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Apparent viscosity during viscometric flow of nematic liquid crystals

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Résumé. — Pour un liquide nématique en cisaillement, il a été montré que l'on peut prédire la forme de la viscosité apparente à la limite des cisaillements élevés. On obtient cette prédiction moyennant deux hypothèses simplificatrices raisonnables. On montre ici que cette même forme pour la viscosité apparente est aussi valable pour d'autres écoulements viscométriques, si la mesure de la force de cisaillement est bien choisie. Les résultats sont en bon accord avec les expériences.

Abstract. — For a nematic in simple shear it has been shown that the asymptotic form of the apparent viscosity for large shearing force can be predicted. This prediction is obtained from two reasonable simplifying assumptions. It is shown here that the same form for the apparent viscosity is also valid for other viscometric flows, provided the appropriate measure of the shearing force is chosen. The results are in good agreement with experiments.

1. Introduction. — MacSithigh and Currie [1] have shown that for a nematic liquid crystal with $\mu_3 < 0$, the apparent viscosity $\eta$ in simple shear (with orientation lying in the plane of shear) has the following asymptotic form for large shearing force:

$$\eta \sim g(\theta_0) \left[ 1 + \text{sgn}(\theta_0 - \theta_1) \xi^{-1} H(\theta_1, \theta_0) \right]^{-1}, \quad (1.1)$$

Here $\xi^2$ is a non-dimensional measure of the shearing force applied to the two plates:

$$\xi^2 = ch^2 / 2k \quad (1.2)$$

where $c$ is the shearing force, $2h$ the gap width and $k$ a typical free energy coefficient. The functions $g$ and $H$ depend on the viscosity coefficients, $\theta_0$ is the Leslie angle and $\theta_1$ the angle of orientation imposed by the strong-anchoring boundary condition.

The description of simple shearing flow given by De Gennes ([2], p. 171) makes plausible the formula (1.1). At large values of $\xi$ the orientation is everywhere close to the Leslie angle $\theta_0$ except near the plates where there are thin boundary layers accommodating the boundary orientation $\theta_1$. De Gennes shows that the thickness of these layers is of order $\xi^{-1}h$. Thus to a first approximation the apparent viscosity $\eta$ is given by the viscosity $g(\theta_0)$ appropriate to the main-stream orientation. Equation (1.1) gives explicitly the first order correction due to the boundary layers.

However, the range of validity of (1.1) will be greater than is suggested by this analysis in terms of boundary layers. The derivation of the formula shows that it will be a good approximation whenever the orientation at the point in the centre of the flow is close to $\theta_0$. This first occurs at values of $\xi$ for which the influence of the boundary orientation is still felt over a substantial portion of the flow.

The purpose of this paper is to show that (1.1) will hold for other viscometric flows provided $\xi$ is defined appropriately. That this is likely to be the case is clear from the boundary-layer descriptions given by De Gennes [2] for plane Poiseuille flow and by Yun and Fredrickson [3] for other flows.

The derivation of (1.1) depends on two simplifying assumptions:

(i) the one-constant approximation for the free energy $k_1 = k_3 = k$;
(ii) the viscosity coefficient $\mu_1 = 0$.

These approximations are justified in [1]. In addition, the Leslie angle $\theta_0$ must exist, i.e. $\mu_3 < 0$. For nematics close to a smectic transition $\mu_3$ is positive [4]. In this case, the present analysis is no longer valid, and the flow patterns are very different [4, 5].

It is assumed that the form of solution adopted here for each flow is stable. Some justification for this
is given by the stability analysis for simple shearing flow [6].

Numerical solutions of the equations governing various flows have been given by various authors [7, 8, 9, 10]. These solutions are valid over the full range of the shearing force. The analytical approach adopted here is complementary to these numerical studies.

2. Plane Poiseuille flow. — A nematic liquid crystal is confined between fixed infinite parallel plates a distance 2\( h \) apart. Cartesian axes are taken with origin half-way between the plates, the \( y \)-axis normal to the plates. The fluid flows in the negative \( x \)-direction driven by a pressure gradient \( c \). The orientation is assumed to lie in the plane of shear. The velocity \( v \) and director \( d \) are taken to have components

\[
\begin{align*}
v_x &= u(y), \quad v_y = v_z = 0, \\
d_x &= \cos \theta, \quad d_y = \sin \theta, \quad d_z = 0, \quad \theta = \theta(y).
\end{align*}
\]

The equations governing \( \theta \) and \( u \) are given by Leslie ([2], eqs. (6.11), (6.13)).

