steady Poiseuille flow of the liquid crystal with an essentially parabolic velocity profile might be expected. Instead of this expected smooth flow with steady average velocity, in a certain pressure interval, the liquid crystal exhibited non-steady flow consisting of long and nearly periodic sequences of jerks. These jerks could be seen as variations in the intensity of light transmitted by the sample, and could be observed both in the nematic and in the smectic A phases.

In the smectic A phase, except for a thin (less than 0.5 mm) region at the air–liquid crystal interface, the alignment of the liquid crystal in the cell was homeotropic with the director perpendicular to the glass walls. When the envelope of the advancing interface reached a critical diameter (typically ~1.5 cm), a flash propagating from the center of cell could be seen. The appearance of the flash is assumed to be due to a disturbance of the director field and the resulting birefringence of the sample between crossed polarizers. The size of the bright region increased rapidly, until its diameter was approximately 20% that of the interface envelope, then decreased as the alignment relaxed back to the homeotropic state. The air–liquid crystal interface advanced continuously during this time, and no clear correlation between the flash and motion of the interface could be discerned. After a few seconds, the flash was repeated, but over a larger region. Provided that the pressure was in the range 10–17 KPa, the above process repeated several times with gradually increasing size of the bright spot and decreasing time between flashes. Eventually the time between flashes is less than the relaxation time of the director alignment, and the entire sample remains birefringent. At this time, the envelope of the interface typically encloses about 2/3 of the cell area.

The time variation of the total light intensity transmitted by the cell is shown in Figure 1a. The build-up time of the flash is very short, ~0.1 s, but its relaxation takes longer than 1 s. For pressures below 9 KPa, the velocity of the air-liquid crystal interface was close to zero, and no flashes could be seen. For pressures above 25 KPa, the interface velocity is so large that only a single continuous flash can be seen. The pressure dependence of the elapsed time between successive flashes, and the transmitted light intensity are plotted in Figure 1b. The temperature dependence of the elapsed time between the first and second observable flashes

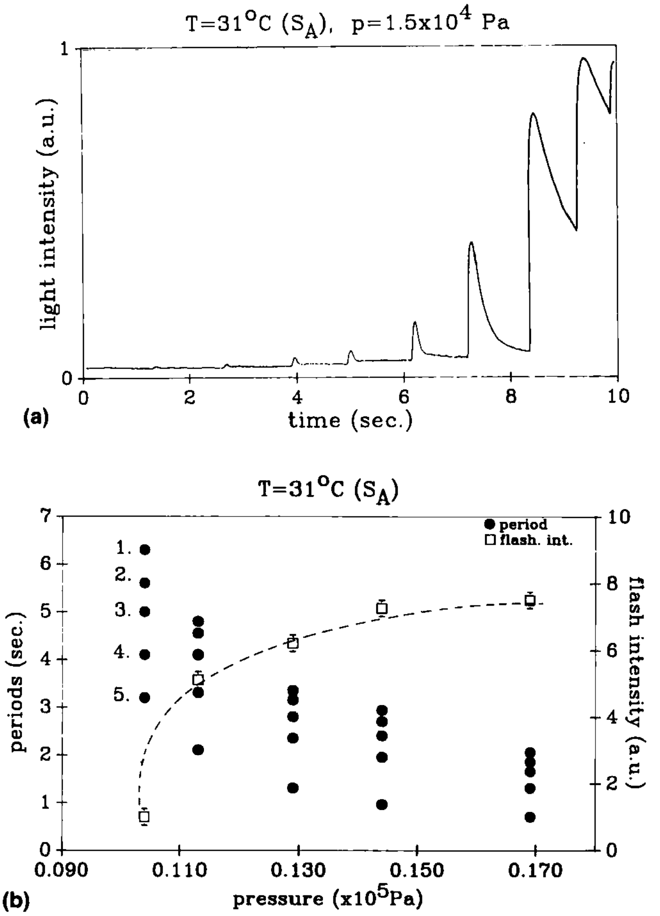

FIGURE 1 (a) Total transmitted light intensity in the smectic A phase as a function of time. (b) Period between flashes (circles) and flash intensity (squares) as function of applied pressure. The circles in the vertical columns at a given pressure refer to successive flashes. The time elapsed after the first flash has the longest period, as the numbering on the left indicates. The intensity is given for the fifth flash at each pressure. The dashed line is to guide the eye.

at the pressure of 15 KPa is shown in Figure 2. On cooling the sample below the nematic–smectic A transition temperature, the period first increases, showing a maximum at T = 29°C, and then decreases to a plateau below T = 27°C.

