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Some Topics in Continuum Theory of Nematics

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Phil. Trans. R. Soc. Lond. A 1983 **309**, 155-165
doi: 10.1098/rsta.1983.0030

Phil. Trans. R. Soc. Lond. A **309**, 155–165 (1983)
Printed in Great Britain

Some topics in continuum theory of nematics

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This paper presents a concise formulation of continuum theory for nematic liquid crystals, both static and dynamic theory being discussed in turn. The emphasis is on the various assumptions behind the theory, and the experimental evidence supporting them. The resulting theory contains a fair number of material parameters, and progress concerning their measurement is assessed. Also, whenever possible, mention is made of problems that remain unresolved.

1. Introduction

My primary aim in this paper is to present a concise but clear derivation of continuum equations commonly employed to describe static and dynamic phenomena in nematic liquid crystals. My starting point is a formulation of static theory due to Ericksen (1962), which clarifies mechanical aspects and leads naturally to dynamic theory as a logical development of the well established static theory. To keep the presentation reasonably brief attention is confined to mechanical behaviour, and thermal considerations are largely excluded. Whenever possible, my presentation attempts to bring out experimental evidence to motivate certain assumptions made in the theory, as well as to serve as a means of either justifying or rejecting the theory.

It is now fairly widely accepted that the continuum theory discussed in this paper describes the mechanical response of nematic liquid crystals reasonably well. This belief stems largely from a number of analyses based on its linearized form which satisfactorily interpret various experimental observations. For example, one can cite in this context light-scattering, flow-induced instabilities, thermal convection and electrohydrodynamic instabilities; accounts of these topics are readily available in the books by de Gennes (1974) and Chandrasekhar (1977), and the reviews by Stephen & Straley (1974), Jenkins (1978) and Leslie (1979). Here, however, my concern is to show that the theory has the correct *nonlinear* form, and therefore my attention is mainly given to predictions and related experimental evidence in support of this claim.

First attempts to measure the various material parameters in the theory led to a fair spread of values in many cases. There were of course certain reasons for this; for example some of the methods of measurement were not wholly satisfactory, or impurities may have been present in the materials studied, or some of the nematics were simply chemically unstable. These factors had the unfortunate effect of encouraging a greater tolerance of experimental error than might normally have been acceptable. However, recently, with more stable materials and better methods, improved results are emerging, and it is of interest to discuss such developments where appropriate.

To present the continuum equations concisely it is convenient to use Cartesian tensor notation from time to time so that repeated indices are subject to the summation convention, a comma followed by an index denotes partial differentiation with respect to the corresponding spatial coordinate, and a superposed dot indicates a material time derivative.

2. Static theory

Formulation of the theory

Oseen (1925) was the first to propose a continuum theory to model equilibrium configurations of liquid crystals. He employed a unit vector field or director $\boldsymbol{n}(\boldsymbol{x})$ to describe the alignment of the anisotropic axis in these transversely isotropic liquids, and assumed that spatial distortions of this axis give rise to an energy density of the form

$$W = W(\boldsymbol{n}, \nabla \boldsymbol{n}), \tag{2.1}$$

quadratic in the gradients of the vector. His theory was later reappraised by Frank (1958), who gave an improved derivation of the energy function, essentially assuming invariance to superposed rigid rotations and to the sense of the director $\boldsymbol{n}$, so that

$$W(\boldsymbol{n}, \nabla \boldsymbol{n}) = W(\boldsymbol{Q}\boldsymbol{n}, \boldsymbol{Q} \nabla \boldsymbol{n}\, \boldsymbol{Q}^T) = W(-\boldsymbol{n}, -\nabla \boldsymbol{n}), \tag{2.2}$$

the second-order tensor $\boldsymbol{Q}$ being proper orthogonal. In nematics, material symmetry further restricts the functional dependence by the inclusion of improper transformations in the above condition. Hence, if the dependence upon gradients is at most quadratic, one finds for nematics that

