Stability of Nematic Liquid Crystals in Couette Flow

P. E. Cladis and S. Torza
Bell Laboratories, Murray Hill, New Jersey 07974
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We have studied the behavior of several nematics in Couette flow with homeotropic boundary conditions. When Leslie's viscosity $\alpha_3>0$, the director tumbles at a critical shear rate, S_c. When $S>S_c$ a *stable, stationary state* is observed. When $S>S_E>S_c$, the flow becomes cellular. We calculate S_c and S_E. Knowing S_c, S_E, the shear viscosity γ_1, and the Frank elastic constants, it is possible to deduce separately the Leslie viscosities α_3 and α_2 when $|\alpha_2|>\alpha_3>0$.

Recently, a great deal of attention[1-4] has been focused on the question of "tumbling" of the nematic director, $\vec{n}$, in shear flow. Tumbling refers to the nonexistence of a stationary state ($\partial\vec{n}/\partial t\neq\vec{0}$) for the director. It is specifically supposed to occur when α_3, a Leslie viscosity,[5-7] becomes positive, whereas α_2, another Leslie viscosity, remains negative. In the absence of pretransitional effects due to a nearby smectic-*A* phase $|\alpha_3|\ll|\alpha_2|$ and they are both negative. Smectic-*A* pretransitional effects are expected[8] to drive α_3 through zero and on to $+\infty$ at a second-order nematic-smectic-*A* transition. They are not expected to affect α_2.

It has been claimed on theoretical grounds[2] that tumbling does not occur and indeed, working in Couette flow[9] [with *p*-*n*-hexyloxybenzylidene-*p*'-aminobenzonitrile (HBAB)], Meiboom and Hewitt[1] found α_3 always negative and no evidence of tumbling. On the other hand, Gähwiller[3] and Pieranski and Guyon,[4] studying the flow between two parallel plates, found that α_3 for HBAB is positive or negative depending upon the temperature. The question is what does the director do when $\alpha_3>0$ and tumbling is supposed to occur?

We have studied the behavior of HBAB and CBOOA (N-*p*-cyanobenzylidene-*p*-*n*-octyloxyaniline) in a small (inner radius $R_1\sim0.25$ mm, outer radius $R_2\sim0.5$ mm) glass Couette which has been treated with the appropriate surfactant[10] so that at rest, $\vec{n}=(1,0,0)$. Our outer cylinder is fixed. With a series of gears, we are able to rotate the inner shaft at speeds ranging from 10^{-3} to 50 revolutions per second. The Couette is epoxied into a Mettler hot stage between microscope slide and cover slip and surrounded with microscope immersion oil. A microscope is used to observe visually the flow field and the director orientation.

We have found that, for Ericksen number[11] $\mathcal{E}$ less than a critical value $\mathcal{E}_c^0$ and $\alpha_3>0$, initial elastic forces can balance the small viscous stresses. When $\mathcal{E}=\mathcal{E}_c^0$, the director tumbles to a new configuration of greater elastic energy and this new configuration is also stationary and in the plane of the velocity and velocity gradient. This is in contrast to the result of Pieranski and Guyon[4] where at large shears, no stable solution in the plane of the velocity and velocity gradient is obtained in the regime $\alpha_3>0$. For CBOOA whenever the temperature was less than ~88°C, this planar solution was the only one we obtained even at our highest shear rates. We also found that whenever $\mathcal{E}\gtrsim\mathcal{E}_E^0>\mathcal{E}_c^0$, a secondary cellular flow developed. The size of the secondary flow cells decreases in the axial direction as $\mathcal{E}$ increases until for $\mathcal{E}\sim5\mathcal{E}_E^0$ disclinations form around the inner shaft and eventually fill the entire gap between the two cylinders. Here we will restrict our attention to the regime of small $\mathcal{E}$ ($\lesssim10^2$) and the case $\alpha_3>0$. By analyzing the balance of viscous and elastic stresses on the director, we are able to arrive at values for α_2 and α_3 for HBAB which are entirely consistent with those of Pieranski and Guyon.

