Two Shear-Flow Regimes in Nematic *p-n*-Hexyloxybenzilidene-*p*′-aminobenzonitrile

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The viscous torque induced in a plane shear flow on the director $\vec{n}$ of *p-n*-hexyloxybenzilidene-*p*′-aminobenzonitrile (nematic below $T_c \sim 102°C$) when $\vec{n}$ is parallel to the velocity changes sign at a temperature $T_c - 8°C$, in agreement with Gähwiller's experiments on a Poiseuille flow. We compare the instability modes obtained near T_c with the similar problem for *p*′-methoxybenzylidene-*p-n*-butylaniline and give the first discussion of the instability modes in the low-temperature domain.

The uniform alignment of a nematic liquid crystal (LC) characterized by a unit vector along the optical axis (the director $\vec{n}$) can be disturbed by a shear flow. If $\vec{n}$ is in the plane of the velocity gradient, $s = dv_x/dz$, the hydrodynamic torque exerted per unit volume[1] is given by

$$\Gamma_y^h = (\alpha_3 \cos^2\theta - \alpha_2 \sin^2\theta)s, \qquad (1)$$

where θ is the angle between $\vec{n}$ and $\vec{v}$ (the geometry is given in the inset of Fig. 1). In practice, $\alpha_2 < 0$ and $|\alpha_2| \gg |\alpha_3|$. Two different hydrodynamic problems are met depending on the sign of α_3. The possibility of a change of sign in α_3 is not surprising. In a molecular model, it can be produced by the mere change of the end shape of elongated rods.[2] It has also been predicted next to a second-order phase transition from a nematic to smectic-*A* LC where α_3 should show a positive divergence.[3] The experiments reported here were done on *p-n*-hexyloxybenzilidene-*p*′-aminobenzonitrile (HBAB) which is nematic below $T_c \sim 102°C$.[4]

In Poiseuille-flow experiments with a planar configuration on HBAB, Gähwiller[5] found that α_3 was negative close to T_c and positive below $\sim T_c - 10°C$. The latter result was questioned later by Meiboom and Hewitt,[6] who used a cylindrical Couette geometry and a capacitive measurement in a magnetic field. However, no direct observation of the alignment could be done in the experiment.

In this experimental Letter, we confirm the result of Gähwiller and present the first discussion of the nature of the hydrodynamic instabilities obtained in the two cases $\alpha_3 < 0, > 0$.

In our work, the shear is obtained by displacing an upper horizontal glass plate with respect to a fixed lower one. The distance between the plates is $d = 190\ \mu m$. Semitransparent Au films evaporated under oblique incidence on the inner sides of the plates provide an initial uniform alignment of $\vec{n}$ in the plane of the film. They also serve as electrodes for applying a high-frequency electric field $\vec{E}$, which gives rise to a destabilizing torque

$$\Gamma_y^E = (\epsilon_a E^2/4\pi) \sin\theta \cos\theta \qquad (2)$$

[we have measured independently the positive dielectric-constant anisotropy $\epsilon_a(T)$; at 75°C, $\epsilon_a = \epsilon_\parallel - \epsilon_\perp = 13.1 \pm 0.5$].

If the field is large enough compared to the Freedericksz threshold (~0.75 V), the molecules are practically aligned along the field over the film thickness. More generally, it is possible in this case to neglect the elastic torques relative to Γ_y^E as the changes in the distortion angles take place over an electric coherence distance small

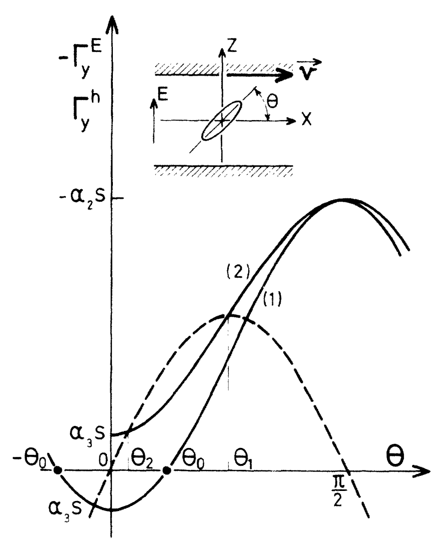

FIG. 1. Electric torque Γ_y^E (dashed line) and hydrodynamic one Γ_y^h (full line) for a uniform molecular distortion θ in the x-z plane (the geometry is given in the inset). In large shear, an equilibrium angle θ_0 is obtained only if $\alpha_3 < 0$ (curve 1).

compared to d. The alignment and the changes of orientation are measured optically using the conoscopic image in monochromatic convergent light.