Following MacSithigh and Currie [1] we assume that the bend and splay free energy coefficients are equal (\( k_3 = k_1 = k \)) and that the viscosity coefficient \( \mu_1 = 0 \). Using Parodi’s relation among the viscosity coefficients (\( \mu_6 = \mu_1 + \mu_3 + \mu_2 \) [11, 12]), the equations given by Leslie may then be written

\[
k\theta'' = cy + g(\theta_0)u', \quad u' = -cyg(\theta),
\]

where the function \( g \) and the Leslie angle \( \theta_0 \) are defined by

\[
\begin{align*}
2g(\theta) &= \mu_4 + \mu_5 - \mu_2 + 2(\mu_4 + \mu_5)\cos^2 \theta, \\
2\theta_0 &= \cos^{-1} \left( (\mu_2 - \mu_3)/(2\mu_1 + \mu_3) \right).
\end{align*}
\]

Only materials for which \( \theta_0 \) exists (\( \mu_3 < 0 \)) are considered.

It is assumed that the strong-anchoring boundary conditions permit an odd solution \( \theta(-y) = -\theta(y) \), i.e. the orientation on the plates is arranged to satisfy

\[
\theta(h) \text{ (mod } \pi) = -\theta(-h) \text{ (mod } \pi) = \theta_1,
\]

for some given \( \theta_1 \). Without loss in generality \( \theta_1 \) is taken to satisfy \( \theta_1 \leq \theta_0 < \pi - \theta_0 \), where \( \theta_0 \) is the critical angle introduced in [6] and defined by

\[
(\theta_0 - \theta_0)(\pi/2)^{1/2} = 2g(\theta_0)\tan^{-1}\{\tan \theta_0(\pi/2)^{1/2}\} - \\
- \tan^{-1}\{\tan \theta_0(\pi/2)^{1/2}\}.
\]

with \( \alpha = g(\pi/2), \beta = g(0) \). For small \( \theta_0, \theta_1 \approx -2\theta_0 \).

As suggested by the analysis for simple shearing flow [6], it is assumed that

\[
\theta_1 \ll \theta(y) < \pi - \theta_0. \quad (2.6)
\]

Even with these restrictions, there are still two types of solution satisfying the boundary conditions (2.4), namely

\[
\theta(y) = -\theta(-y) + \pi/2 \pm \pi/2.
\]

With the + sign, \( \theta(0) = \pi/2 \) while with the - sign \( \theta(0) = 0 \). The second of these alternatives seems more likely at high flow rates. Indeed, for radial flow between parallel discs with homeotropic boundary orientation (\( \theta_4 = \pi/2 \)), Hiltrop and Fischer [13] have observed a transition from the first regime to the second with increasing flow rate.

We shall see below that the asymptotic formula is insensitive to the differences between these alternative solutions.

From (2.2) the flux of fluid \( Q \) per unit width of the flow is given by

\[
Q = \int_{-h}^{h} u \, dy = -\int_{-h}^{h} yu' \, dy
\]

\[
= \frac{1}{g(\theta_0)}\int_{-h}^{h} (cy - k\theta') \, dy
\]

\[
= \frac{1}{g(\theta_0)}\left\{ \frac{2ch^3}{3} + 2kh\theta'(h) + k[\theta(h) - \theta(-h)] \right\},
\]

since \( \theta'(h) = \theta'(-h) \). Also, from (2.2)

\[
\frac{1}{2}k[\theta'(h)]^2 = ch\int_{\theta_0}^{\theta_1} \frac{g(\phi) - g(\theta_0)}{g(\phi)} \, d\phi - cy \times
\]

\[
\times \int_{\theta_0}^{\theta_1} \frac{g(\phi) - g(\theta_0)}{g(\phi)} \, d\phi - c \int_{\theta_0}^{\theta_1} \frac{g(\phi) - g(\theta_0)}{g(\phi)} \, d\phi \, dy.
\]

As \( c \to \infty \) it follows from (2.2) that, for \( y > 0 \), \( \theta \to \theta_0 \) everywhere except for a thin boundary layer on the wall and an adjustment layer at the centre. This is observed optically by Gahwiller [14], and discussed by De Gennes ([2], page 171). Take \( y \) in (2.9) to be a position in the main-stream, outside both these layers. Then \( \theta(y) \to \theta_0 \), \( \theta'(y) \to 0 \) as \( c \to \infty \).

Moreover, the third integral in (2.9) becomes negligible compared with the first. Thus

\[
\frac{1}{2}k[\theta'(h)]^2 \sim ch[H(\theta_1, \theta_0)]^2,
\]
where as in [1]

\[
\left( H(\theta_1, \theta_0) \right)^2 = \int_{0}^{\theta_1} \frac{g(\varphi) - g(\theta_0)}{g(\varphi)} \, d\varphi = \frac{2}{A} \tan^{-1} \left\{ \frac{A \tan (\theta_0 - \theta_1)}{2 g(\theta_0) + [\mu_2 + \mu_3] \tan (\theta_0 - \theta_1)} \right\} + \theta_1 - \theta_0 ,
\]

