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T. Carlsson^a, F. M. Leslie^{a b} & J. S. Lavery^{a b}

^a Institute of Theoretical Physics, Chalmers University of Technology, S-412, 96, Göteborg, Sweden

^b Department of Mathematics, University of Strathclyde, Livingstone Tower, Glasgow, G1 1XH, Scotland

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BIAXIAL NEMATIC LIQUID CRYSTALS - FLOW PROPERTIES AND EVIDENCE OF BISTABILITY IN THE PRESENCE OF ELECTRIC AND MAGNETIC FIELDS

T.CARLSSON¹, F.M.LESLIE² AND J.S.LAVERY²

1) Institute of Theoretical Physics, Chalmers University of Technology, S-412 96 Göteborg, Sweden

2) Department of Mathematics, University of Strathclyde, Livingstone Tower, Glasgow G1 1XH, Scotland

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ABSTRACT The stress tensor for the biaxial nematic phase is given in such a form that it is easily seen that the dynamic theory of this phase can be formulated as a natural generalization of the Leslie-Ericksen theory of uniaxial nematics. With the biaxial nematic stress tensor as a starting point we introduce the concept of viscous torques and discuss the qualitative nature of the flow properties of the system, showing that three rotational viscosities and nine effective shearing viscosities have to be defined in order to characterize the viscous behaviour of the biaxial nematic phase completely. We also discuss the behaviour of a biaxial nematic liquid crystal in the presence of electric and magnetic fields and show that in some cases the system can develop bistability if certain conditions for the magnitudes of the electric and magnetic fields are fulfilled.

Keywords: biaxial nematic, liquid crystal, flow alignment, rotational viscosity, hydrodynamics, rheology, bistability, electro-magnetic properties

I INTRODUCTION

The biaxial nematic liquid crystalline phase has recently been reported to exist in lyotropic¹ as well as in thermotropic^{2,3} systems. In order to present a simple microscopic picture of this phase, Figure 1 compares the uniaxial nematic and the biaxial nematic phases. The microscopic picture one most often makes of a uniaxial nematic is a rotationally symmetric ellipsoid, the orientational order of which is described by a unit vector \hat{n} , commonly denoted the director (c.f. Figure 1a). When studying a biaxial nematic the biaxiality of the system can be thought of as a breaking of the rotational symmetry of the ellipsoid around \hat{n} , and instead of studying a system of rotationally symmetric rods one can imagine a system consisting of plates with sides of lengths a , b and c for which we assume $a \gg b \gg c$. The description of such a system requires two directors. Thus we introduce \hat{n} , the "long director" corresponding to the director of the uniaxial nematic and \hat{m} , the "transverse director" describing the rotation of the biaxial plate around the long director (c.f. Figure 1b).

During the ten years which have passed since the biaxial nematic phase was discovered¹ a number of approaches to the description of the rheological properties of this system have been presented⁴⁻⁶. Even if it is not always done in this way, it is always possible to express such a theory as a generalization of the corresponding theory of the uniaxial nematic phase. In this paper, however, we show how to write down the equations governing the flow behaviour of biaxial nematics in such a way⁸ that it is easily seen how the dynamic theory of this phase can be formulated as a generalization of the Leslie-Ericksen theory⁹ of uniaxial nematics. With this formulation as a starting point, and neglecting elasticity, we then discuss some qualitative features of the flow behaviour of the biaxial nematic phase. A brief summary of the behaviour of biaxial nematics in the presence of two perpendicular electric and magnetic fields is also given, and it is shown that under certain circumstances this system can develop bistability⁷.

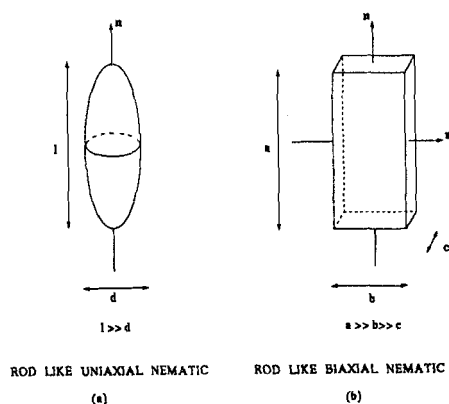


FIGURE 1 Comparison between a rod-like uniaxial nematic (a) and a rod-like biaxial nematic (b).