$$\begin{aligned} 2W = {} & k_{11}(\operatorname{div} \boldsymbol{n})^2 + k_{22}(\boldsymbol{n} \cdot \operatorname{curl} \boldsymbol{n})^2 + k_{33}(\boldsymbol{n} \times \operatorname{curl} \boldsymbol{n}) \cdot (\boldsymbol{n} \times \operatorname{curl} \boldsymbol{n}) \\ & + (k_{22} + k_{24}) \operatorname{div}\{(\boldsymbol{n} \cdot \operatorname{grad}) \boldsymbol{n} - (\operatorname{div} \boldsymbol{n}) \boldsymbol{n}\}, \end{aligned} \tag{2.3}$$

the coefficients being the familiar Frank constants, which are at most functions of temperature. One approach commonly followed obtains equilibrium configurations by considering the Euler–Lagrange equations for the minimization of this energy, a further contribution being included to describe the influence of external magnetic or electric fields if present. The boundary conditions generally adopted assume that the solid surfaces confining the liquid crystal dictate a particular known orientation to the anisotropic axis at the surfaces, this depending upon their prior treatment.

It is of interest to interpret the above in mechanical terms, this being helpful for the development of a theory to describe flow and other dynamic effects. To this end, Ericksen (1961) postulates a principle of virtual work of the form

$$\delta \int_V W \, \mathrm{d}v = \int_V (\boldsymbol{F} \cdot \delta \boldsymbol{x} + \boldsymbol{G} \cdot \Delta \boldsymbol{n}) \, \mathrm{d}v + \int_{\partial V} (\boldsymbol{t} \cdot \delta \boldsymbol{x} + \boldsymbol{s} \cdot \Delta \boldsymbol{n}) \, \mathrm{d}a, \tag{2.4}$$

where

$$\Delta \boldsymbol{n} = \delta \boldsymbol{n} + (\delta \boldsymbol{x} \cdot \operatorname{grad}) \boldsymbol{n}. \tag{2.5}$$

Here V denotes a volume of liquid crystal with surface ∂V, $\boldsymbol{F}$ and $\boldsymbol{t}$ are body and surface forces, and $\boldsymbol{G}$ and $\boldsymbol{s}$ generalized body and surface forces, respectively. Since incompressibility is assumed and $\boldsymbol{n}$ remains a unit vector, the variation in displacement $\delta \boldsymbol{x}$ and the material variation in the director $\Delta \boldsymbol{n}$ are subject to the constraints

$$\operatorname{div} \delta \boldsymbol{x} = 0, \quad \boldsymbol{n} \cdot \Delta \boldsymbol{n} = 0. \tag{2.6}$$

As Ericksen (1970) has shown, consideration of an arbitrary, infinitesimal, rigid translation in which $\Delta \boldsymbol{n}$ is zero leads to the somewhat obvious balance of forces

$$\int_V \boldsymbol{F} \, \mathrm{d}v + \int_{\partial V} \boldsymbol{t} \, \mathrm{d}a = 0. \tag{2.7}$$

However, similar consideration of an arbitrary, infinitesimal, rigid rotation $\boldsymbol{\omega}$, in which

$$\delta \boldsymbol{x} = \boldsymbol{\omega} \times \boldsymbol{x}, \quad \Delta \boldsymbol{n} = \boldsymbol{\omega} \times \boldsymbol{n}, \tag{2.8}$$

leads to a balance of couples

$$\int_V (\boldsymbol{x} \times \boldsymbol{F} + \boldsymbol{n} \times \boldsymbol{G})\, \mathrm{d}v + \int_{\partial V} (\boldsymbol{x} \times \boldsymbol{t} + \boldsymbol{n} \times \boldsymbol{s})\, \mathrm{d}a = 0. \tag{2.9}$$

Consequently the generalized forces are related to a body couple $\boldsymbol{K}$ and a couple stress $\boldsymbol{l}$ through

$$\boldsymbol{K} = \boldsymbol{n} \times \boldsymbol{G}, \quad \boldsymbol{l} = \boldsymbol{n} \times \boldsymbol{s}. \tag{2.10}$$