The flow behaviour in the nematic phase also showed non-steady behaviour, however, some details were different. Flashes were again observed, as shown in Figure 3a, but these could be correlated with a non-steady, jerky advance of the air-liquid crystal interface. In one cycle, the interface velocity could be seen to increase, then the flash would appear, followed by an abrupt slowing down of the interface. This would be followed by a period of gradual acceleration, terminated by a new flash and jerk. Although the amplitude of this jerky motion was very small, it was clearly visible when viewing the video image on the monitor. During the jerk, the color of the transmitted light in front of the interface also abruptly

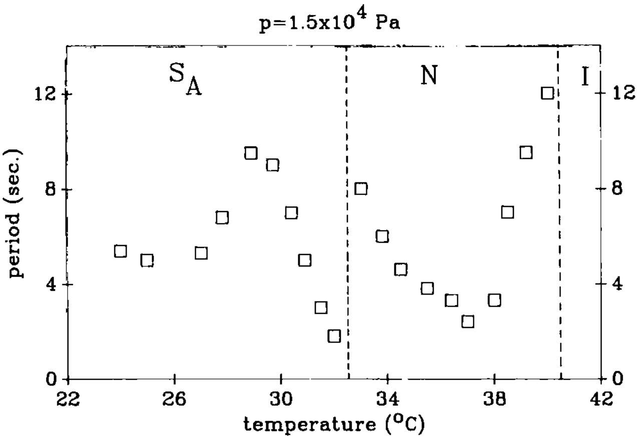

FIGURE 2 Period between flashes as function of temperature.

changed. Frequently, after the jerk, tip-splitting of the interface could be observed. As in the smectic case, the periods between the flashes were in the range of several seconds. Unlike in the smectic case, however, the period in the nematic phase is very nearly constant. The temperature and pressure dependence of the average period is plotted in Figure 2 and Figure 3b. The periodic pulsating flow behaviour was observed in the entire nematic range, but close to both nematic–isotropic and nematic–smectic A transitions, the period increased considerably. The minimum period occurred at T = 37°C; the pressure dependence of the average period is shown at this temperature in Figure 3b. At pressures below 7 KPa no appreciable flow could be induced, while above 20 KPa, only one jerk could be seen.

Although the details of the behaviour in the nematic and smectic phases differ, it seems likely that the observed non-steady flow is associated with transitions between states with different director configurations. In nematics such transitions are already described,[2–6] but similar behaviour has not yet been observed in smectic A phases.

As air is injected into the cell at constant pressure and the air–liquid crystal interface advances, some liquid crystal flows out of the cell at its outer edges. The magnitude of the pressure gradient in the radial direction therefore increases with time, and so does the flow velocity of the liquid crystal. Since the apparent viscosity depends on the director configuration which depends on the shear, the apparent viscosity of the sample is expected to change in time.

Assuming a parabolic velocity profile, and assuming that the director remains in the shear plane defined by the velocity and the velocity gradient, in the one elastic constant approximation the torque balance equation for the director is

$$\frac{\partial \theta^2}{\partial z^2} = \eta\, z \left(\cos^2\theta - \frac{\alpha_3}{\gamma_2}\right) \qquad (1)$$

where θ is the angle between the director and the normal to the plates, and z is the coordinate along the plate normal, measured in units of the plate separation