(2.11)

with

\[
A^2 = (\mu_4 + \mu_5 - \mu_2) (\mu_4 + \mu_5 + \mu_2 + 2 \mu_3) .
\]

The apparent viscosity \( \eta = 2 c h^3/3 Q \). Define \( \xi \) by

\[
\xi^2 = c h^3/18 k .
\]

Taking the appropriate root in (2.10), it is found from (2.8) that

\[
\eta^{-1} \sim (g(\theta_0))^{-1} \left\{ 1 - \text{sgn} (\theta_0 - \theta_1) \xi^{-1} H(\theta_1, \theta_0) + \frac{1}{6} \xi^{-2} \theta(\theta) \right\} .
\]

(2.13)

Clearly for large \( \xi \) the behaviour of the apparent viscosity is given by (1.1). The difference between the two alternative solutions (2.7) will not be discernible to first order in \( \xi^{-1} \). The reason for this is that \( u' = 0 \) at \( y = 0 \), by (2.2). It follows from (2.8) that the adjustment layer at the centre of the flow has less influence on \( Q \) than the boundary layers as the pressure gradient increases, and so the precise form of the adjustment layer is unimportant in the asymptotic limit.

### 3. Other flows.

Since (1.1) holds for plane Poiseuille and shearing flows we expect it to hold for similar flows with cylindrical boundaries. The results for Couette flow between concentric cylinders and shearing flow between sliding co-axial cylinders are summarized in the table I. The analysis for these flows is very similar to that for shearing flow. In both, it is assumed that the orientation lies in the plane of shear and adopts the same value on each cylinder.

<table>
<thead>
<tr>
<th>Flow and geometry and boundary orientation</th>
<th>( c )</th>
<th>( \eta )</th>
<th>( \xi^2 )</th>
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<tr>
<td>Simple shearing flow, gap width ( 2 h ), relative velocity of plates ( 2 V ). Same orientation on plates.</td>
<td>shearing force on plates per unit area</td>
<td>( c h/V )</td>
<td>( c h^2/2k )</td>
</tr>
<tr>
<td>Couette flow between concentric cylinders, radii ( R_1 ), ( R_2 ), relative angular speed of cylinders ( \Omega_2 - \Omega_1 ). Same orientation on cylinders.</td>
<td>torque on cylinders per unit length</td>
<td>( c(R_1^{-2} - R_2^{-2})/4\pi(\Omega_2 - \Omega_1) )</td>
<td>( c/16\pi k \left[ R_2^{-2} + R_1^{-2} \right] ^2 )</td>
</tr>
<tr>
<td>Sliding coaxial cylinders, radii ( R_1 ), ( R_2 ), relative velocity of cylinders ( 2V ). Same orientation on cylinders.</td>
<td>axial force on cylinders per unit length</td>
<td>( c \ln (R_2/R_1)/4\pi V )</td>
<td>( c/4\pi k \left[ \ln (R_2/R_1) \right] ^2 )</td>
</tr>
<tr>
<td>Plane Poiseuille flow, gap width ( 2h ), flux of fluid per unit width ( Q ). Opposite orientation on cylinders.</td>
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<td>( 2c h^3/3Q )</td>
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<tr>
<td>Pressure driven flow between co-axial cylinders, radii ( R_1 ), ( R_2 ), volume flux ( Q ). Opposite orientation on cylinders.</td>
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<td>( \frac{\pi c}{8 Q} \left{ R_2^4 - R_1^4 - \frac{(R_2^2 - R_1^2)^2}{\ln (R_2/R_1)} \right} )</td>
<td>see eq. (3.6)</td>
</tr>
<tr>
<td>Poiseuille flow in cylinder radius ( R ), volume flux ( Q ).</td>
<td>axial pressure gradient</td>
<td>( \frac{\pi c R^4}{8 Q} )</td>
<td>( c R^3/64k )</td>
</tr>
</tbody>
</table>
3.1 Pressure-driven flow between co-axial cylinders. — Two co-axial cylinders are held fixed and the nematic flows between them, driven by a pressure gradient $c$ in the negative $z$-direction (where $r$, $\psi$, $z$ are cylindrical polars). The radii are 

$$R_1, R_2 (R_1 < R_2).$$

The velocity and director components are

$$v_z = -u(r), \quad v_r = v_\phi = 0,$$

$$d_z = \cos \theta, \quad d_r = \sin \theta, \quad d_\phi = 0, \quad \theta = \theta(r).$$

(3.1)

The governing equations are given by Atkin [[15], eqs. (3.11), (3.15)]. With the simplifications $k_1 = k_3 = k$, $\mu_1 = 0$ they become

$$k(r^2 \theta'' + r \theta' - \sin \theta \cos \theta) = r^2 u' [g(\theta_0) - g(\theta)],$$

$$u' = -\frac{c(r^2 - 2b)}{2} r g(\theta),$$

(3.2)

where the total axial force required to hold the cylinder $R_2$ fixed is $\pi(cR_2^2 - 2b)$.