With the usual tetrahedron argument, (2.7) and (2.9) become in point form

$$t_i = t_{ij} \nu_j, \quad F_i + t_{ij,j} = 0, \tag{2.11}$$

and

$$l_i = l_{ij} \nu_j, \quad K_i + e_{ijk} t_{kj} + l_{ij,j} = 0, \tag{2.12}$$

respectively, $\boldsymbol{\nu}$ denoting the unit surface normal and e_{ijk} being the alternating tensor. Also, returning to (2.4) one finds that

$$t_{ij} = -p\delta_{ij} - (\partial W/\partial n_{k,j})\, n_{k,i}, \quad l_{ij} = e_{ipk} n_p\, \partial W/\partial n_{k,j}, \tag{2.13}$$

where p is an arbitrary pressure arising from the assumed incompressibility, and δ_{ij} is the familiar Kronecker delta. These latter results entail some manipulation of the left-hand side of (2.4), and details are available in Ericksen's paper.

Alternatively, however, if one rewrites the couple stress tensor l_{ij} as

$$l_{ij} = e_{ipk} n_p s_{kj}, \quad s_{ij} = n_i \beta_j + \partial W/\partial n_{i,j}, \tag{2.14}$$

where $\boldsymbol{\beta}$ is an arbitrary vector, (2.12) with (2.10) becomes

$$e_{ijk} n_j (G_k + s_{kp,p}) + e_{ijk} (t_{kj} + s_{kp} n_{j,p}) = 0. \tag{2.15}$$

Hence it is possible to introduce a vector $\boldsymbol{g}$ defined by

$$e_{ijk} n_j g_k = e_{ijk} (t_{kj} + s_{kp} n_{j,p}), \tag{2.16}$$

and the balance of couples (2.9) takes the alternative point form

$$G_i + g_i + s_{ij,j} = 0, \tag{2.17}$$

since from its definition $\boldsymbol{g}$ is indeterminate to the extent of an arbitrary scalar multiple of the director $\boldsymbol{n}$. Moreover, the invariance assumption (2.2) yields the following identity

$$e_{ijk} (n_j\, \partial W/\partial n_k + n_{j,p}\, \partial W/\partial n_{k,p} + n_{p,j}\, \partial W/\partial n_{p,k}) = 0, \tag{2.18}$$

first noted by Ericksen (1961). This, combined with (2.13), (2.14) and the definition of the intrinsic body force $\boldsymbol{g}$, gives

$$g_i = \gamma n_i - (n_i \beta_j)_{,j} - \partial W/\partial n_i, \tag{2.19}$$

with γ an arbitrary multiplier.

As for example Ericksen (1962) discusses, the body force and couple arising from an external magnetic or electric field can be expressed in the form

$$F_i = \partial \psi/\partial x_i, \quad G_i = \partial \psi/\partial n_i, \tag{2.20}$$

where for a magnetic field $\boldsymbol{H}$

$$2\psi = 2\psi_{\mathrm{m}} = \chi_\perp \boldsymbol{H} \cdot \boldsymbol{H} + \chi_{\mathrm{a}} (\boldsymbol{n} \cdot \boldsymbol{H})^2, \tag{2.21}$$

and for an electric field $\boldsymbol{E}$

$$2\psi = 2\psi_e = \epsilon_\perp \boldsymbol{E}\cdot\boldsymbol{E} + \epsilon_a(\boldsymbol{n}\cdot\boldsymbol{E})^2, \tag{2.22}$$

the coefficients χ and ϵ being material parameters. This being so, Ericksen shows that the balance of forces (2.11) integrates with the aid of (2.13), (2.14), (2.17) and (2.19) to yield

$$p + W - \psi = p_0 \tag{2.23}$$

where p_0 is an arbitrary constant. Also, the balance of couples (2.17) becomes

$$(\partial W/\partial n_{i,j})_{,j} - \partial W/\partial n_i + \gamma n_i + G_i = 0, \tag{2.24}$$

the generalized body force having the respective forms

$$G_i = \partial\psi_m/\partial n_i = \chi_a n_j H_j H_i, \quad G_i = \partial\psi_e/\partial n_i = \epsilon_a n_j E_j E_i, \tag{2.25}$$

should a magnetic or electric field be present. Clearly, therefore, Ericksen's formulation identifies the Euler–Lagrange equation mentioned above with a local balance of couples in the liquid crystal.