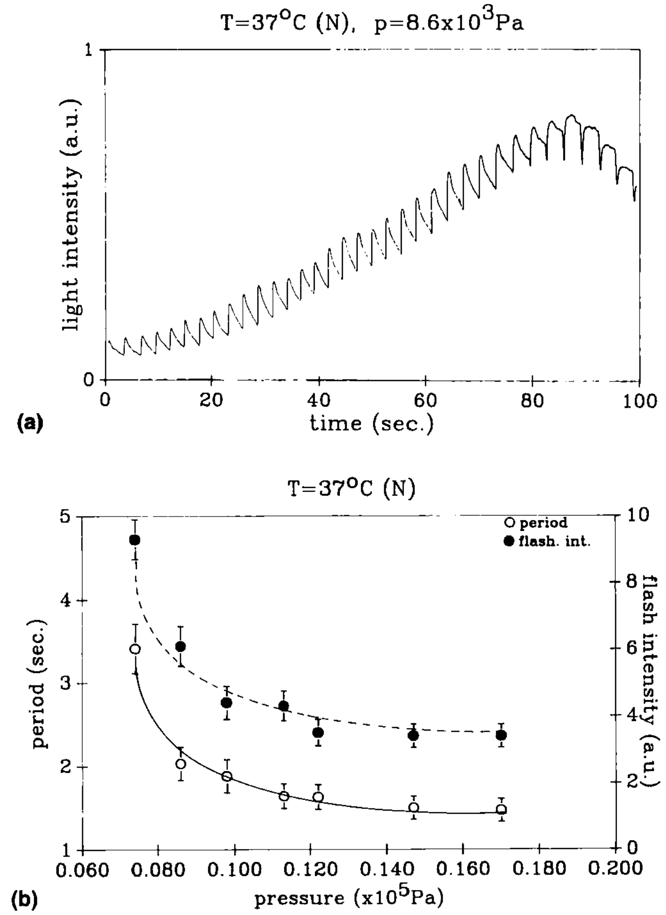

FIGURE 3 (a) Total transmitted intensity in the nematic phase as a function of time. (b) Average period (open circles) and flash intensity (solid circles) as function of applied pressure. The solid and dashed lines are best fits to the experimental data.

d. $\gamma_2 = \alpha_2 + \alpha_3$ where α_2 and α_3 are Leslie viscosity coefficients. If v is the average velocity (averaged in the z direction) and K is the curvature elastic constant, $\eta = 12vd\gamma_2/K$ is the Ericksen number and is a measure of the contribution of viscous torques due to shear relative to elastic torques due to curvature of the director field. We have solved Equation (1) numerically using the shooting method, subject to the boundary condition $\theta(-\frac{1}{2}) = \theta(\frac{1}{2}) = 0$, and found that when η reaches the threshold value η_c, two distinct director configurations can satisfy the equation. Increasing η increases the number of solutions; this behaviour was found in the entire nematic range independent of whether α_3 was negative or positive. Using values of the elastic constants and viscosity coefficients from the literature,[8,9] we find that $K \simeq 10^{-11}$ N and $\gamma_2 \simeq 5 \times 10^{-2}$ Pa s over the whole nematic range. We estimate $v = 1$ mm/s from the motion of the interface, and since $d = 15$ μm, we find that typically $\eta \approx 1000$ in our experiment. On calculating η_c for different temperatures we find that $\eta_c(T = 36°) = 383$ and $\eta_c(T = 38°C) = 475$; and therefore in our experiment the system was in the multiple solution regime.[10]

We have also calculated the apparent sample viscosities $\langle\mu\rangle$ corresponding to the different configurations. For $\langle\mu\rangle$ we have[7]

$$\langle\mu\rangle = \int_{-1/2}^{1/2} (\eta_2 - \gamma_2 \cos^2\theta)dz \tag{2}$$

where η_2 is one of the Miesowicz viscosities. Using our two solutions for $\theta(z)$ at $\eta = \eta_c$, we found that the corresponding apparent viscosities varied appreciably, by approximately 20%. For larger values of η, larger differences in apparent viscosity are possible.

