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INFLUENCE OF AN ELECTRIC FIELD ON THE FLOW ALIGNMENT ANGLE
IN SHEAR FLOW OF NEMATIC LIQUID CRYSTALS

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Abstract Two nematic liquid crystals are studied in a torsional shear flow apparatus. The nematics are MBBA (neg.diel.anisotropy) and 5CB (pos.diel.anisotropy). The director align in the shear at a certain angle θ_n to the normal to the flow direction, except at two boundary layers near the surfaces. The influence of an electric field on the flow alignment angle and on the size of the boundary layers is studied both theoretically and experimentally.

I - INTRODUCTION

The director orientation in a nematic is strongly dependent on e.g. applied electric or magnetic fields, treatment of the bounding surfaces or flows in the liquid. The nematic is orientationally "soft", since the elasticity associated with deformations in the director field is very weak. In flow experiments this softness makes flow alignment of the director occur even at extremely low flow rates.

In simple flow geometries, like the shear flow studied in this paper, a stationary director configuration in the nematic can exist as a result of the balance between two counteracting viscous torques on the director. Far from the walls and at sufficiently high shear rates (so that we can neglect boundary and elastic effects) the angle between the normal to the shear direction and the director θ_n , is given by the simple expression (in the absence of an electric field)¹

$$\tan^2 \theta_n = \alpha_2 / \alpha_3$$

where α_2 and α_3 are two viscosity constants to be discussed later. The phenomenon of flow alignment has been observed in different situations (simple shear flow, Pouseuille flow, Couette flow) and for different liquid crystals.²⁻⁶ The angle θ_n is found to be close to 90 degrees, and to decrease with increasing temperature.

We assume in this work that the viscosities α_2 and α_3 are negative. In this case we have a stable flow alignment of the director up to a certain shear rate, where disclinations start to form from the walls. We work in a disclination free flow situation, and apply an electric a.c. field simultaneously with the shear flow. A stable flow alignment is observed also in this case. The flow alignment angle will depend on the strength of the electric field and on the shear rate. We study in particular the flow alignment angle as a function of applied electric field in the limit of small fields. In this limit we can measure the viscosity constants α_2 and α_3 experimentally.

The use of an a.c. electric field is of experimental importance. A d.c. field may cause injection of charges into the sample, and a voltage drop will occur in the liquid crystal-glass plate interface which we know very little about. It is also well known that d.c. fields and a.c. fields of low frequency may cause instabilities in the sample and give rise to an entirely new situation. Liquid crystals with negative and positive dielectric anisotropy (MBBA and 5CB respectively) are used in the experiments.

Generally, at arbitrary shear rates and strengths of the applied electric field, the director orientation in the nematic is very hard to get from the theory. However, for high enough shear rates we have a simple director configuration in the bulk of the nematic and the influence of the boundaries can be treated as a small perturbation in the theory. It is work in this regime we present in this paper.

II - EXPERIMENTAL

The experimental method of torsional shear flow was introduced by Wahl and Fischer⁶. The nematic is held between two circular glass plates (diameter 50 mm), the lower of which rests on a tube rotatable around a vertical axis and connected to a ten-step-gear synchronous motor. Typical distances between the plates are a few hundred μm . The angular velocity ω of the lower plate relative the upper, fix, can be varied between $2 \times 10^{-5} \text{ s}^{-1}$ and $2 \times 10^{-2} \text{ s}^{-1}$. Because of this rather low angular velocity we can consider the flow as a simple shear flow. At a given radius r we then have a linear shear velocity of $v = \omega r$.

