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F. M. Leslie^a

^a Mathematics Department, Strathclyde University, Livingstone Tower, 26 Richmond St., Glasgow, G 1 1XH, Scotland

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Viscometry of Nematic Liquid Crystals†

F. M. LESLIE

Mathematics Department, Strathclyde University, Livingstone Tower, 26 Richmond St., Glasgow, G1 1XH, Scotland.

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Viscometric experiments have played a significant part in establishing continuum theory for nematic liquid crystals. Such theory supports the hypothesis that observed non-Newtonian behaviour stems from competition between the aligning influences of flow and solid surfaces, and as a consequence predicts rather unusual scaling for the apparent viscosity. This paper first describes such scaling and its subsequent experimental confirmation emphasising the full implications of the latter. The relevance of the theory thus established, there follows a simple analysis of alignment in shear flow which leads to conditions on material coefficients necessary to ensure consistency with observations. Our discussion turns next to an account of solutions exhibiting non-Newtonian behaviour, and also mention of a recent stability analysis which attempts to discriminate between the different solutions that are possible. The final section considers oscillatory shear flow and presents an analysis of an instability which can occur in certain nematics.

1 INTRODUCTION

Viscosity measurements have always played a prominent role in studies of physical properties of liquid crystals, even although a greater variety of dynamic effects is possible in these materials. The review of rheological behaviour by Porter and Johnson¹ written more than ten years ago cites numerous papers concerned with such measurements, and gives an indication of interest in the topic. It is now apparent, however, that the great majority of these earlier viscometric investigations missed features of interest by using too high shear rates and also rather large viscometer dimensions, although they do provide useful data concerning behaviour in these limits. Several more recent experimental studies attempt to correct this by employing more meaningful ranges of shear rates and viscometer dimensions, and also known surface orientations with optical checks upon alignment. Two recent reviews discuss these developments in some detail, one by Jenkins² which is devoted

† Invited lecture, presented at Eighth International Liquid Crystal Conference, Kyoto (Japan), June 30–July 4, 1980.

solely to viscometry of nematics, while the other by Leslie³ attempts to provide a more comprehensive coverage of flow phenomena in general. Given the availability of these detailed accounts, we can therefore be somewhat more selective here and discuss topics of more current interest, or some deserving greater attention than they have received.

Porter, Barrall and Johnson⁴ first suggested some fifteen years ago that non-Newtonian behaviour of nematic liquid crystals might stem from competition between flow and solid boundaries to dictate alignment. Continuum theory commonly employed supports this contention, and moreover predicts an unusual relationship for the apparent viscosity in that it is not simply a function of the relative shear rate. Subsequent experimental confirmation of this prediction largely vindicates several assumptions behind the continuum theory and establishes its relevance. In view of its importance and also because its implications are not widely appreciated, we turn first to this topic. Before progressing to more detailed theoretical predictions, it is helpful to examine initially the influence of shear flow upon alignment in a nematic, other factors such as solid surfaces being insignificant, and we discuss this at some length giving certain results which appear to be new. These preliminaries over, there follows an account of solutions of the continuum equations which demonstrate the non-Newtonian behaviour that arises from the competing influence of flow and viscometer surfaces upon alignment. Here, however, there is a variety of solutions available in even the simplest cases, and therefore we consider next some recent work by Currie and MacSithigh⁵ which limits the possibilities through a stability analysis. Our final section presents a brief description of a flow instability which can occur in oscillatory shear. While the analysis is perhaps over-simplified, it does indicate a new flow induced instability in nematics. A more detailed account of this recent study is given in a paper presented by Clark, Leslie, Saunders and Shanks⁶ at this conference.

It is natural in such a review to assume that the reader is familiar with the physical properties of nematics and also relevant continuum theory. For the former, the books by de Gennes⁷ and Chandrasekhar⁸ provide comprehensive introductions, and also give useful accounts of continuum theory, although fuller reviews of the latter are available in the articles by Ericksen,⁹ Jenkins² and Leslie.³

2 CONTINUUM EQUATIONS

Before examining specific solutions it is of interest to consider a more general property of the continuum equations proposed by Ericksen¹⁰ and Leslie¹¹ for flow phenomena in nematic liquid crystals. Consequently we first sum-

marise the appropriate form of their theory employing Cartesian tensor notation, and proceed to a topic which has not received the attention that it merits.

The aforesaid theory employs two independent vector fields, the velocity \mathbf{v} and an additional kinematic variable \mathbf{n} , a unit vector or director describing alignment of the anisotropic axis. With the assumption of incompressibility, the equations are the constraints

$$v_{i,i} = 0, \quad n_i n_i = 1, \quad (2.1)$$

and balance laws for linear and angular momentum

$$\rho \dot{v}_i = t_{ij,j}, \quad s_{ij,j} + g_i = 0, \quad (2.2)$$

respectively, where ρ is density, \mathbf{t} the stress tensor, \mathbf{s} the director stress tensor, and \mathbf{g} an intrinsic director body force. The superposed dot denotes a material time derivative, a comma preceding a suffix implies partial differentiation with respect to the corresponding spatial coordinate, and repeated indices are subject to the summation convention. Since effects associated with external body forces and couples are beyond the scope of this review, we omit corresponding terms, and also "director inertia" since it is generally believed to be insignificant in the problems discussed.