Freedericksz transitions and Frank constants

The first experiments to be candidates for analysis in terms of the above theory were conducted by Freedericksz & Zolina (1933). These experiments require that one first uniformly align a thin layer of nematic liquid crystal between parallel plates by suitable surface treatments, and then apply a magnetic (or electric) field perpendicular to this initial alignment. For small field strengths, the influence of the surfaces dominates, but as the field strength increases a critical value is reached beyond which the field becomes the stronger influence and distortion occurs. In their experiments, Freedericksz & Zolina found that the critical field strength varies inversely with distance between the plates for a given nematic. Other variations of the experiment are possible, but in all cases that use parallel plates the critical field strength is apparently inversely proportional to the layer thickness for a given material (see, for example, Ericksen (1976) or Deuling (1978)).

For such problems the above theory reduces to an analysis of solutions of (2.24) subject to some prescribed alignment at the boundaries, with of course the function W given by (2.3). Rather clearly, if one introduces dimensionless variables by

$$\boldsymbol{x} = l\boldsymbol{x}^*, \quad \boldsymbol{H} = H\boldsymbol{h}, \tag{2.26}$$

and suitably scales the multiplier γ, the field strength H, assumed constant, and the gap width l enter the problem solely through the product Hl, and therefore the predicted scaling necessarily agrees with that observed. Thus this simple exercise supports the choice of the quadratic energy (2.3), since presumably any additional nonlinearity in this function must lead to some predictions at odds with observations. Equally, the choice of boundary conditions and body couple is apparently vindicated.

Detailed calculations lead to precise predictions of the critical field strengths, these naturally involving the coefficients in the energy function (2.3). Not surprisingly, this led initially to attempts to evaluate these material parameters from experimental measurements of the critical field strengths. However, determination of the precise point at which distortion of the alignment commences is not an easy matter, and extrapolations from data above the thresholds can present difficulties, as Tough & Raynes (1979) discuss. Consequently, more recent attempts to evaluate these material parameters use measurements over a wide range above the critical fields, and

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determine values of the coefficients that provide a best fit to the experimental data. Such studies on capacitance measurements by Bradshaw *et al.* (1981) and on optical data by Scheuble *et al.* (1981) and Bunning *et al.* (1981) appear to have led to more accurate and consistent values for the coefficients in the energy function.

One exception to the above remarks is of course the fourth energy coefficient k_{24}. Since the corresponding term does not contribute to (2.24), this coefficient does not appear in any of the predicted critical field strengths, nor in the description of behaviour above threshold. It does, however, appear in the couple stress and the energy density in certain cases, but at present no method appears to be available for its measurement.

3. Dynamic theory

Formulation of theory

Given that static theory describes equilibrium configurations rather well, one naturally seeks to generalize it when proposing a theory for dynamic effects in nematic liquid crystals. One such approach is to replace the principle of virtual work by a corresponding energy balance and derive equations for conservation of linear and angular momenta from it by invariance arguments, as for example Leslie (1979) describes. This leads to the rather natural extensions of (2.11) and (2.12)

$$\rho \dot{v}_i = F_i + t_{ij,j} \tag{3.1}$$

and

$$\sigma \dot{\omega}_i = K_i + e_{ijk} t_{kj} + l_{ij,j}, \tag{3.2}$$

where $\boldsymbol{v}$ denotes velocity and $\boldsymbol{\omega}$ local angular velocity given by

$$\boldsymbol{\omega} = \boldsymbol{n} \times \dot{\boldsymbol{n}}, \tag{3.3}$$

any component parallel to $\boldsymbol{n}$ itself being immaterial in a director description. On account of the assumption of incompressibility, the velocity $\boldsymbol{v}$ is subject to the constraint

$$\operatorname{div} \boldsymbol{v} = 0, \tag{3.4}$$

and the density ρ is simply a constant. The coefficient σ is a constant inertial coefficient, but the inertial term in angular momentum is frequently neglected, being generally considered unimportant. As before, (3.2) can be recast as

$$\sigma \ddot{n}_i = G_i + g_i + s_{ij,j}, \tag{3.5}$$

where

$$e_{ijk} n_j g_k = e_{ijk}(t_{kj} + s_{kp} n_{j,p}), \tag{3.6}$$

this again by using (2.10) and (2.14).