Figure 1 shows our measurement of the velocity profile, $\omega(r)$, in the gap between the two cylinders. We found this by suspending Sefadex particles (2–10 μm diam) in HBAB, filming them in rotation, then projecting the film frame by frame. It shows that, excluding the boundary layers[12] $R_1\leq r\leq R_1^*$ and $R_2^*\leq r\leq R_2$, the flow field is the usual Couette flow for a Newtonian fluid (solid line in Fig. 1), i.e.,

$$r\frac{d\omega}{dr}=-\frac{2\omega'(R_1^*R_2^*)^2}{(R_2^*)^2-(R_1^*)^2}\times r^{-2},\qquad(1)$$

where R_i^* depend upon $\mathcal{E}$. As $\mathcal{E}$ increases $R_i^*\to R_i$. The inner boundary layer appears to be in rigid rotation whereas the outer one is stationary. The director adjusts from the orientation dictated by the walls (homeotropic) to the one dic-

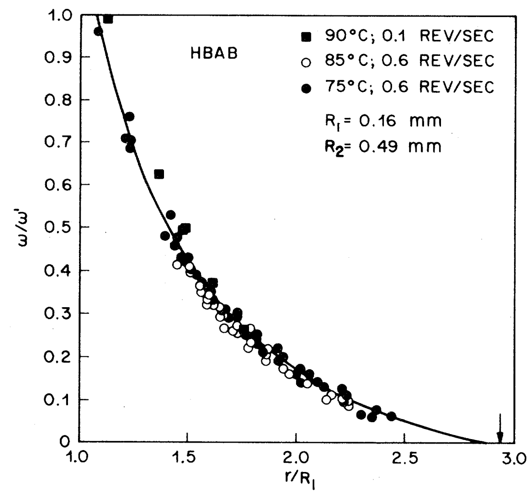

FIG. 1. ω/ω' versus r/R_1 found by observing the rotation of Sefadex particles suspended in the liquid crystals. Note the boundary regions. The solid line is the ordinary Couette-flow velocity profile [Eq. (1)].

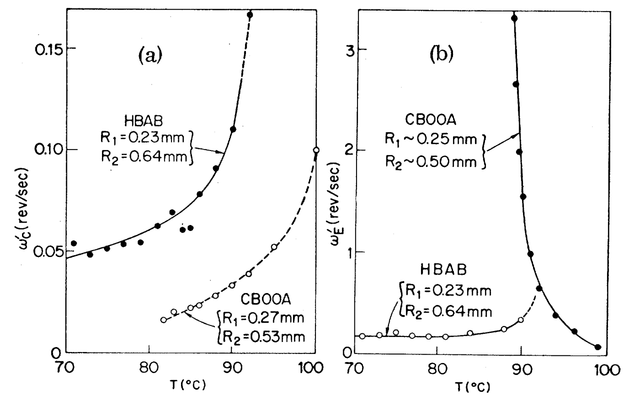

FIG. 2. (a) ω_c' versus temperature for HBAB and CBOOA. The circles represent our measured values. The solid line for HBAB is calculated from Eq. (4) using measured values for R_i^*, α_3, α_2 (Ref. 4), and K_3/χ_a (Ref. 16). We used $\chi_a = 1.14\times 10^{-7}$ cgs. (b) ω_E versus temperature for CBOOA and HBAB. When 86°C $< T <$ 88°C, for CBOOA the planar Couette flow appears to be completely stable even at very large shears. $\omega_E' \to \infty$ when $\alpha_3 = |\alpha_2|$ (CBOOA) and $\alpha_3 \to 0^+$ (HBAB) [Eq. (5)].

tated by the flow field in the boundary layers, where we have observed $n_z \neq 0$ when $\omega' > \omega_c'$ (ω' being the rotation rate of the inner cylinder). In the effective gap, $R_1^* \leq r \leq R_2^*$, the director configuration appeared planar ($n_z = 0$) when $\omega' < \omega_E'$ (or $\mathcal{E} < \mathcal{E}_E{}^0$). We checked this using the technique of double images.[13]