As for plane Poiseuille flow, the strong anchoring boundary conditions are assumed to impose the conditions $\theta(R_2) = -\theta(R_1) = \theta$, and $\theta_1$ is restricted by (2.6). From (3.2) the significant terms in the expression for the flux are

$$Q = -\int_{R_1}^{R_2} \pi^2 u' \, dr \sim \pi g(\theta_0) \times \left[ \frac{c r^4}{8} - \frac{b r^2}{2} \theta' \right]_{R_1}^{R_2}. \quad (3.3)$$

Similarly the condition of zero velocity on both walls gives

$$0 = -\int_{R_1}^{R_2} u' \, dr \sim \left[ \frac{c r^2}{4} - b \log r - k \theta' \right]_{R_1}^{R_2}.$$  

(3.4)

An argument similar to that leading to (2.10) shows that

$$k[\theta'(R_2)]^2 \sim (cR_2 - 2b/R_2) [H(\theta_1, \theta_0)]^2,$$

(3.5)

and similarly at $R_1$. If $b$ is now eliminated between (3.4) and (3.5), the linear terms in $\theta'$ arising from (3.4) are found to be insignificant, and hence $\theta'(R_2)$ can be determined. Similarly for $\theta'(R_1)$. Eliminating $b$ between (3.3) and (3.4) and substituting for $\theta'$ we then find that the apparent viscosity

$$\eta\{R_2^4 - R_1^4 - (R_2^2 - R_1^2)^2/ln(R_2/R_1)\} \times \pi c/8 Q$$

is given by (1.1) with

$$\xi^2 = \frac{c}{64k} \left[ \frac{R_2^2 - R_1^2}{\psi(R_2) + \psi(R_1)} \right]^{1/2} - \frac{R_2^{-1/2} - R_1^{-1/2}}{\psi(R_1)}^{1/2}. \quad (3.6)$$

3.2 Poiseuille flow in circular pipe. — It can be verified by independent calculation that the results for a circular pipe can be found by letting $R_1 \to 0$ in (3.6). As noted by Atkin [15], $b = 0$ in this case, and the condition (3.4) is no longer appropriate. The results are summarized in the table I.

4. Conclusion. — We have shown that the asymptotic formula (1.1) holds for a number of viscometric flows, provided $\xi$ is chosen correctly. We emphasize the following points:

a) (1.1) gives the behaviour of $\eta$ only for large $\xi$. It cannot be generally valid. For simple shear flow it seems to be valid for $\xi > 1.5$ [1].

b) At all boundaries there must be the same disposition between the local shear and the boundary orientation. Thus for pressure-driven flows the boundary orientations must be different on opposite boundaries.

It has been assumed in this paper that the orientation will not pass through the critical angle $\theta_c$ (c.f. (2.6)). The justification for this assumption comes from the stability and dissipation analysis given by Currie and MacSithigh [6] for simple shear. This assumption must be regarded as tentative, in the absence of experimental conformation. However, it is clear that the analysis presented here does not depend crucially on this assumption.

It is worth noting that for all these flows some information can be easily obtained merely by reversing the direction of flow. This determines successively $H(\theta_1, \theta_0)$, $H(\pi - \theta_1, \theta_0)$ if $\theta_0 < \theta_1 < \pi + \theta_e$, or $H(\theta_1, \theta_0)$, $H(-\theta_1, \theta_0)$ if $\theta_e < \theta_1 < \theta_0$.

The asymptotic formula (1.1) predicts that $\eta^{-1}$ behaves linearly with $\xi^{-1}$ for large $\xi$. It has been shown [1] that the formula correlates well with numerical solutions of the governing equations. Most published experimental data is for flows in the presence of magnetic fields. Exceptions to this are the experiments of Peter and Peters [16] and Fisher and Fredrickson [17], both for Poiseuille flow of PAA in a capillary. In the experiments of Peter and Peters the boundary was untreated and the orientation unknown.
Nevertheless, from their data it is found that $n^{-1}$ varies linearly with $c^{-1/2}$ as predicted (this correlation is not shown here). In the figure 1 the data of Fisher and Fredrickson for $n^{-1}$ is plotted against $$(4 Q n R)^{-1/2} = (32 k_2)^{-1/2}.$$  

The points close to line A are taken from the universal curve found for various values of the capillary radius for perpendicular boundary orientation ([17], Fig. 7). It is seen that these points lie on a straight line provided $\xi$ is large enough. Curves B and C correspond to data for supposedly parallel orientation ([13], Fig. 8). It has already been argued [18] that this data does not correspond to the same boundary orientation in each case, and hence two lines result. However, they are both straight and pass through the same point on the axis at line A, as required.

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