Through the tube holding the lower glass plate an expanded, parallel, He-Ne laser beam (wavelength 632.8 nm) is incident from below. The liquid crystal film is observed between crossed polarizers. The experimental set up is shown schematically in fig. 1a. When the laser beam has passed the liquid crystal film and the lower plate is rotating, an interference pattern consisting of dark rings is observed, corresponding to the geometrical feature that the shear velocity increases radially outwards from the center. This pattern is recorded directly on a photographic film. Example of an interference pattern is shown in fig. 1b. As can be seen from this figure it is very difficult to

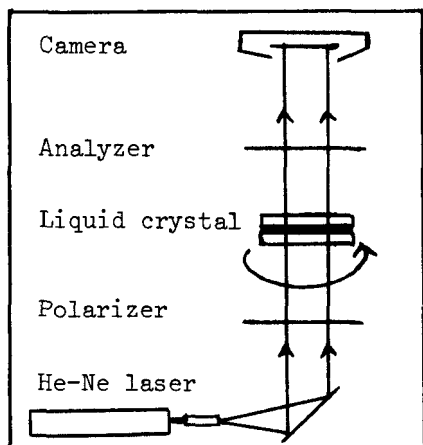


FIGURE 1a A schematic picture of the experimental set up. A beam expander is placed after the laser.

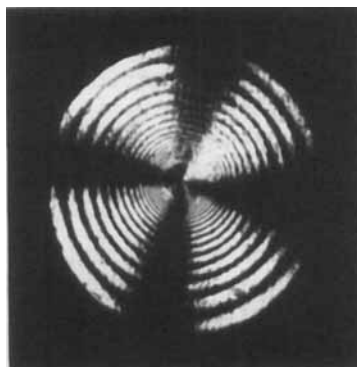


FIGURE 1b Interference pattern of flowing 5CB.

optically resolve the dark rings in the central region, and it is therefore not possible to assign the correct ring number to the outer rings that actually can be resolved and whose radii are measured. To overcome this difficulty we place a photocell outside the largest ring, and stop the motor. Then the rings on their outward motion pass the photocell as the director lines up normal to the glass plates subject only to the boundary conditions, and the ring numbers can be counted by recording the photocurrent⁶.

If we rotate the lower plate for a longer time than about five minutes, disclination lines start to form from the walls as pointed out by Wahl and Fischer. The lines are circular and begin to form in the outer region of the plates, where the velocity is greatest, and spread inwards. Since we want to study disclination-free flow, we must do the experiments before the disclinations have formed. When they have been created by the flow, the motor is stopped and we wait until they have disappeared. For the biphenyl the time until we again have a nematic "single crystal" could be reduced by applying an electric a.c. field over the plates. Even with an applied field the waiting time was about half an hour.

III - THEORETICAL STUDY OF THE FLOW ALIGNMENT ANGLE AND THE BOUNDARY LAYER IN THE PRESENCE OF AN ELECTRIC FIELD

In paragraph II the experiment is described. Here we shall study the theory behind it. In IIIa and IIIb expressions for the

flow alignment angle and the boundary layer in the presence of an electric field will be derived. In IIIc we will expand these to linear order in E_r^2 .

The equation which we will study is the equation that describes the motion of the director. This is given by the Leslie-Ericksen - Parodi theory as

$$I D_t D_t n_i = g_i + \partial_j \Pi_{ji} \quad (1)$$

The quantity I is the moment of inertia of the molecules, g_i includes electromagnetic and other effects and Π_{ji} is a stress tensor. If we now seek stationary solutions to our problem, the director equation (1) can be rewritten as follows⁷ (all derivatives are with respect to z).

$$\begin{aligned} & \theta''(K_{11}\sin^2\theta + K_{33}\cos^2\theta) + \theta'^2(K_{11} - K_{33})\sin\theta\cos\theta + \\ & + \frac{1}{2} u'(-\gamma_1 + \gamma_2\cos 2\theta) - \epsilon_a E_r^2 \cos\theta\sin\theta = 0 \end{aligned} \quad (2)$$