A natural starting point in the search for appropriate constitutive relations is simply to add dynamic contributions to the relations (2.13), (2.14) and (2.19) of static theory, and therefore one writes

$$\left.\begin{aligned} t_{ij} &= -p\delta_{ij} - (\partial W/\partial n_{k,j})\, n_{k,i} + \tilde{t}_{ij}, \\ s_{ij} &= n_i \beta_j + \partial W/\partial n_{i,j} + \tilde{s}_{ij}, \\ g_i &= \gamma n_i - (n_i \beta_j)_{,j} - \partial W/\partial n_i + \tilde{g}_i, \end{aligned}\right\} \tag{3.7}$$

where from the definition (3.6) and the identity (2.18)

$$e_{ijk} n_j \tilde{g}_k = e_{ijk}(\tilde{t}_{kj} + \tilde{s}_{kp} n_{j,p}), \tag{3.8}$$

and these additional terms $\tilde{\boldsymbol{t}}$, $\tilde{\boldsymbol{s}}$ and $\tilde{\boldsymbol{g}}$ all vanish in equilibrium. The early viscosity measurements by Miesowicz (1936) clearly indicate that the dynamic contribution to the stress tensor depends upon the alignment, and the somewhat related measurements by Zwetkoff (1939) suggest that it also depends upon the rate of change of the anisotropic axis. Hence for a relevant theory it seems necessary to assume that the dynamic contributions are all functions of the director, its material time derivative, and the velocity gradients. Initially it is not obvious that one should not include certain other variables in these relations, but those cited appear sufficient to cover many effects.

It seems reasonable to ask that the dynamic terms vanish in a rigid rotation, and this implies dependence upon the velocity gradients and the director velocity solely through the rate of strain tensor $\boldsymbol{A}$ and a vector $\boldsymbol{N}$, where

$$2A_{ij} = v_{i,j} + v_{j,i}, \quad N_i = \dot{n}_i - W_{ik}n_k, \quad 2W_{ij} = v_{i,j} - v_{j,i}. \tag{3.9}$$

It is also natural to consider a linear dependence upon both, given the experimental evidence mentioned above. Further, because this theory does not distinguish between $\boldsymbol{n}$ and $-\boldsymbol{n}$, the stress tensor must be unaffected by a change of sign in the vector $\boldsymbol{n}$, but generalized stress and the intrinsic body force must change sign with it. Such arguments coupled with invariance to superposed rigid rotations and appeal to nematic symmetries lead to

$$\tilde{t}_{ij} = \alpha_1 n_k n_p A_{kp} n_i n_j + \alpha_2 N_i n_j + \alpha_3 N_j n_i + \alpha_4 A_{ij} + \alpha_5 A_{ik} n_k n_j + \alpha_6 A_{jk} n_k n_i, \tag{3.10}$$

$$\tilde{s}_{ij} = 0, \quad \tilde{g}_i = -\gamma_1 N_i - \gamma_2 A_{ik} n_k, \tag{3.11}$$

where

$$\gamma_1 = \alpha_3 - \alpha_2, \quad \gamma_2 = \alpha_6 - \alpha_5, \tag{3.12}$$

the coefficients being at most functions of temperature. Earlier derivations of these relations by Leslie (1968, 1979) show that the dynamic part of the generalized stress is zero by other arguments. Of course thermodynamic considerations place some restriction upon possible values for the coefficients in the above relations. Leslie (1968) argues that positive entropy production requires that

$$\tilde{t}_{ij} A_{ij} - \tilde{g}_i N_i \geqslant 0, \tag{3.13}$$

and Parodi (1970) adds one further restriction, namely

$$\gamma_2 = \alpha_3 + \alpha_2 \tag{3.14}$$

by appeal to an Onsager relation.