For nematics, the constitutive relations are

$$\begin{aligned} t_{ij} &= -p\delta_{ij} - \frac{\partial W}{\partial n_{k,j}} n_{k,i} + \tilde{t}_{ij}, \\ s_{ij} &= n_i \beta_j + \frac{\partial W}{\partial n_{i,j}}, \quad g_i = \gamma n_i - (n_i \beta_j)_{,j} - \frac{\partial W}{\partial n_i} + \tilde{g}_i, \end{aligned} \quad (2.3)$$

where p represents an arbitrary pressure arising from the assumed incompressibility, and similarly the scalar γ and vector $\boldsymbol{\beta}$ stem from the constraint imposed upon the director. The form of the energy function W is that due to Oseen¹² and Frank,¹³ and thus

$$\begin{aligned} 2W &= k_1(n_{i,i})^2 + k_2(n_i e_{ijk} n_{k,j})^2 + k_3 n_i n_j n_{k,i} n_{k,j} \\ &\quad + (k_2 + k_4)[n_{i,j} n_{j,i} - (n_{i,i})^2]. \end{aligned} \quad (2.4)$$

Also, the dissipative contributions $\tilde{\mathbf{t}}$ and $\tilde{\mathbf{g}}$ are given by

$$\begin{aligned} \tilde{t}_{ij} &= \alpha_1 A_{kp} n_k n_p n_i n_j + \alpha_2 N_i n_j + \alpha_3 N_j n_i + \alpha_4 A_{ij} \\ &\quad + \alpha_5 A_{ik} n_k n_j + \alpha_6 A_{jk} n_k n_i, \\ \tilde{g}_i &= -\gamma_1 N_i - \gamma_2 A_{ik} n_k, \quad \gamma_1 = \alpha_3 - \alpha_2, \quad \gamma_2 = \alpha_6 - \alpha_5, \\ 2A_{ij} &= v_{i,j} + v_{j,i}, \quad 2N_i = 2\dot{n}_i - (v_{i,k} - v_{k,i})n_k. \end{aligned} \quad (2.5)$$

Here the coefficients in these relationships are simply constants, since we

ignore thermal effects. Ericksen¹⁴ restricts possible values for the k 's by a stability argument, and Leslie¹⁵ limits the range of values of the α 's by the thermodynamic inequality

$$\tilde{t}_{ij}v_{i,j} - \tilde{g}_i\dot{n}_i \geq 0. \quad (2.6)$$

Lastly, one commonly adds the relationship

$$\gamma_2 = \alpha_3 + \alpha_2 \quad (2.7)$$

due to Parodi.¹⁶

Ericksen¹⁷ draws attention to a rather surprising feature of the above equations. If one scales space and time by a transformation

$$\mathbf{x} = h\mathbf{x}^*, \quad t = h^2t^*, \quad (2.8)$$

where h is some constant, the new equations are *identical* to the original provided that one chooses

$$p = h^{-2}p^*, \quad \gamma = h^{-2}\gamma^*. \quad (2.9)$$

In this scaling, the velocity and stress transform according to

$$\mathbf{v}^* = h\mathbf{v}, \quad \mathbf{t}^* = h^2\mathbf{t}, \quad (2.10)$$

but the director being a unit vector is unaltered. In the context of simple shear flow it is natural to choose the constant h to be the gap width. As a result, if V denotes the relative shearing speed, the original problem transforms into one with unit gap width and relative speed V^* , the latter given by

$$V^* = Vh. \quad (2.11)$$

However, the usual strong anchoring boundary condition on the director remains unchanged.

Given a unique solution to the problem, it therefore follows that

$$\mathbf{n} = \mathbf{n}^* = \mathbf{n}^*(\mathbf{x}^*, V^*) = \mathbf{n}^*(h^{-1}\mathbf{x}, Vh), \quad (2.12)$$

and thus one predicts that in simple shear flow an optical property must depend upon the parameter V through the product Vh . Further, introducing an apparent viscosity η as is commonly done by

$$\eta = \frac{\sigma h}{V}, \quad (2.13)$$

where σ is the shear stress applied in the direction of shear, one readily verifies that

$$\eta = \eta^* = \mathcal{F}(V^*) = \mathcal{F}(Vh), \quad (2.14)$$

the function of course unknown. Thus the same simple scaling must occur in

viscosity measurements if the theory applies. Wahl and Fischer¹⁸ find such scaling in optical measurements with a flow closely approximating simple shear, but unfortunately no experimental evidence appears to exist with which to check the latter prediction.