Here K_{11} and K_{33} are the Frank elastic constants and

$$\gamma_1 = \alpha_2 - \alpha_3 \quad \gamma_2 = -(\alpha_2 + \alpha_3) \quad (3)$$

where α_2 and α_3 are the Leslie coefficients of viscosity and $\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp}$ is the dielectric anisotropy of the molecules. We now do the one-constant approximation setting $K_{11} = K_{33} = K$, and eq. (2) then simplifies to

$$K\theta'' - \frac{1}{2} u'\gamma_1 + \frac{1}{2} u'\gamma_2\cos 2\theta - \frac{1}{2} \epsilon_a E_r^2 \sin 2\theta = 0 \quad (4)$$

IIIa - Study of the flow alignment angle

Let us consider first the solution of eq. (4) far from the walls. At a sufficiently high shear rate u' we can set $\theta'' = 0$ and this together with the relations

$$\cos 2\theta = \frac{1 - \tan^2\theta}{1 + \tan^2\theta} \quad \sin 2\theta = \frac{2\tan\theta}{1 + \tan^2\theta} \quad (5)$$

will make it possible to rewrite eq. (4) as

$$\tan^2\theta_n + \frac{2\epsilon_a E_r^2}{u'(\gamma_1 + \gamma_2)} \tan\theta_n + \frac{\gamma_1 - \gamma_2}{\gamma_1 + \gamma_2} = 0 \quad (6)$$

which has the solution

$$\tan\theta_n = -\frac{\epsilon_a E_r^2}{u'(\gamma_1 + \gamma_2)} + \sqrt{\frac{\epsilon_a^2 E_r^4}{u'^2(\gamma_1 + \gamma_2)^2} + \frac{\gamma_1 - \gamma_2}{\gamma_1 + \gamma_2}} \quad (7)$$

We shall now argue that it is the upper sign in eq. (7) which gives the correct solution. Setting $E_r = 0$ in eq. (7) we get

$$\tan\theta_n = \frac{+\gamma_2 - \gamma_1}{(-)\gamma_2 + \gamma_1} = \frac{+\alpha_2}{(-)\alpha_3} \quad (8)$$

If we compare this expression with the known solution to the shear flow problem with $E_r = 0^1$, we confirm our choice of the + sign in eq. (7).

IIIb - Study of the boundary layer

We now expand eq. (4) to linear order in $\delta\theta = \theta - \theta_n$. First we get the relations

$$\theta'' = \delta\theta'', \quad \sin 2\theta \approx \sin 2\theta_n + 2\cos 2\theta_n \delta\theta, \quad \cos 2\theta_n \approx \cos 2\theta_n - 2\sin 2\theta_n \delta\theta \quad (9)$$

Eq. (9) together with eq. (4) now give

$$\begin{aligned} & -\frac{1}{2} u' \gamma_1 + \frac{1}{2} u' \gamma_2 \cos 2\theta_n - \frac{1}{2} \epsilon_a E_r^2 \sin 2\theta_n + K \delta\theta'' - \\ & - \delta\theta u' \gamma_2 \sin 2\theta_n - \delta\theta \epsilon_a E_r^2 \cos 2\theta_n = 0 \end{aligned} \quad (10)$$

The sum of the first three terms in eq. (10) equals zero according to eq. (4) and we can now write eq. (10) as

$$K \delta\theta'' = \{u' \gamma_2 \sin 2\theta_n + \epsilon_a E_r^2 \cos 2\theta_n\} \delta\theta = Q \delta\theta \quad (11)$$

which has the solution

$$\delta\theta = \delta\theta_0 e^{-z/\xi}, \quad \xi = \sqrt{K/Q} \quad (12)$$

This tells us that the effect of the wall will die out in a distance given by ξ which will be our boundary layer.