Scaling property

As Ericksen (1969) points out, the above equations have a rather surprising property when external body forces and couples are absent. If one scales spatial coordinates and time by

$$\boldsymbol{x} = l\boldsymbol{x}^*, \quad t = l^2 t^*, \tag{3.15}$$

where l is some constant, the transformed equations are identical to the original provided that one neglects the inertial term in (3.5) and selects

$$p = l^{-2} p^*, \quad \gamma = l^{-2}\gamma^*. \tag{3.16}$$

In this scaling, the director's being a unit vector is not altered, but velocity and stress transform according to

$$\boldsymbol{v}^* = l\boldsymbol{v}, \quad \boldsymbol{t}^* = l^2 \boldsymbol{t}. \tag{3.17}$$

This rather simple property leads quickly to certain conclusions that go a long way towards vindicating the special constitutive assumptions outlined above.

For example, consider a simple shear flow between parallel plates a distance l apart and subject to a relative shear velocity V. The above scaling at once transforms this given problem into simple shear flow between parallel plates with unit gap and relative shear velocity V^*, where

$$V^* = Vl, \tag{3.18}$$

but the prescribed surface alignments are unchanged. Given a unique solution to the problem, it follows that

$$\boldsymbol{n} = \boldsymbol{n}^* = \boldsymbol{n}^*(\boldsymbol{x}^*, V^*) = \boldsymbol{n}^*(l^{-1}\boldsymbol{x}, Vl), \tag{3.19}$$

because V^* is the sole parameter in the transformed problem. From this one notes that in simple shear an optical property can depend upon V only through the product Vl. Rather encouragingly, Wahl & Fischer (1973) find such a dependence in optical measurements on a flow that closely approximates to simple shear.

Moreover, if one defines an apparent viscosity η for simple shear by

$$\eta = \tau l/V, \tag{3.20}$$

where τ is the shear stress applied per unit area of plate, it follows that

$$\eta = \tau l^2/Vl = \tau^*/V^* = \eta^*. \tag{3.21}$$

Hence one finds that

$$\eta = \eta^* = \mathscr{F}(V^*) = \mathscr{F}(Vl), \tag{3.22}$$

$\mathscr{F}$ being some unknown function. Unfortunately no experimental data are available to allow verification or otherwise of this novel prediction.

However, a similar analysis is feasible for Poiseuille flow, and for this flow Atkin (1970) predicts that the flux Q per unit time from a capillary of radius R is related to the pressure gradiant a by

$$Q = R\mathscr{K}(aR^3), \tag{3.23}$$

the function $\mathscr{K}$ being unknown. Moreover, if one defines as apparent viscosity in this instance by

$$\eta = \pi a R^4/8Q, \tag{3.24}$$

it immediately follows that

$$\eta = \mathscr{G}(Q/R), \tag{3.25}$$

the function $\mathscr{G}$ again being unknown. This result is in marked contrast with the corresponding one for an isotropic liquid where the dependence is upon Q/R^3, *but* it has been confirmed experimentally by Fisher & Fredrickson (1969). These authors initially plot the apparent viscosity as a function of the latter and obtain distinct curves for each of the four capillaries used. However, they proceed to show that their data neatly fit a single curve when plotted according to Atkin's prediction.

As Leslie (1979) argues, this experimental confirmation of Atkin's novel scaling, and also that by Wahl & Fischer for simple shear, are particularly important because they largely vindicate the particular form of the theory, especially the functional dependence assumed in the constitutive theory. Any generalization of the above constitutive relations must quickly lead to predictions at odds with these experimental observations. Also, as Leslie points out, given the rapid changes of alignment in the narrow capillaries employed by Fisher & Fredrickson, it is hard to imagine that additional terms omitted in the above derivation could be negligible in these experiments. In view of the relative importance of this topic, it is somewhat surprising that essentially only two experimental investigations examine it. However, any generalization or extension of the above theory for nematic liquid crystals seems premature until further experimental evidence points to violations of the scaling described above.