By a somewhat similar analysis for Poiseuille flow, Atkin¹⁹ predicts that the flux per unit time Q from a capillary of radius R is related to the pressure gradient α by

$$Q = R\mathcal{H}(\alpha R^3), \quad (2.15)$$

the function \mathcal{H} unknown. As a consequence, the apparent viscosity η defined in the usual manner satisfies the functional relationship

$$\eta = \mathcal{G}\left(\frac{Q}{R}\right). \quad (2.16)$$

This is also in contrast with the corresponding result for an isotropic liquid where the dependence upon the radius differs. For this flow, however, the experiments of Fisher and Frederickson²⁰ with narrow capillaries verify the prediction of such novel scaling. As Leslie³ discusses in some detail, this confirmation is an important vindication of several assumptions made in the formulation of the above theory. For example, one can readily verify that various generalizations of the above constitutive assumptions, with say gradients of the director or its velocity in the dissipative terms, must lead to predictions in conflict with these experimental observations, and therefore such generalizations are unnecessary at least at present.

3 FLOW ALIGNMENT

Before turning to more complex aspects, it is helpful initially to investigate the influence of flow upon alignment, all other factors being absent. Consequently, we ignore below complications due to surface and other effects, and simply examine rather idealized situations that are likely to improve our understanding of flow alignment in nematics. The resulting analysis is therefore rather similar to that given earlier by Ericksen²¹ in the context of a simpler theory of anisotropic liquids.

In the first instance we seek to determine how the director aligns in simple shear flow, and thus consider solutions in which the velocity and director take the particular forms referred to Cartesian axes

$$\begin{aligned} v_x &= \kappa z, & v_y &= v_z = 0, & \kappa &> 0, & n_x &= \cos \theta \cos \phi, \\ n_y &= \cos \theta \sin \phi, & n_z &= \sin \theta, & \theta &= \theta(t), & \phi &= \phi(t), \end{aligned} \quad (3.1)$$

which are consistent with the constraints (2.1). As indicated the angles θ and

ϕ depend solely upon time t and κ is a positive constant. In this event, the equations of the preceding section reduce to

$$\gamma_1 \frac{d\theta}{dt} + \kappa m(\theta) \cos \phi = 0, \quad \gamma_1 \cos \theta \frac{d\phi}{dt} - \kappa l(\theta) \sin \phi = 0, \quad (3.2)$$

where

$$m(\theta) = \alpha_3 \cos^2 \theta - \alpha_2 \sin^2 \theta, \quad l(\theta) = \alpha_2 \sin \theta, \quad (3.3)$$

the latter employing the Parodi relation (2.7).

Whatever the values of the various coefficients, the Eqs. (3.2) have always a steady solution of the form

$$\theta = 0, \quad \phi = \frac{\pi}{2}, \quad (3.4)$$

which corresponds to uniform alignment normal to the plane of shear. Since the theory does not distinguish between orientations \mathbf{n} and $-\mathbf{n}$, it is of course superfluous to cite the other solution physically equivalent to the above. In addition, provided that

$$\alpha_2 \alpha_3 > 0, \quad (3.5)$$

there are essentially two steady solutions with the director in the plane of shear, namely

$$\theta = \pm \theta_0, \quad \phi = 0, \quad (3.6)$$

the acute angle θ_0 being defined by

$$\tan^2 \theta_0 = \frac{\alpha_3}{\alpha_2}. \quad (3.7)$$

Since the thermodynamic inequality (2.6) requires γ_1 to be positive, two possibilities emerge. Either one has

$$\alpha_2 < \alpha_3 < 0, \quad 0 < \theta_0 < \frac{\pi}{4}, \quad (3.8)$$

or

$$\alpha_3 > \alpha_2 > 0, \quad \frac{\pi}{4} < \theta_0 < \frac{\pi}{2}. \quad (3.9)$$

For consistency with observations of flow alignment at relatively small angles to the streamlines, one must of course select the former option.

In order to examine the behaviour of time dependent solutions of Eqs. (3.2), it is necessary first to obtain an integral relating the angles θ and ϕ . To

this end, one notes that

$$\frac{d\theta}{d\phi} = \frac{d\theta}{dt} \frac{d\phi}{dt} = \frac{\cos^2 \theta (\tan^2 \theta - \alpha_3/\alpha_2)}{\tan \theta \tan \phi}, \quad (3.10)$$

and integration quickly yields

$$\sin^2 \phi = C \left(\tan^2 \theta - \frac{\alpha_3}{\alpha_2} \right), \quad (3.11)$$

where C is a constant. With the aid of this integral, a detailed examination of Eqs. (3.2) shows that all solutions must tend to one of the in-plane solutions (3.6) whenever the condition (3.5) applies, the other steady solutions being unstable. In particular, if α_2 and α_3 satisfy (3.8), the stable solution is that with the positive sign, but, when they satisfy (3.9), it is the other solution that is stable. However, even when α_2 and α_3 do not satisfy the condition (3.5), the out of plane solution (3.4) remains unstable.