The sign of α_3 is obtained qualitatively at once (without an applied field). Starting from an oriented film with $\vec{n}$ parallel to $\vec{v}$, a shear is applied. The center of the conoscopic image is initially displaced in the direction of $\vec{v}$ when $T < T_c - 8°C$ and in the opposite one when $T_c - 8°C < T < T_c$. The latter case is the one obtained in most nematics. In this range, if the shear is large enough, the molecules are aligned in the bulk at an angle θ_0 such that[1]

$$\tan^2\theta_0 = \alpha_3/\alpha_2. \tag{3}$$

This can be seen easily from the solid curve 1 of Fig. 1. The solution $-\theta_0$ is an unstable state. On the other hand, if α_3 is positive (curve 2), Γ_y^h never vanishes. At large shears, no stable solution in the plane of the velocity and velocity gradient is obtained. However, in a small enough shear Γ_y^h can be balanced by the elastic "splay" torque

$$K_1 d^2\theta/dz^2 = \alpha_3 s,$$

or

$$\theta(z) = \tfrac{1}{2}(\alpha_3 s/K_1)(z^2 - \tfrac{1}{4}d^2). \tag{4}$$

The elastic constant K_1 is found using the Freedericksz transition in an electric field $\vec{E}$. The expressions (3) and (4) give two determinations of $\alpha_3(T)$. Both imply small distortions (typically a change of optical path of one wavelength for a 200-μm film). The weak change of birefringence is measured by placing a quarter-wavelength plate at 45° with $\vec{n}$ after the LC and measuring the rotation of the analyzer. The values of θ_0 agree with the determinations of Gähwiller.[5] Typically, θ_0 decreases from 12° at $T_c - 4°C$ to zero at $T_c - 8°C$.

Direct measurements of $\alpha_3(T)$ are obtained for both signs of $\alpha_3(T)$ in small shears using (3) and the measurement of the change in the optical path in the presence of the distortion,

$$\delta = \tfrac{1}{2}n_e(1 - n_e^2/n_0^2)\int_{-d/2}^{+d/2}\theta^2(z)\,dz. \tag{5}$$

Our determination of the birefringence $n_e - n_0$ agrees with that of (4). The optical path varies as s^2 only for very low shears (typically $s < 0.1$ sec^{-1}); for higher shears [typically when $\theta(z=0) > 5°$] the α_2 contribution of (1) which was neglected in (4) is larger than the α_3 term. The values of $\alpha_3(T)$ (points of Fig. 2) agree reasonably with those of Gähwiller.

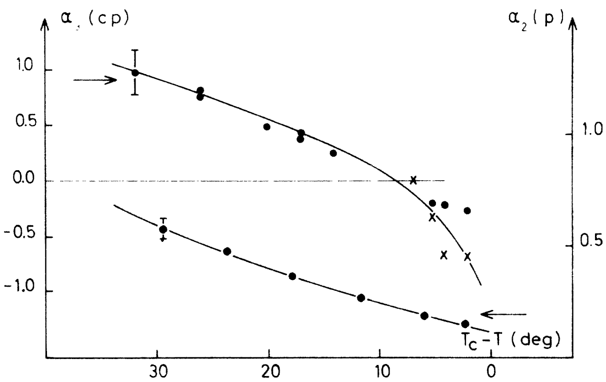

FIG. 2. The constant α_2 is determined from the equilibrium angle θ_1 in Fig. 1. One determination of α_3 (crosses) uses the values of θ_0. The other one (dots) is obtained from the distortion in low shears. The accuracy of α_3 is poor but the change of sign of α_3 is unambiguously obtained. Note the different scales for α_2 and α_3.