II THE BIAxIAL NEMATIC STRESS TENSOR

The equations governing the flow behaviour of a biaxial nematic liquid crystal have been derived and discussed by us in detail elsewhere⁸, and below we simply give a brief summary of these, omitting the elastic terms which need not concern us here. In this theory the two basic equations are these representing the conservation of momentum and angular momentum, respectively,

$$\rho \dot{v}_i = \rho F_i + t_{ij,j}, \quad \rho K_i + \epsilon_{ijk} t_{kj} = 0, \quad (\text{II.1})$$

where in the second of these equations we have neglected director inertia. Thus this equation can be interpreted as a balance of torque equation, in which the two terms correspond to the torque exerted on the director due to external fields and to the viscous torque, respectively. Introducing

$$D_{ij} = \frac{1}{2}(v_{i,j} + v_{j,i}), \quad W_{ij} = \frac{1}{2}(v_{i,j} - v_{j,i}), \quad N_i = \dot{n}_i - W_{ik}n_k, \quad M_i = \dot{m}_i - W_{ik}m_k, \quad (\text{II.2})$$

where D_{ij} is the strain rate tensor, W_{ij} the vorticity tensor and N_i and M_i represent the rate of change of the directors with respect to the moving fluid, we can write the biaxial nematic stress tensor as⁸

$$\begin{aligned}
 t_{ij} = & \alpha_1 D_{kp} n_k n_p n_i n_j + \alpha_2 N_i n_j + \alpha_3 N_j n_i + \alpha_4 D_{ij} + \alpha_5 D_{ik} n_k n_j + \alpha_6 D_{jk} n_k n_i \\
 & + \beta_1 D_{kp} m_k m_p m_i m_j + \beta_2 M_i m_j + \beta_3 M_j m_i + \beta_5 D_{ik} m_k m_j + \beta_6 D_{jk} m_k m_i \\
 & + N_p m_p (\mu_1 m_i n_j + \mu_2 n_i m_j) + D_{kp} n_k m_p (\mu_3 m_i n_j + \mu_4 n_i m_j) - p \delta_{ij}.
 \end{aligned} \quad (\text{II.3})$$

In Eq.(II.3) we have introduced fifteen viscosity coefficients into the dynamic theory of biaxial

nematics. These are related⁸ by three Onsager relations

$$\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5, \quad \beta_2 + \beta_3 = \beta_6 - \beta_5, \quad \mu_1 + \mu_2 = \mu_4 - \mu_3, \quad (\text{II.4})$$

and thus altogether twelve linearly independent viscosity coefficients are needed in order to describe the system completely. This is the same number as was given by Saupe in his derivation⁴ of the biaxial nematic stress tensor.

The form of the stress tensor given by Eq.(II.3) is easily interpreted in the light of Figure 1. The six α_i -terms, which are the ones acting on the long director correspond exactly to the Leslie stress tensor⁹ of uniaxial nematics. The five β_i -terms have the same structure as the five anisotropic α_i -terms, only \hat{n} is replaced by \hat{m} and thus these terms are acting on the transverse director. Finally there are four μ_i -terms which reflect coupling effects between the long and the transverse directors. These terms do not have any counterpart in the Leslie-Ericksen theory of uniaxial nematics.

III THE ROTATIONAL VISCOSITIES

The viscous torque $\Gamma^v = \epsilon_{ijk} t_{kj}$ is most conveniently divided into two parts: the shearing torque Γ^s which is the torque acting on the directors due to velocity gradients and the rotational torque Γ^r which appears if $\dot{\hat{n}} \neq 0$ or $\dot{\hat{m}} \neq 0$. Being interested in the rotational torque, we can write this as $\Gamma_i^r = \epsilon_{ijk} t_{kj}^r$ where t_{kj}^r is the rotational part of the stress tensor

$$t_{kj}^r = \alpha_2 \dot{n}_k n_j + \alpha_3 \dot{n}_j n_k + \beta_2 \dot{m}_k m_j + \beta_3 \dot{m}_j m_k + \dot{n}_p m_p (\mu_1 m_k n_j + \mu_2 n_k m_j). \quad (\text{III.1})$$

Introducing the Euler angles to describe the biaxial plate, i.e. a spherical polar coordinate system (θ, ϕ) to describe the long director \hat{n} and an angle ψ describing the rotation of the transverse director \hat{m} around the long director (c.f. Figure 3), we derive⁸ the following expression for the rotational torque

$$\begin{aligned} \Gamma_\phi^r &= -(\beta_3 - \beta_2) (\dot{\phi} \cos \theta + \dot{\psi}) \\ \Gamma_\theta^r &= (\alpha_3 - \alpha_2) \dot{\phi} \sin \theta + (\beta_3 - \beta_2 + \mu_2 - \mu_1) (\dot{\phi} \sin \theta \cos^2 \psi - \dot{\theta} \sin \psi \cos \psi) \\ \Gamma_\psi^r &= -(\alpha_3 - \alpha_2) \dot{\theta} - (\beta_3 - \beta_2 + \mu_2 - \mu_1) (\dot{\theta} \sin^2 \psi - \dot{\phi} \sin \theta \sin \psi \cos \psi). \end{aligned} \quad (\text{III.2})$$