Naturally the above is of interest in that it indicates likely behaviour of a nematic in shear flow. However, the related surface forces are in general rather complex, and therefore perhaps not realisable in practice. For example, in the experiments by Pieranski and Guyon²² with alignment out of the plane of shear, transverse flow occurs with apparently no transverse component of surface stress. For this reason, it seems equally appropriate to include transverse flow, and consequently we extend the above discussion to somewhat more general solutions in which

$$\begin{aligned} v_x &= \kappa z, & v_y &= \tau z, & v_z &= 0, & n_x &= \cos \theta \cos \phi, \\ n_y &= \cos \theta \sin \phi, & n_z &= \sin \theta, \end{aligned} \quad (3.12)$$

with κ , τ , θ and ϕ all functions of time only, and in addition require that

$$t_{xz} = X(t), \quad t_{yz} = 0, \quad t_{zz} = Z(t), \quad (3.13)$$

corresponding to a shear stress applied in a given direction. In this event, the Eqs. (3.2) are replaced by

$$\begin{aligned} \gamma_1 \frac{d\theta}{dt} + m(\theta)\xi &= 0, & \gamma_1 \cos \theta \frac{d\phi}{dt} - l(\theta)\zeta &= 0, \\ \xi &= \kappa \cos \phi + \tau \sin \phi, & \zeta &= \kappa \sin \phi - \tau \cos \phi, \end{aligned} \quad (3.14)$$

and one must now determine the flow from the first of Eqs. (2.2). For reasons given by Pieranski, Brochard and Guyon,²³ it seems reasonable to neglect the inertial term in this equation, and thus one readily finds that

$$g(\theta)\xi + m(\theta)\frac{d\theta}{dt} = X \cos \phi, \quad h(\theta)\zeta - l(\theta)\cos \theta \frac{d\phi}{dt} = X \sin \phi, \quad (3.15)$$

where

$$\begin{aligned} 2g(\theta) &= \alpha_4 + (\alpha_3 + \alpha_6)\cos^2 \theta + (\alpha_5 - \alpha_2)\sin^2 \theta + 2\alpha_1 \sin^2 \theta \cos^2 \theta, \\ 2h(\theta) &= \alpha_4 + (\alpha_5 - \alpha_2)\sin^2 \theta. \end{aligned} \quad (3.16)$$

Elimination of the velocity gradients finally yields

$$\gamma_1 G(\theta) \frac{d\theta}{dt} + X m(\theta) \cos \phi = 0, \quad \gamma_1 H(\theta) \cos \theta \frac{d\phi}{dt} - X l(\theta) \sin \phi = 0, \quad (3.17)$$

in which

$$G(\theta) = g(\theta) - \frac{m^2(\theta)}{\gamma_1}, \quad H(\theta) = h(\theta) - \frac{l^2(\theta)}{\gamma_1}. \quad (3.18)$$

As Leslie³ discusses, the functions $G(\theta)$ and $H(\theta)$ are both positive on account of the thermodynamic inequality (2.6).

Comparing Eqs. (3.17) with (3.2), one notes that they differ only in two respects; the stress component X replaces the velocity gradient κ , and the additional factors $G(\theta)$ and $H(\theta)$ appear in the leading terms. Therefore, in this case also, we recover the same steady solutions cited above. Also, proceeding as before, it follows that

$$\frac{d\theta}{d\phi} = -\frac{m(\theta)H(\theta)}{\alpha_2 \tan \theta \tan \phi G(\theta)}, \quad (3.19)$$

and with the substitution

$$u = \tan^2 \theta, \quad (3.20)$$

one ultimately obtains

$$2 \cot \phi \frac{d\phi}{du} = \frac{((u+1)^2 + 2\beta u)}{(u+\varepsilon)(u+\lambda)(u+1)}, \quad (3.21)$$

where the parameters β , ε and λ are defined by

$$\begin{aligned} 2\gamma_1 \eta \beta &= \gamma_2^2 + \alpha_1 \gamma_1, & \alpha_2 \varepsilon &= -\alpha_3, & 2\eta \lambda &= \alpha_4, \\ 2\eta \gamma_1 &= \gamma_1(\alpha_4 + \alpha_3 + \alpha_6) - 2\alpha_3^2. \end{aligned} \quad (3.22)$$