The α_2 torque is measured using the following techniques: First, a large electric field ($E = 10$ V) is applied across the sample. (By applying a shear temporarily, we prepare the initial conditions for the Freedericksz transition so that we get a $\theta > 0$ distortion and avoid the formation of domains.) A dc shear is subsequently applied and an equilibrium distortion results from the balance between Γ_y^E and Γ_y^h. The intercept of the two corresponding curves in Fig. 1 shows the two solutions θ_1 and θ_2. Only the higher one θ_1 is a stable state. The sign of θ is such that the director $\vec{n}$ remains in the x-z plane (fluctuations off this plane give a vertical restoring component to the hydrodynamic torque). The angle θ_1 can be measured easily by comparing the count of interference fringes p, from the undistorted state, to the total number of fringes N when the planar structure is converted practically to a homeotropic one in the presence of a large enough E. We have calculated θ numerically as a function of p and N.

By doing the measurements for two different shears and solving (1) and (2), it should be possible in principle to get both α_2 and α_3. However, the measurements involve relatively large distortion angles where the α_3 contribution is negligible. The variation of $\alpha_2(T)$ given in Fig. 2 agrees reasonably well with that of Gähwiller. In the domain $\alpha_3 < 0$, we use the values of θ_0 and α_2 to get a new determination of $\alpha_3(T)$ (crosses in Fig. 2).

We come now to the problem of hydrodynamic stability. In a recent work,[7,8] we analyzed the stability of a nematic LC under a shear flow such that $\vec{n}$ ($\parallel \hat{y}$) was perpendicular to $\vec{v}$ ($\parallel \hat{x}$) and the velocity gradient ($\parallel \hat{z}$). Here we extend this study to the case of HBAB. Near T_c, the problem is similar to the one studied with p'-methoxybenzylidene-p-n-butylaniline (MBBA) where α_2 and α_3 are also negative. Two instability regimes were found. In a first mode[7] a uniform fluctuation of $\vec{n}$ in the x-y plane, away from the y axis, is amplified by the coupled destabilizing action of hydrodynamic torques measured by α_2 and α_3, if the shear is large enough. Another instability mode[8] involves rolls having their axis parallel to $\vec{v}$ and associated with a fluctuation in $\vec{n}$: $n_z(y)$, $n_y(y)$. The main destabilizing torque off the horizontal plane, due to the term $\alpha_3 n_y$ in the first case, is now associated with the periodic distortion of $n_y(z)$. This term has been described as a hydrodynamic-focusing effect[9] and is essentially associated with the anisotropy of the viscosity in the x-y plane. The roll instability exists in HBAB for both signs of α_3. But, the homogeneous one is only found near T_c where α_3 is negative. (It is easily seen that the mechanism invoked in this regime is stabilizing when $\alpha_2\alpha_3 < 0$.) A detailed discussion of the instability threshold is beyond the scope of this Letter. Let us note however that E plays a role complementary to the case of MBBA where ϵ_a is negative.

We come back to the geometry where $\vec{n}$ is parallel to $\vec{v}$ and α_3 negative. If no electric field is present or if the shear is too large so that there exists no solution of $\Gamma_y^E = \Gamma_y^h$, the molecules distort towards negative values of θ. However, such a state is unstable with respect to fluctuations of $\vec{n}$ out of the x-z plane. Once a component n_y is established, the vertical torque $\alpha_2\theta n_y$ twists the molecules further off the plane and the splay distortion along z is reduced as the twist increases. In practice, it is found that for large enough shears (typically $s \sim 1$ sec^{-1}) the molecules are practically perpendicular to $\vec{v}$. The configuration is very similar to that discussed above with $\vec{n}$ parallel to $\hat{y}$. As expected, we find that if we increase s further ($s \sim 4$ sec^{-1}), a one-dimensional instability with the roll axis parallel to $\vec{v}$ takes place. This is a rather remarkable example of a cascade of instabilities.

The HBAB was synthesized in Orsay and had a $T_c = 102°$C. The same experiment was done with an "old" sample with a T_c reduced to 95°C with similar results. In particular, a change of sign in α_3 was obtained at $T_c - 8°$C. As the quality of the material is not crucial, it is not likely to explain the result of (5). However, we feel that no unambiguous conclusion could be drawn from it as no direct observation of the distortion pattern could be obtained.

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