IIIc - Study of the flow alignment angle and the boundary layer for small fields

We see from eq. (7), (11) and (12) that the expressions for θ_n and ξ are rather complicated functions of E_r^2 . Here, we shall study how these functions simplify in the limit of small fields. Eq. (3) and (7) give

$$\tan \theta_n = \frac{\epsilon_a E_r^2}{2u' \alpha_3} + \sqrt{\frac{\alpha_2}{\alpha_3}} \sqrt{1 + \frac{\epsilon_a^2 E_r^4}{4u'^2 \alpha_2 \alpha_3}} \quad (13)$$

$$\text{If now } E_r^2 \ll \frac{2u' \alpha_2 \alpha_3}{|\epsilon_a|} \quad (14)$$

we can expand eq. (13) to linear order in E_r^2 and we get

$$\tan \theta_n \approx \sqrt{\frac{\alpha_2}{\alpha_3}} + \frac{\epsilon_a}{2u' \alpha_3} E_r^2 \quad (15)$$

The expression for ξ in the same limit will after some calculations turn out to be

$$\xi \approx \sqrt{\frac{K}{2u' \alpha_2 \alpha_3}} \left\{ 1 + \frac{\epsilon_a}{4u'} \left(\frac{1}{\alpha_2 \alpha_3} + \frac{1}{\alpha_2 \sqrt{\alpha_2 \alpha_3} + \alpha_2 \sqrt{\alpha_2 \alpha_3} / \alpha_3} \right) E_r^2 \right\} \quad (16)$$

IV - MEASURING THE FLOW ALIGNMENT ANGLE AND THE BOUNDARY LAYER

When you do a shear flow experiment you put a thin (100-500 μm) sample of a normally oriented nematic between two parallel glass plates. If you move the lower plate with a velocity v relative the upper one, you will introduce a shear flow with shear rate $u' = v/d$. Generally, at arbitrary shear rates, the angle between the flow direction and the director is given as a function of z by the theory as a second order, non-linear differential equation (our eq.4). At the high shear rate we use, the situation simplifies, and the angle is very nearly constant throughout the liquid crystal layer (along z except at the boundaries. This is the assumption used setting $\theta'' = 0$ when solving eq.(5). In figure 2 is shown an idealized picture of the director field in this case.

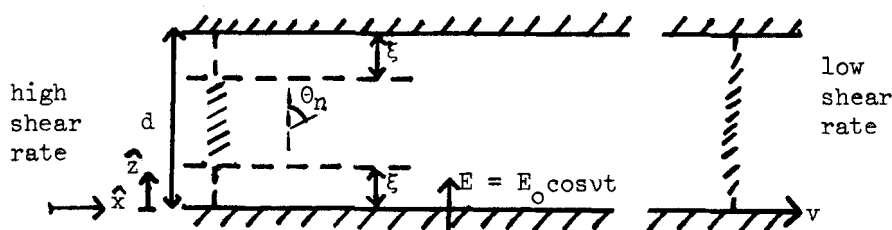


FIGURE 2 An idealized picture of the director field for large u' . ξ is the boundary layer and θ_n is the flow alignment angle. The situation for low shear rate is shown to the right.

In our experiment we have put the sample between two circular glass plates, the lower one rotating with a constant angular velocity ω . We then get a director field as a function of $u' = r\omega/d$ according to eq. (7) and (12) (r is the radial distance to the center of the plates). We then observe the nematic between crossed polarizers, and now we see interference pattern consisting of concentric dark rings. The condition for ring n will be⁶

$$2\pi n = \eta(d - 2\xi) \quad (17)$$

$$\text{Where } \eta = \frac{2\pi n_1}{\lambda_0} \left\{ \frac{1}{\sqrt{1 - \delta \sin^2 \theta_n}} - 1 \right\}, \quad \delta = 1 - n_1^2/n_2^2 \quad (18)$$

n_1 and n_2 are the refractive indices perpendicular and parallel to the molecule axes and λ_0 is the wavelength of the polarized light.