From Eqs.(III.2) one can define three rotational viscosities of the biaxial nematic phase as

$$\gamma_n = \alpha_3 - \alpha_2 > 0, \quad \gamma_m = \beta_3 - \beta_2 > 0, \quad \gamma_{nm} = \alpha_3 - \alpha_2 + \beta_3 - \beta_2 + \mu_2 - \mu_1 > 0. \quad (\text{III.3})$$

Each of these rotational viscosities is related to the rotation of the biaxial plate around one of its three principal axes as is pictured in Figure 2 and must always be positive by thermodynamic considerations⁸.

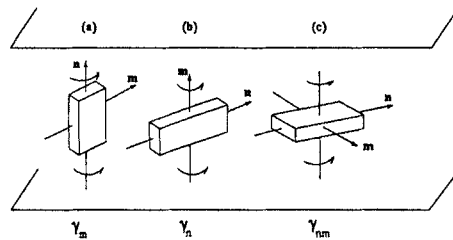


FIGURE 2 Definition of the three rotational viscosities of the biaxial nematic phase.

IV THE SHEARING TORQUE AND THE FLOW ALIGNMENT ANGLES

In this section we study the behaviour of the biaxial nematic system when subjected to shear flow. Confining the liquid crystal between two glass plates, both parallel to the xy -plane, and moving the upper one in the x -direction with the velocity v_0 , the coordinates describing the system are defined in Figure 3. The directors are described by the Euler angles (θ, ϕ, ψ) as discussed in the previous section, $\theta = 0$ corresponding to the long director pointing in the z -direction. Neglecting the possibility of transverse flow (this assumption is justified in reference 8), a shear $v' = dv_x/dz$ will be induced in the fluid. From Eqs.(II.1) - (II.3) the spherical polar components of the shearing torque $\Gamma^v = \epsilon_{ijk} t_{kj}$ can be calculated⁸ to be

$$\Gamma_z^v = v' [\beta_3(\sin\theta \sin\phi \cos^2\psi + \sin\theta \cos\theta \cos\phi \sin\psi \cos\psi) + \beta_2(\sin\theta \cos\theta \cos\phi \sin\psi \cos\psi - \sin\theta \sin\phi \sin^2\psi)]$$

$$\Gamma_\theta^v = v' [-\alpha_2 \cos\theta \sin\phi - \beta_2 \sin^2\theta \cos\phi \sin\psi \cos\psi + \beta_3(\cos\theta \sin\phi \cos^2\psi + \cos^2\theta \cos\phi \sin\psi \cos\psi) - \mu_1 \cos\theta \sin\phi \cos^2\psi + \cos\phi \sin\psi \cos\psi (\mu_2 \sin^2\theta - \mu_1 \cos^2\theta)]$$

$$\Gamma_\phi^v = v' [(\alpha_3 \sin^2\theta - \alpha_2 \cos^2\theta) \cos\phi - \beta_2 \sin^2\theta \cos\phi \sin^2\psi + \beta_3(\cos\theta \sin\phi \sin\psi \cos\psi + \cos^2\theta \cos\phi \sin^2\psi) - \mu_1 \cos\theta \sin\phi \sin\psi \cos\psi + \cos\phi \sin^2\psi (\mu_2 \sin^2\theta - \mu_1 \cos^2\theta)] . \quad (\text{IV.1})$$

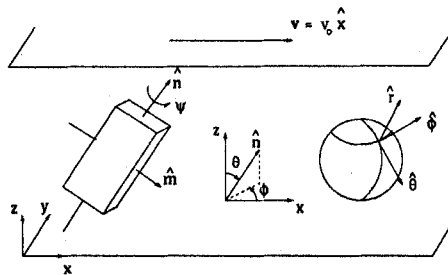


FIGURE 3 Definition of the coordinates describing the biaxial plate in the shear flow setup.