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Flow alignment

Flow can exert a strong influence upon alignment in a nematic, and therefore a brief résumé of this topic is helpful before proceeding to a discussion of methods used to measure the various viscous coefficients. For most purposes it is sufficient to consider simple shear flow and to ignore complications arising from the competing influences of boundaries and external fields. Here, therefore, one examines solutions of the above equations in which the velocity and director have Cartesian components of the form

$$\left.\begin{aligned} v_x = \kappa z, \quad v_y = v_z = 0, \quad \kappa > 0, \\ n_x = \cos\theta\cos\phi, \quad n_y = \cos\theta\sin\phi, \quad n_z = \sin\theta, \end{aligned}\right\} \tag{3.26}$$

where κ, θ and ϕ are simply constants. This choice at once satisfies (3.1) and (3.4) rather trivially, and reduces (3.5) to

$$\kappa(\alpha_3\cos^2\theta - \alpha_2\sin^2\theta)\cos\phi = 0, \quad \kappa\alpha_2\sin\theta\sin\phi = 0, \tag{3.27}$$

the final expressions by using the relations (3.12) and (3.14). Whatever the values of α_2 and α_3, the above equations always have a solution

$$\theta = 0, \quad \phi = \tfrac{1}{2}\pi, \tag{3.28}$$

representing alignment normal to the plane of shear. In addition, if α_2 and α_3 have the same sign, there are two steady solutions in the plane of shear, namely

$$\theta = \pm\theta_0, \quad \phi = 0, \tag{3.29}$$

where the acute angle θ_0 is defined by

$$\tan^2\theta_0 = \alpha_3/\alpha_2. \tag{3.30}$$

The thermodynamic inequality requires that γ_1 be positive and so two possibilities emerge: either

$$\alpha_2 < \alpha_3 < 0 \quad (0 < \theta_0 < \tfrac{1}{4}\pi), \tag{3.31}$$

or

$$\alpha_3 > \alpha_2 > 0 \quad (\tfrac{1}{4}\pi < \theta_0 < \tfrac{1}{2}\pi). \tag{3.32}$$

For the frequently observed alignment at a small angle to the streamlines, the viscous coefficients must clearly satisfy the former.

An examination of time-dependent perturbations of the director shows that the solution (3.28) is always unstable, and this configuration and its instability have been studied extensively by Pieranski & Guyon (see, for example, Dubois-Violette *et al.* (1977)). If the solutions (3.29) are possible, one is stable to such perturbations, and the other unstable. When the coefficients satisfy (3.31) the stable configuration corresponds to that with the positive sign, but, if (3.32) applies, the stable solution has the negative sign. It is of course possible to measure this flow alignment angle θ_0 experimentally, and Gähwiller (1971, 1973) describes such measurements. While there are solutions of the equations that demonstrate the competition between flow and boundaries on alignment, their description is not attempted here, given their complexity. None the less, the response of such flow-aligning nematics in shear flow is now reasonably well understood (see, for example, Leslie (1981)).

However, as first discovered by Gähwiller (1972), there are nematics that do not align in shear flow, this being ascribed to the coefficient α_3 changing sign for at least part of its nematic temperature range, this generally preceding a transition to a smectic phase. For this type of

nematic, the behaviour in shear flow is not as well understood. Two experimental studies, one by Pieranski *et al.* (1976) and the second by Cladis & Torza (1975), investigate this and some differences emerge. A variety of flow-induced instabilities certainly occur, possibly accompanied by other complications, but theoretical investigations appear to be limited to an approximate analysis by Pikin (1974), what is available in the two papers just cited, and a discussion of oscillatory shear by Clark *et al.* (1981). Certainly this topic deserves more attention than it has received, although progress may be difficult.

The behaviour in shear flow certainly typifies the response in other viscometric flows as far as one can tell at present. Little has been yet attempted with regard to flows that are not viscometric, presumably because there appears to have been little need for it. However, studies of other anisotropic systems tend to look to liquid crystal theory for guidance, and such interests may begin to motivate investigations of other flows.