Figure 2(a) shows ω_c' versus temperature for HBAB and CBOOA. When $\omega' < \omega_c'$, the director appears to be nearly constant in the effective gap. We call this configuration the mode $k=0$. When $\omega' = \omega_c$, the director tumbles to a new stationary configuration where optically one observes two lines parallel to the cylinder axis forming in the effective gap (these we call tumbling lines) indicating orientation changes of the director of π and $-\pi$. We call this new configuration the mode $k=1$. Once the mode $k=1$ is achieved $n_z \neq 0$ in the boundary layers. The larger elastic energy stored in the mode $k=1$ can balance a larger viscous stress than the mode $k=0\,[\mathcal{E}_c{}^0 < \mathcal{E}_c{}^1]$, and thus this configuration is stationary for $\mathcal{E} < \mathcal{E}_c{}^1$. Presumably when $\mathcal{E} = \mathcal{E}_c{}^1$, the director can tumble again to an even more energetic configuration ($k=2$) which will once again maintain $\vec{n}$ stationary. This phenomenon has already been envisioned by de Gennes[14] and we confirm his prediction here for the first time. In our experiments, however, we observed that before the configuration (mode $k=2$) appeared, as we increased ω', the distance between the two tumbling lines increased (i.e., $k \to 0$ in this region and an additional tumble in this region would have led to the mode $k=2$) and a secondary cellular flow developed when $\mathcal{E} = \mathcal{E}_E{}^0 \lesssim \mathcal{E}_c{}^1$. The director orientation became nonplanar ($n_z \neq 0$), but not uniformly so, in the effective gap. This again is in contrast to the results of Pieranski and Guyon.[4] Figure 2(b) shows ω_E' as a function of temperature for HBAB and CBOOA.

This secondary cellular flow resembles the Taylor vortex flow of isotropic liquids,[15] however, the Taylor number for this flow is about three orders of magnitude *smaller* than the Taylor number (~42) for isotropic liquids and is the result of unbalanced elastic torques and not of centrifugal forces. We have also observed classic Taylor vortices in N-[p-methoxybenzylidine]-p-butylaniline and CBOOA (Taylor number ~95 as predicted by Leslie[9] for anisotropic liquids).

To calculate $\mathcal{E}_c$ and $\mathcal{E}_E$, we write the balance of viscous and elastic torques as[5-9]

$$\partial\vec{n}/\partial t = \tfrac{1}{2}(\nabla\times\vec{v})\times\vec{n} - \vec{v}\cdot\nabla\vec{n} + \lambda[\vec{A}\cdot\vec{n} - (\vec{n}\cdot\vec{A}\cdot\vec{n})\vec{n}] + \beta[(\vec{h}\cdot\vec{n})\vec{n}], \tag{2}$$

where $\lambda = -\gamma_2/\gamma_1 = -[(\alpha_3+\alpha_2)/(\alpha_3-\alpha_2)]$ using Parodi's relation,[16] $\beta = 1/\gamma_1 = (\alpha_3-\alpha_2)^{-1}$, $\vec{h}$ is the molecular field,[7] $\vec{v}$ is the velocity field [$\vec{v}=(0, r\omega, 0)$ in cylindrical co-ordinates], $\bar{A}$ is one-half the symmetric part of $\nabla\vec{v}$. To simplify the interpretation of our results we use Eq. (1) and we take $\vec{n}=(\rho\cos\psi, \rho\sin\psi, (1-\rho^2)^{1/2})$; $d\rho/dr=0$ and $r\,d\psi/dr=$ const; in the effective gap we put $\psi=(\psi_c{}^0-k\pi)[1-\ln(r/\bar{R}^*)]$, where $\psi_c{}^0<0$ for $\omega'>0$, $k=0,1,2,\ldots$, and $\bar{R}^{*2}=(R_1{}^*R_2{}^*)^2/(R_2{}^{*2}-R_1{}^{*2})$. This configuration shows that the orientation ψ decreases from its maximum absolute value at $r=R_1{}^*$, where $(r\,d\omega/dr)$ is also maximum [Eq. (1)], to its minimum absolute value at $r=R_2{}^*$, where $(r\,d\omega/dr)$ is also minimum. We calculate $\partial\rho^2/\partial t$ and $\partial\psi/\partial t$ as a function of λ, ψ, and the Frank constants[17] K_i of splay ($i=1$), twist ($i=2$), and bend ($i=3$). In the limit $n_z=0$, we find a stationary solution ($\partial\psi/\partial t=0$) for $\bar{\psi}$ (ψ averaged over the effective gap) when