Since η coincides with the value of $G(\theta)$ when θ is zero, it is therefore positive, and thus λ is also positive. The integral of (3.21) depends of course upon the values of λ and ε and the following cases arise:

i) when $\lambda \neq \varepsilon$, $\lambda \neq 1$, $\varepsilon \neq 1$,

$$\begin{aligned} \sin^2 \phi &= C(1 + \tan^2 \theta)^k (\varepsilon + \tan^2 \theta)^l (\lambda + \tan^2 \theta)^{1-k-l}, \\ k &= \frac{2\beta}{(1-\varepsilon)(\lambda-1)}, & l &= \frac{((1-\varepsilon)^2 - 2\beta\varepsilon)}{(1-\varepsilon)(\lambda-\varepsilon)}, \end{aligned} \quad (3.23)$$

ii) when $\lambda = \varepsilon \neq 1$,

$$\sin^2 \phi = C(\lambda + \tan^2 \theta)^{m+1}(1 + \tan^2 \theta)^{-m} \exp \left[\frac{n \tan^2 \theta}{(\lambda + \tan^2 \theta)} \right],$$

$$m = \frac{2\beta}{(\lambda - 1)^2}, \quad n = \frac{2\beta}{(\lambda - 1)} - \frac{(\lambda - 1)}{\lambda}; \quad (3.24)$$

iii) when $\varepsilon = 1$, $\lambda \neq 1$, (similarly $\lambda = 1$, $\varepsilon \neq 1$),

$$\sin^2 \phi = C(1 + \tan^2 \theta)^s(\lambda + \tan^2 \theta)^{1-s} \exp(-r \sin^2 \theta),$$

$$r = \frac{2\beta}{(\lambda - 1)}, \quad s = \frac{2\beta\lambda}{(\lambda - 1)^2}; \quad (3.25)$$

iv) when $\varepsilon = \lambda = 1$,

$$\sin^2 \phi = C(1 + \tan^2 \theta) \exp(\beta \sin^4 \theta). \quad (3.26)$$

In all cases C is a constant of integration. With the aid of these integrals, one can again make identical statements concerning the stability of the steady solutions. Consequently, even in this more general case our conclusions are exactly as before.

While the above is perhaps too idealized, it does give insight into likely behaviour in more complex situations, and also indicates restrictions upon the viscous coefficients α_2 and α_3 necessary to ensure agreement with observed flow alignment. Another reason for the inclusion of the latter more general solution becomes apparent in the final section.

4 SIMPLE SHEAR FLOW

The simplest solution of the continuum equations for shear flow of a nematic is that with the anisotropic axis uniformly aligned normal to the plane of shear. From the discussion of the previous section this orientation is compatible with the flow, and also with possible prescribed surface alignments. However, as one might anticipate from the behaviour of time dependent solutions in shear flow, this particular configuration becomes unstable when the shear torques are sufficiently strong to overcome the influence of the solid surface. Pieranski and Guyon²⁴ were first to appreciate this flow induced instability, demonstrating it experimentally, and subsequently various investigators have examined this phenomenon in some detail. Consequently, we do not consider it further here, but refer the reader to the reviews by Jenkins² and Leslie³ and the more recent papers by Manneville^{25,26} for further details and references.

Instead we elect to discuss solutions which exhibit directly the non-Newtonian behaviour that arises from competition between flow and

boundaries to dictate alignment of the anisotropic axis. One reason for this choice is that the topic is rather more complicated than is widely appreciated, and secondly there has been some recent relevant work of interest. Our discussion naturally requires that the appropriate viscous coefficients satisfy the condition (3.5) in order that flow alignment occur, and here for definiteness we assume that they satisfy the inequalities (3.8). However, consideration of the case of nematics that do not align in shear is beyond the scope of this paper, chiefly because less progress has been made with these problems.

Other solutions of the continuum equations for simple shear flow take the two-dimensional forms

$$\begin{aligned} v_x &= u(z), & v_y &= v_z = 0, & p &= p(z), \\ n_x &= \cos \theta(z), & n_y &= 0, & n_z &= \sin \theta(z), \end{aligned} \quad (4.1)$$

which clearly satisfy the constraints (2.1). The first of the balance laws (2.2) determines the pressure p and also yields

$$t_{xz} = g(\theta)u' = \alpha, \quad (4.2)$$

where the prime denotes differentiation with respect to z , the function $g(\theta)$ is defined in equations (3.16), and α is a positive constant. The second of Eqs. (2.2) reduces to

$$2f(\theta)\theta'' + \frac{d}{d\theta}f(\theta)\theta'^2 = 2m(\theta)u', \quad (4.3)$$

with

$$f(\theta) = k_1 \cos^2 \theta + k_3 \sin^2 \theta \quad (4.4)$$

and the function $m(\theta)$ as in Eqs. (3.3). If one combines the above and integrates, it follows that

$$f(\theta)\theta'^2 = 2\alpha \int_{\theta_m}^{\theta} \frac{m(\phi)}{g(\phi)} d\phi = \alpha F(\theta, \theta_m), \quad (4.5)$$

where θ_m is an arbitrary constant. On account of the inequalities proposed by Ericksen,¹⁴ the left hand side of this last equation is always positive and this places some restrictions on possible solutions.