Viscosity measurements

Viscosity measurements commonly employ a magnetic field to control alignment of the nematic, while measuring some quantity that yields a value for the corresponding viscosity. The assumption is that a sufficiently strong field completely aligns the liquid crystal, and the test of sufficient strength is generally that measurements appear to reach some limiting value. The theory employed is somewhat limited, simply assuming uniform alignment with possibly some assessment of likely errors due to variations at boundaries.

To examine the information obtainable from such measurements it suffices to consider simple shear flow with some arbitrary alignment (3.26). If one defines a viscosity by

$$\eta = t_{xz}/\kappa, \tag{3.33}$$

it follows straightforwardly that

$$\eta = \eta_c \sin^2\theta + (\eta_a \sin^2\phi + \eta_b \cos^2\phi)\cos^2\theta + \alpha_1 \sin^2\theta \cos^2\theta \cos^2\phi, \tag{3.34}$$

where

$$2\eta_a = \alpha_4, \quad 2\eta_b = \alpha_4 + \alpha_3 + \alpha_6, \quad 2\eta_c = \alpha_4 + \alpha_5 - \alpha_2. \tag{3.35}$$

Hence from such measurements one hopes to obtain three viscosities corresponding to particular alignments as follows:

$$\left.\begin{array}{llll} \text{(i)} & \theta = 0, & \phi = \tfrac{1}{2}\pi, & \eta = \eta_a, \\ \text{(ii)} & \theta = 0, & \phi = 0, & \eta = \eta_b, \\ \text{(iii)} & \theta = \tfrac{1}{2}\pi, & \phi = 0, & \eta = \eta_c, \end{array}\right\} \tag{3.36}$$

and the coefficient α_1 by a further measurement with alignment in the plane of shear.

Miesowicz (1936) was the first to attempt such viscosity measurements. He measured the damping of vertical oscillations of a plate suspended in a nematic, calibrating his measurements with a liquid of known viscosity. Because his magnetic field was horizontal he determined η_a and η_c. Subsequently, Gähwiller (1971, 1973) carried out a complete set of such measurements by measuring the flux through a rectangular capillary, again using a standard liquid for calibration. More recently Kneppe & Schneider (1981) and Skarp *et al.* (1980) have also used capillaries to make such measurements on nematics.

As de Jeu (1978) points out, there is some concern that the field strengths are sufficiently large to ensure complete alignment, particularly for η_c. Certainly many of the newer materials are more viscous than those first studied, and consequently greater fields are necessary to overcome

the stronger flow alignment. A further cause for concern is the lack of any theory to guide extrapolation to limiting values. As Clark *et al.* (1980) show in a somewhat related situation, extrapolation without some theory to guide can be rather uncertain.

Given the above quantities accurately determined, the Parodi relation (3.14) and (3.12) yield

$$\eta_b - \eta_c = \gamma_2, \tag{3.37}$$

and hence γ_2 is also found. If the flow alignment angle is known, this provides the fifth measurement and the α are determined. However, if the nematic does not align in shear, some other measurement is necessary. One candidate is the experiment first employed by Zwetkoff (1939) in which he suspended some nematic in a cylinder and rotated a horizontal magnetic field about the axis of suspension. A measurement of the torque transmitted to the cylinder yields a value for γ_1. Here again the theory used is relatively simplistic, and details are given by Gasparoux & Prost (1971). More recently Gerber (1981) has described a similar experiment that appears to remove most of the problems associated with Zwetkoff's method.

At present insufficient measurements have been performed on a given material to allow meaningful comparisons and checks for consistency. It is to be hoped that the progress made with regard to the measurement of the elastic constants will encourage further efforts to determine the viscous constants more accurately. However, there is clearly a need to develop alternative methods, preferably with the use of small samples, although light scattering (see Orsay Liquid Crystal Group 1971) and shear wave reflectance (Martinoty & Candau 1971) are possible techniques meeting this requirement.

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