As the pressure is applied and the flow velocity increases sufficiently, in the region of the greatest speed near the interface η increases beyond η_c. We expect that at this time, a new high viscosity state propagates from the interface into the less stable low viscosity state. This increases the apparent viscosity, leads to a decrease in the flow velocity and η, and to the subsequent vanishing of the new solution. This process may result in the observed oscillatory motion, provided that there is inertia in the system, which allows the pressure to overshoot its purely viscous value. We believe that the mass attached to the plunger of the syringe used to inject air into the cell provides most of this inertia.

The plunger of the syringe with the attached mass m, the air in the barrel, and the liquid crystal in the sample cell with velocity and time dependent viscosity form a nonlinear oscillator. This can be modelled by a spring with force mg acting on one end, and a nonlinear friction force acting on the other. If $\eta < \eta_c$, the system behaves as a damped harmonic oscillator with natural frequency $\omega_0 = 50\ s^{-1}$ and damping coefficient $\beta = 4\ s^{-1}$. When the plunger is first released, the air-liquid crystal interface oscillates with period $T_0 = 0.12$ s as it advances. This oscillation is rapidly damped, and after the first second the pressure acting on the liquid crystal is constant. During this stage the interface advances with an average velocity given approximately by

$$v \simeq \frac{mgd}{12\langle\mu\rangle A(t)} \tag{3}$$

where $A(t)$ is the area of cell occupied by the liquid crystal. Since $A(t)$ is monotonically decreasing in time, v is increasing. When $\eta = 12vd\gamma_2/K$ reaches the value η_c, at $t = 0$, a new solution can invade the cell and the apparent viscosity increases. If this increase is linear in time, then

$$\langle\mu(t)\rangle = \langle\mu_1\rangle + \frac{t}{\tau}(\langle\mu_2\rangle - \langle\mu_1\rangle) \tag{4}$$

where $\langle\mu_1\rangle$ and $\langle\mu_2\rangle$ are the apparent viscosities of the two states. τ is the build-up time of the new solution, and although in principle this could be calculated approximately,[6] from the observed increase in flash intensity (see Figure 3a) we estimate here that it is of the order of 0.1 s. Solving the resulting equations of motion, we find that after reaching η_c, the dimensionless velocity η further increases by an amount $\Delta\eta\uparrow$ during the time period $0 < t < T_0/4$, then drops below η_c as $\langle\mu\rangle$ decreases towards $\langle\mu_1\rangle$. η decreases below η_c by an amount $\Delta\eta\downarrow$ ($\Delta\eta\downarrow > \Delta\eta\uparrow$), and after small oscillations with angular frequency ω_0 which decay, the velocity increases again as given by Equation (3). The physical interpretation of this solution is as follows. As the viscosity increases, the air in the syringe is compressed, and causes the velocity of the liquid crystal to exceed its critical value. This takes place in a time interval $T_0/2$. When the air subsequently expands, the velocity decreases, falls well below its critical value and the viscosity decreases. After a few decaying oscillations which do not increase the velocity to its critical value, the velocity increases slowly as more material is pushed out of the cell. When the critical velocity is again reached, the process is repeated. It is likely that the role of the interface is to facilitate the formation of the new state.

In order to verify that the inertia of the air injection system plays a role in determining the response, we studied the effect of a smaller injecting system, where the barrel cross sectional area and mass were both 15 times smaller than in the original system. With this syringe, we also observed oscillations, but with reduced intensity and frequency. Finally, injecting nitrogen into the cell via a pressure regulator produced no oscillations at all.

We have observed novel non-steady pulsating flow in the nematic and smectic phases of the liquid crystal 8CB in a radial Hele-Shaw cell. The responsible mechanism is likely the velocity dependent apparent viscosity of the liquid crystal; however, the properties of the injection system play a role in determining the dynamics of the response. We propose that the velocity dependence of the viscosity of the liquid crystal originates in transitions between stationary states with different director configurations. Simple calculations support this picture, as do our observations of periodic changes in sample color and transmittance. Detailed study of this effect is likely to lead to a better understanding of the flow properties of liquid crystals and other systems, such as sands and seeds, consisting of anisotropic particles.

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