$$\tan\bar{\psi}=(\mathrm{sgn}\,\omega')\left(\frac{1+\lambda}{1-\lambda}\right)^{1/2}\left\{\frac{\mathcal{E}_c{}^k}{\mathcal{E}}+\left[\left(\frac{\mathcal{E}_c{}^k}{\mathcal{E}}\right)^2-1\right]^{1/2}\right\},\quad -1<\lambda<1, \tag{3}$$

where

$$\mathcal{E}_c{}^k=\frac{[1+(\psi_c{}^0-k\pi)^2]}{2(1-\lambda^2)^{1/2}}\,\frac{(K_3-K_1)}{K_3},\quad -1\le\lambda\le1. \tag{4}$$

When $\mathcal{E}>\mathcal{E}_c{}^k$, i.e., $\omega'>\omega_c'$, Eq. (3) shows that there are no real solutions for ψ and tumbling first occurs when $k=0$, $\tan\bar{\psi}=(-\alpha_2/\alpha_3)^{1/2}$, and the director configuration changes modes from $k=0$ to $k=1$. In the region where $k\to0$ we estimate the stability of the planar Couette using Ericksen's original analysis of stability[5] and Eqs. (1) and (2). We find that the planar solution ($n_z=0$) is unstable for Ericksen numbers smaller than a critical $\mathcal{E}_E{}^0$ (for which $\omega'=\omega_E'$) when

$$\mathcal{E}_E{}^0=\frac{1}{2\lambda}\left[\left(\frac{K_1}{K_3}\cot\bar{\psi}+\frac{K_2}{K_3}\tan\bar{\psi}\right)\left(1+\psi_c{}^{02}\right)+\frac{2}{K_3}\left(K_3-K_2\right)\frac{(\psi_c{}^0\cos\bar{\psi}+\sin\bar{\psi})^2}{\cos\bar{\psi}\sin\bar{\psi}}\right],\ \tan\bar{\psi}>0 \text{ when } \omega'>0,\ 0\le\lambda\le1. \tag{5}$$

When $\alpha_3\ge|\alpha_2|$ and $\alpha_3\to0^+$, Eq. (5) shows that $\mathcal{E}_E{}^0\to\infty$ ($\omega_E{}^0\to\infty$) and we no longer see the cellular flow. Thus qualitatively our simple model shows that $\omega_E'\to\infty$ for CBOOA at 88°C because here $\alpha_3=|\alpha_2|$.

Using Eqs. (3)–(5), our measured values for $\mathcal{E}_c{}^0$, $\mathcal{E}_E{}^0$, and K_i,[18] we now evaluate α_2 and α_3 separately. We find at 85°C for HBAB, $-\alpha_2=0.38$ P and $\alpha_3=0.23\times10^{-2}$ P. These values agree fairly well with the measurement of Pieranski and Guyon.[4] The agreement is independent of temperature as can be seen from the solid curve calculated from Eq. (4) with $k=0$ shown in Fig. 2(a).

In summary, we have observed a new flow regime for nematic liquid crystals when $\alpha_3>0$ where boundary layers are not negligibly small. We have used a simple model for the director configuration in the effective gap as a first step towards understanding our experimental results.