Doing two experiments with different sample thicknesses d_1 and d_2 but with the same shear rate u' and electric field E , you will according to eq. (7), (11) and (12) get the same flow alignment angle θ_n and the same boundary layer ξ . If you call the corresponding ring numbers n_1 and n_2 respectively you get from eq. (17)

$$2\pi n_1 = \eta(d_1 - 2\xi) \quad , \quad 2\pi n_2 = \eta(d_2 - 2\xi) \quad (19)$$

Solving eq. (19) for ξ and η gives

$$\xi = \frac{n_2 d_1 - n_1 d_2}{2(n_2 - n_1)} \quad , \quad \eta = \frac{2\pi n_1}{d_1 - 2\xi} \quad (20)$$

Combining eq. (18) and (20) you get

$$\sin^2 \theta_n = \frac{\eta^2 \lambda_0^2 + 4\pi n_1 \eta \lambda_0}{(1 - n_1^2/n_2^2)(\eta \lambda_0 + 2\pi n_1)^2} \quad (21)$$

V - RESULTS AND DISCUSSION

Our main result is eq. (15). It gives the angle θ_n as a function of applied electric field E_a and shear rate $u' = v/d$ for fields fulfilling the condition of eq. (14). In eq. (15) we see, that for zero applied field, the angle is independent of the shear rate and given by $\arctan \sqrt{\alpha_2/\alpha_3}$, which is the result found in the literature.

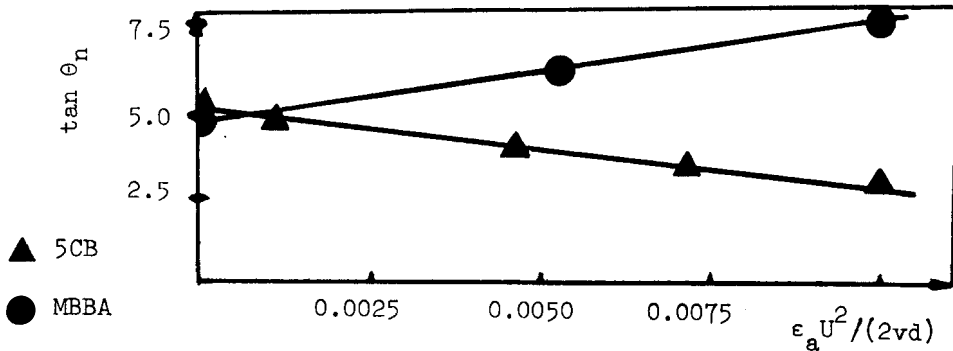


FIGURE 3 θ_n as a function of electric field.

In figure 3 we show our experimental result for θ_n , as a function of applied electric field. From the slope of the curve at low voltage we can compute α_3 from eq. (15). From the intercept with the vertical axes we get the ratio α_2/α_3 , and combining these two values we get α_2 and α_3 for MBBA and 5CB. The experiments were done at 20 °C and we used the following data for the refractive indices and the dielectric anisotropy^{8,9}:

MBBA: $n_1 = 1.555$, $n_2 = 1.787$, $\epsilon_a = -0.7$

5CB: $n_1 = 1.528$, $n_2 = 1.713$, $\epsilon_a = 12$

With these values we obtain:

MBBA: $\alpha_2 = -93$ cp $\alpha_3 = -4$ cp 5CB: $\alpha_2 = -130$ cp $\alpha_3 = -5$ cp

Existing data in the literature for MBBA is $\alpha_2 = -78$ cp and $\alpha_3 = -1$ cp⁸.

Of course, the director configuration shown in figure 2 is only an approximation of the true one. Better values for α_2 and α_3 would be obtained if one used a director configuration which corresponded to the full solution of eq. (4). Furtheron, doing the one-constant approximation influences the expression for ξ given by eq. (12). However as is seen when going from eq. (2) to eq. (6) this will not influence the value for Θ_n . Examination of our method of measuring Θ_n in paragraph IV also shows that the values for α_2 and α_3 does not suffer from the limitations of the one-constant approximation.

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