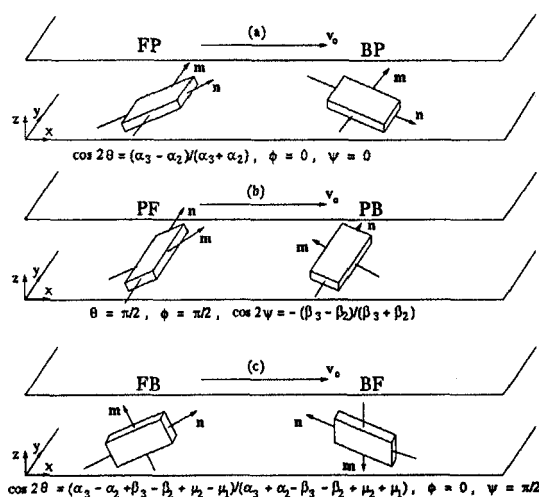


FIGURE 4 Equilibrium orientations of the biaxial plate in shear flow.

Depending on the values of the viscosity coefficients, the shearing torque will vanish for six, four, two or no different orientations of the biaxial plate, all occurring in pairs, leading to the possibility of flow alignment. The six possible orientations, and the corresponding values of the coordinates describing the biaxial plate, are shown in Figure 4. We have introduced a notation, also shown in the figure, of these six orientations in such a way that the first letter indicates whether the long director is confined within the shearing plane, pointing in the forward (F) or backward (B) direction, or if it is pointing in the y-direction (P). In the same manner the second letter is defined by the orientation of the transverse director. The analysis of the stability of the six equilibrium angles is rather involved and is discussed by us elsewhere⁸.

V THE EFFECTIVE VISCOSITIES

When shearing the system as shown in Figure 3 one has to apply a force τ per unit area to the moving plate in order to maintain its constant velocity v_0 . If we have oriented the two directors homogeneously throughout the whole sample, by the means of applying some external fields, the shear rate v' will be uniform in space, and we can define an effective viscosity η_{ij} according to $\eta_{ij} = \tau/v'$, where the indices refer to the particular configuration studied. Assuming a velocity profile $\mathbf{v} = v(z)\hat{x}$, the first of Eqs.(II.1) reduces to $t_{xz,x} = 0$ in the steady state, and can be integrated to read $t_{xz} = \tau$. Thus we can introduce a viscosity function $g(n_i, m_j) = \tau/v'$ which is calculated from Eq.(II.3),

$$g = \frac{1}{2} [2\alpha_1 n_x^2 n_z^2 - \alpha_2 n_z^2 + \alpha_3 n_x^2 + \alpha_4 + \alpha_5 n_z^2 + \alpha_6 n_x^2 + 2\beta_1 m_x^2 m_z^2 - \beta_2 m_z^2 + \beta_3 m_x^2 + \beta_5 m_z^2 + \beta_6 m_x^2 + \mu_1 (n_x m_z - n_z m_x) n_x m_x + \mu_2 (n_x m_z - n_z m_x) n_z m_z + \mu_3 (n_x m_z + n_z m_x) n_x m_x + \mu_4 (n_x m_z + n_z m_x) n_z m_z] .$$

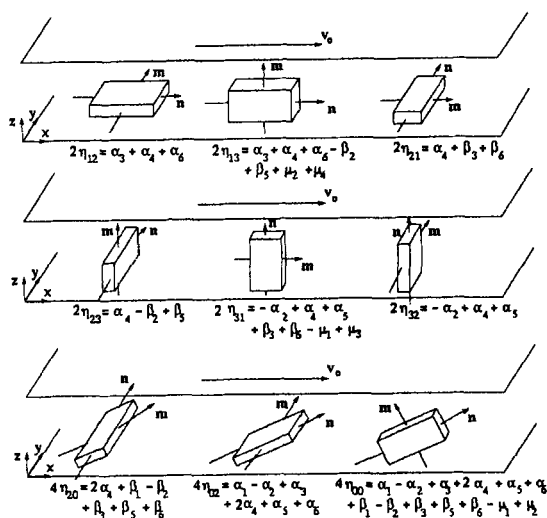


FIGURE 5 The nine linearly independent effective viscosities of biaxial nematics.

In Figure 5 we show the nine different orientations of the biaxial plate which are used to define nine linearly independent effective viscosities. We also introduce a specific notation for these effective viscosities and show how to express them in terms of the fifteen viscosity coefficients introduced by the stress tensor (II.3). As the flow must always be in the same direction as the driving force, we conclude that each of the effective viscosities must be positive. Thus we have shown how the twelve independent viscosity coefficients introduced by the stress tensor can be determined experimentally by the measurement of three rotational viscosities and nine effective shearing viscosities.