To fix ideas let us initially consider the case of parallel alignment and assume that at the solid surfaces

$$\theta(\ell) = \theta(-\ell) = 0, \quad (4.6)$$

ℓ denoting half the gap width. For this surface alignment, equation (4.5) has a simple symmetric solution in which θ increases monotonically to a maximum value θ_m at the centre of the gap that is less than the flow alignment

angle θ_0 , defined by Eq. (3.7). Thus one has

$$\theta(z) = \theta(-z), \quad 0 \leq \theta \leq \theta_m < \theta_0, \quad \theta_m = \theta(0), \quad (4.7)$$

and integration of Eq. (4.5) yields

$$a^{1/2}|z| = \int_{\theta}^{\theta_m} \left[\frac{f(\phi)}{F(\phi, \theta_m)} \right]^{1/2} d\phi, \quad (4.8)$$

from which follows that

$$a^{1/2}h = \int_0^{\theta_m} \left[\frac{f(\phi)}{F(\phi, \theta_m)} \right]^{1/2} d\phi. \quad (4.9)$$

This last expression provides a rather involved equation for the angle θ_m in terms of the parameters a and h . Rather clearly, when the shear stress or gap width is sufficiently small, θ_m is close to the boundary value, but with increasing stress or gap width θ_m increases to θ_0 , at which value the integral becomes singular. Hence this solution is possible for all shear stresses and gap widths.

With the familiar non-slip assumption, the boundary conditions for the flow are

$$u(h) = V, \quad u(-h) = 0, \quad (4.10)$$

V a given constant, and one can readily integrate (4.2) to obtain

$$V = a \int_{-h}^h \frac{dz}{g(\theta)} = 2a \int_0^h \frac{dz}{g(\theta)}. \quad (4.11)$$

However, by inversion of the relationships (4.8) and (4.9) it follows that

$$\theta = \hat{\theta}(a^{1/2}|z|, \theta_m), \quad \theta_m = \mathcal{F}(ah^2), \quad (4.12)$$

and hence (4.11) yields

$$Vh = 2ah \int_0^h \frac{dz}{g[\hat{\theta}(a^{1/2}z, \theta_m)]} = 2a^{1/2}h \int_0^{a^{1/2}h} \frac{ds}{g[\hat{\theta}(s, \theta_m)]} = \mathcal{G}(ah^2). \quad (4.13)$$

Also, defining an apparent viscosity as before, it follows that

$$\eta = \frac{ah}{V} = \frac{ah^2}{Vh} = \frac{\mathcal{G}^{-1}(Vh)}{Vh} = \mathcal{H}(Vh), \quad (4.14)$$

as predicted earlier. For sufficiently large values of the product Vh , the apparent viscosity essentially takes the value $g(\theta_0)$.

However, as Currie²⁷ discusses, an examination of the associated phase-plane diagram shows that the above solution of Eq. (4.5) is not unique. For example, if one assumes a phase-plane diagram similar to that given by

Currie and MacSithigh⁵ and defines angles θ_c and θ_a by

$$F(\theta_0, \theta_c) = 0, \quad F(0, \theta_a) = 0, \quad \theta_0 - \pi < \theta_c < \theta_a < -\theta_0, \quad (4.15)$$

there is in general a second symmetric solution subject to the boundary conditions (4.6) in which θ decreases monotonically to a minimum value θ_m at the centre of the gap, and

$$\theta_m \leq \theta \leq 0, \quad \theta_c \leq \theta_m \leq \theta_a. \quad (4.16)$$

While this type of solution is available only for a rather limited range of values of applied stress and gap width, one can also have in such cases a further symmetric solution with θ decreasing monotonically to a minimum value θ_m , but in this case

$$\theta_0 - \pi < \theta_m < \theta_c. \quad (4.17)$$

This type of solution can occur once the shear stress or gap width is sufficiently large, and remains an option for larger values. Somewhat similarly, other symmetric, monotonically decreasing solutions become possible for sufficiently large shear stress and gap width, and in these

$$\theta_0 - (r + 1)\pi < \theta_m < \theta_c - r\pi, \quad (4.18)$$

where r is a positive integer. In addition, one can construct more complex symmetric and non-symmetric solutions by essentially combining our original solution (4.7) with others of the type (4.16).

It is of course compatible with parallel surface alignment to relax the boundary conditions (4.6) to

$$\theta(\ell) = m\pi, \quad \theta(-\ell) = n\pi, \quad (4.19)$$

where m and n are arbitrary integers. This permits an even greater variety of non-symmetric solutions, some formed by extending one end of the range of θ in solutions of the types (4.17) and (4.18) by an integral multiple of π , and others simply comprising monotonic rotations of θ through arbitrary integral multiples of π .