We have found that for Ericksen numbers $\mathcal{E}<\mathcal{E}_c{}^0$ [Eq. (4)], the elastic deformations in the effective gap can stabilize the viscous torques. When $\mathcal{E}\sim\mathcal{E}_c{}^0$, the director configuration is momentarily unstable (it tumbles) and changes modes from $k=0$ to $k=1$ and the critical Ericksen number changes from $\mathcal{E}_c{}^0$ to $\mathcal{E}_c{}^1>\mathcal{E}_c{}^0$. Tumbling in the sense $\partial\vec{n}/\partial t\ne\vec{0}$ forever did not occur. Apart from the boundary layers we found that even after tumbling $n_z=0$ in the effective gap provided $\mathcal{E}<\mathcal{E}_E{}^0$ [Eq. (5)]. When $\mathcal{E}\gtrsim\mathcal{E}_E{}^0>\mathcal{E}_c{}^0$, the planar Couette flow ($n_z=0$) in unstable and we have observed a cellular flow which resembles but is not "similar" to the Taylor vortices observed at much higher shears. When $\alpha_3\ge|\alpha_2|$, $\mathcal{E}_E{}^k=\infty$ and the planar configuration is stable for finite shears. Knowing $\mathcal{E}_c{}^k$, $\mathcal{E}_E{}^k$, and the Frank elastic constants it is possible to deduce α_3 and α_2 separately.

A more complete account of this work is in preparation.

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[11]We define the Ericksen number, $\mathcal{E}$, to be the ratio of the viscous ($\sim \gamma_1 S$) to elastic forces ($\sim K/\bar{R}^2$), where $\gamma_1 = \alpha_3 - \alpha_2$ is the shear viscosity, S a shear rate, K an elastic constant, and $\bar{R}$ a characteristic length. This is essentially the same as the Ericksen number, Er, proposed by de Gennes [see P. G. de Gennes, in lectures at l'Ecole d'Eté, Les Houches (to be published)] defined as the ratio of convective transport to diffusive transport of the director orientation. The convenient Ericksen number for our problem is specifically $\mathcal{E} = (\mathrm{sgn}\,\omega')\gamma_1\omega'\bar{R}^{*2}/K_3$, where K_3 is the bend elastic constant, ω' the angular rotation of the inner shaft, and $\bar{R}^*$ the characteristic length. As we use it $\mathcal{E}$ is always positive in order that the viscous and elastic stresses oppose each other.

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Superfluid Density in Porous Vycor Glass*

C. W. Kiewiet and H. E. Hall
Physics Department, Schuster Laboratory, The University, Manchester M13 9PL, England

and

J. D. Reppy
Physics Department, Schuster Laboratory, The University, Manchester M13 9PL, England, and Laboratory of Atomic and Solid State Physics and Materials Science Center, Cornell University, Ithaca, New York 14853

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The superfluid density of ^{4}He confined to porous Vycor glass has been determined from fourth-sound measurements over a temperature range from below 0.1 K to within 10 mK of the transition. For temperatures below 1.4 K the normal-fluid fraction can be described in terms of a roton contribution with gap $\Delta/k_B = 5.85$ K and a "one-dimensional," T^2, phonon contribution. Near the transition temperature, $T_c = 1.955$ K, the superfluid density is found to vary as $(T_c - T)^\zeta$ with $\zeta = 0.65 \pm 0.03$.

The study of helium confined to the narrow (approximately 60 Å diameter) channels of porous Vycor glass[1] provides an excellent opportunity to examine the effect of restricted geometry on the excitations of the liquid and on the character of the superfluid transition. In the work reported here we consider both of these questions.

In the past, extensive determinations have been made of the superfluid density using fourth-sound[2] and persistent-current[3] techniques in both packed-powder and Vycor systems. The temperature for which superflow is first seen, the "onset" temperature, is observed to decrease with decreasing pore size. For onset temperatures on the order of 2 K the fourth-sound measurements in packed powders indicate that the superfluid density approaches zero in a nearly linear fashion.[4] Both the depression of the onset temperature[5] and the temperature dependence of the superfluid density near onset[6] have been discussed in terms of the temperature-dependent healing length of the Ginzburg-Pitaevskii-Mamaladze theory.[7] In this theory the healing length, ξ, is given by $\xi = \xi_0(1 - T/T_\lambda)^{-2/3}$. The experiments indicate that ξ_0 is in the range of 1 to 2 Å. A key assumption for this analysis is the requirement that the or-