To discuss other surface alignments, it is convenient to consider separately two distinct cases,

$$\theta_c < \theta_s < \theta_0 \quad (4.20)$$

and

$$\theta_0 - \pi \leq \theta_s \leq \theta_c, \quad (4.21)$$

where θ_s denotes the prescribed surface value, and there is no loss of generality in so restricting the value. For the former range of values, the multiplicity of solutions is exactly as described above. However, for the latter, the number

of possibilities is somewhat reduced. When the surface orientation satisfies (4.21), there is no counterpart to the solution (4.7), nor to those of the type corresponding to (4.16). Thus symmetric solutions are similar to those represented by conditions (4.17) and (4.18), and non-symmetric solutions require relaxation of the boundary conditions as in the previous paragraph. While it is possible to consider cases in which the prescribed surface alignments differ, we do not attempt such a discussion here.

To discriminate between the above solutions, Currie and MacSithigh⁵ examine their stability, and therefore consider perturbations of the original solutions (4.1) of the form

$$\begin{aligned} v_x &= u(z) + U(z)\exp(-\omega t), & v_y &= V(z)\exp(-\omega t), & v_z &= 0, \\ n_x &= \cos \theta \cos \phi, & n_y &= \sin \phi, & n_z &= \sin \theta \cos \phi, \\ \theta &= \theta(z) + \Psi(z)\exp(-\omega t), & \phi &= \Phi(z)\exp(-\omega t), \end{aligned} \quad (4.22)$$

where the functions $U(z)$, $V(z)$, $\Psi(z)$ and $\Phi(z)$ are infinitesimal and ω is a constant. Not unreasonably, they discuss the special case in which the material coefficients satisfy

$$k_1 = k_2 = k_3 = k, \quad \alpha_1 = 0, \quad (4.23)$$

this presumably giving some indication of likely behaviour in general. With these simplifications, the equations for the perturbations Ψ and Φ prove to be

$$\begin{aligned} k\Psi'' - a \frac{d}{d\theta} \left(\frac{m(\theta)}{g(\theta)} \right) \Psi + \frac{\gamma_1 \omega G(\theta) \Psi}{g(\theta)} &= 0, \\ k\Phi'' + (k\theta'^2 + aP(\theta))\Phi + \frac{\gamma_1 \omega H(\theta) \Phi}{h(\theta)} &= 0, \end{aligned} \quad (4.24)$$

where

$$P(\theta) = \sin 2\theta \frac{(\gamma_2 + \alpha_2(\alpha_3 + \alpha_6)/2h(\theta))}{2g(\theta)}, \quad (4.25)$$

and otherwise our notation is that of the previous section. The perturbations are of course subject to the boundary conditions

$$\Psi(\pm \ell) = \Phi(\pm \ell) = 0. \quad (4.26)$$

Given the complex nature of the coefficients in the above equations, Currie and MacSithigh invoke the variational form of this eigen value problem, which leads them to consider two integrals

$$I_1 = \int_{-\ell}^{\ell} \left[k\Psi'^2 + a \frac{d}{d\theta} \left(\frac{m(\theta)}{g(\theta)} \right) \Psi^2 \right] dz, \quad (4.27)$$

and

$$I_2 = \int_{-h}^h [k\Phi'^2 - (k\theta'^2 + \alpha P(\theta)\Phi^2)] dz, \quad (4.28)$$

where Ψ and Φ are any two functions subject to the boundary conditions (4.26). If either of these integrals turns out to be negative for a suitable choice of the test functions Ψ and Φ , this implies a negative eigen value ω , and hence instability of the solution under consideration.

In this way Currie and MacSithigh are able to discount many of the above solutions, but cannot narrow the options down to a unique solution. Nevertheless, they proceed to select a preferred solution from those that remain by choosing that with least viscous dissipation as the most likely to occur in practice. The solution with this property depends upon the prescribed surface alignment, being the counterpart of our original solution (4.7) when (4.20) applies, but that corresponding to the condition (4.17) for surface alignments in the range (4.21). This suggests the likelihood of an interesting transition as the surface alignment passes through the value θ_c . If it is just greater than this value, θ increases monotonically, but, when it is less, θ decreases monotonically. As Currie and MacSithigh remark, it would be of interest if such a transition is observed experimentally.

5 OSCILLATORY SHEAR FLOW

In this final section we present a brief account of a preliminary analysis of the response of a nematic to oscillatory shear flow. Our approach is similar to that of Section 3, and thus concentrates upon an assessment of the influence of flow upon alignment assuming that that of the solid surfaces is of secondary importance. The measure of agreement between our predictions and related experimental observations suggests that our rather simplistic calculation may contain the essential features of the problem discussed.

Here we consider a nematic confined between parallel plates with its initial alignment uniform and parallel to the solid surfaces, and subject to oscillatory shear produced by one plate executing sinusoidal oscillations of amplitude a and frequency ω parallel to the initial alignment, while the other remains at rest. With an appropriate choice of Cartesian axes, it is therefore of interest to consider solutions of the continuum equations in which

$$\begin{aligned} v_x &= \kappa(t)z, & v_y &= v_z = 0, & \kappa(t) &= \frac{a\omega \cos \omega t}{\ell}, \\ n_x &= \cos \theta(t), & n_y &= 0, & n_z &= \sin \theta(t), \end{aligned} \quad (5.1)$$

where h denotes the gap width. Rather clearly, the forms selected ignore complications arising from non-uniformity of alignment due to surface effects. With this choice, the equations of Section 2 reduce to

$$\gamma_1 \frac{d\theta}{dt} + \kappa(t)m(\theta) = 0, \quad (5.2)$$

the function $m(\theta)$ as in Eqs. (3.3). For reasons mentioned earlier, it also seems reasonable to neglect inertial effects in this calculation. The Eq. (5.2) may be re-expressed in a more convenient form

$$\frac{d\theta}{dt} = -(\sin^2 \theta + \varepsilon \cos^2 \theta) A \omega \cos \omega t, \quad A = \frac{|\alpha_2| a}{\gamma_1 h}, \quad (5.3)$$

where ε is as defined in equations (3.22) and α_2 is assumed negative.

Consistent with the initial parallel alignment, the appropriate solution of the above equation is either

$$\tan \theta = \delta \tanh(\delta A \sin \omega t), \quad \delta^2 = |\varepsilon|, \quad (5.4)$$

when α_2 and α_3 satisfy (3.8), or

$$\tan \theta = -\delta \tan(\delta A \sin \omega t), \quad \delta^2 = \varepsilon, \quad (5.5)$$

otherwise, in both the zero of time coinciding with θ zero. In the above, the angle θ oscillates about its initial value with an amplitude independent of frequency but increasing with that of the plate. When the material aligns in shear, the amplitude has an upper limit, but, when flow alignment is not possible, the oscillations increase without bound. In this latter case, however, when δ is small compared with unity, the amplitude remains small until that of the plate approaches a critical value a_c given by

$$a_c = \frac{\gamma_1 h \pi}{2\delta |\alpha_2|}, \quad (5.6)$$

when it increases rather sharply.

To examine the stability of the above Clark, Leslie, Saunders and Shanks⁶ consider solutions of the form (3.12), but assume that the angle ϕ and the transverse flow gradient τ are infinitesimal, and further that the transverse component of stress is zero. On account of these assumptions, they retain the equation (5.2) or (5.3) for the angle θ and consequently the solutions (5.4) or (5.5). Moreover, with the aid of essentially the results (3.23–26), they obtain the corresponding solutions for the angle ϕ .

In order to simplify details, let us consider cases in which

$$\lambda \neq 1, \quad |\varepsilon| \ll 1, \quad (5.7)$$

and the relationship (3.23) leads to

$$\frac{n_y^2}{(n_y^2)_{t=0}} = (1 + \tan^2 \theta)^{k-1} \left(1 + \frac{\tan^2 \theta}{\varepsilon}\right)^l \left(1 + \frac{\tan^2 \theta}{\lambda}\right)^{1-k-l}, \quad (5.8)$$

$$k = \frac{2\beta}{(\lambda - 1)}, \quad l = \frac{1}{\lambda}.$$

For nematics that align in shear, one therefore finds

$$\frac{n_y^2}{(n_y^2)_{t=0}} = [1 - \tanh^2(\delta A \sin \omega t)]^l, \quad (5.9)$$

and from this it follows that a small deviation of the anisotropic axis from the plane of shear remains small throughout the cycle in such materials. On the other hand, if α_3 is positive, the corresponding result is

$$\frac{n_y^2}{(n_y^2)_{t=0}} = \left[\frac{1 + T^2}{1 + \varepsilon T^2/\lambda} \right]^l \left[\frac{1 + \varepsilon T^2}{1 + \varepsilon T^2/\lambda} \right]^{k-1}, \quad T = \tan(\delta A \sin \omega t), \quad (5.10)$$

and in this case a perturbation of the anisotropic axis from the plane of shear can increase considerably. For example, if the amplitude of the plate's oscillation approaches the critical value (5.6),

$$\max \left[\frac{n_y^2}{(n_y^2)_{t=0}} \right] \sim \varepsilon^{-1/2}, \quad (5.11)$$

indicating that deviations from the shear plane increase sharply with the angle θ near this critical value. It seems reasonable to interpret this behaviour as a flow instability. A rather similar phenomenon can occur when the parameter ε is large and positive, but in this case the threshold is less pronounced. These predictions are in rather good agreement with experimental observations described by Clark, Leslie, Saunders and Shanks,⁶ and we refer readers to that paper for fuller details.

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