VI INTERACTION WITH ELECTRIC AND MAGNETIC FIELDS - EVIDENCE OF BISTABILITY

In this section we discuss the orientational effects which the application of two perpendicular electric and magnetic fields will impose on the two directors of the system. In order to describe the dielectric and magnetic properties of the medium we have to introduce three dielectric permittivities ($\epsilon_n, \epsilon_m, \epsilon_l$) and three magnetic susceptibilities (χ_n, χ_m, χ_l), one for each principal axis of the biaxial plate, employing the notation l for the third principal axis defined by $\hat{l} = \hat{n} \times \hat{m}$. The corresponding dielectric and magnetic anisotropies are $\epsilon_{ij} = \epsilon_i - \epsilon_j$ and $\chi_{ij} = \chi_i - \chi_j$. Furthermore it is convenient to introduce a notation which incorporates the coupling constants of the fields into the susceptibilities,

$$\bar{\epsilon}_i = \epsilon_0 E^2 \epsilon_i, \quad \bar{\epsilon}_{ij} = \epsilon_0 E^2 \epsilon_{ij}, \quad \bar{\chi}_i = \mu_0^{-1} B^2 \chi_i, \quad \bar{\chi}_{ij} = \mu_0^{-1} B^2 \chi_{ij}. \quad (\text{VI.1})$$

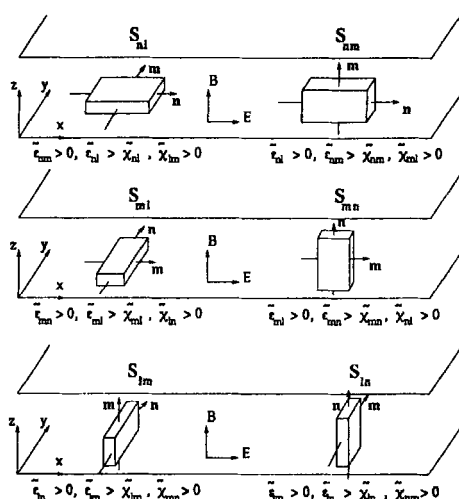


FIGURE 6 The six equilibrium configurations of biaxial nematics in the presence of two perpendicular electric and magnetic fields. The conditions for each of the solutions to be stable are also displayed in the figure.

Assuming that the fields are given by $\mathbf{E} = E\hat{x}$ and $\mathbf{B} = B\hat{z}$, respectively, the Cartesian components of the electro-magnetic torque are given by⁷

$$\begin{aligned}\Gamma_x^{\mathcal{E}} &= \chi_{nl} n_y n_z + \chi_{ml} m_y m_z \\ \Gamma_y^{\mathcal{E}} &= -\chi_{nl} n_x n_z - \chi_{ml} m_x m_z + \bar{\epsilon}_{nl} n_x n_z + \bar{\epsilon}_{ml} m_x m_z \\ \Gamma_z^{\mathcal{E}} &= -\bar{\epsilon}_{nl} n_x n_y - \bar{\epsilon}_{ml} m_x m_y\end{aligned}\quad (\text{VI.2})$$

From Eqs.(VI.2) we immediately see that all configurations for which two of the three vectors \hat{n} , \hat{m} and \hat{l} are parallel to the fields will be possible equilibrium configurations of the system. In Figure 6 we show these solutions and also give the appropriate conditions, which can be derived⁷ from Eqs.(VI.2), for each of these to be stable. It is helpful to introduce a notation for the six solutions in the following way. The solution S_{ij} corresponds to that for which the i -director points in the direction of the electric field and the j -director is in the direction of the magnetic field. Thus the solution for which $\hat{n} = \hat{x}$ and $\hat{m} = \hat{z}$ is denoted S_{nm} and so on. This notation is shown in Figure 6.

The interpretation of the stability conditions given in Figure 6 is not entirely straightforward. It turns out⁷ that we have to divide the biaxial nematic compounds into three different classes, depending on how the magnitudes of ϵ_i and χ_i are mutually ordered. The compounds belonging to each class will exhibit a different qualitative behaviour in the presence of the fields. In order to classify a specific compound we will introduce a state vector ψ_{pqr}^{ijk} in the following way. The three upper indices of ψ refer to the ordering of the magnitudes of the dielectric permittivities, while the three lower indices refer to the ordering of the magnitudes of the magnetic susceptibilities. Thus the state vector ψ_{pqr}^{ijk} is used to describe a compound for which $\epsilon_i > \epsilon_j > \epsilon_k$ and $\chi_p > \chi_q > \chi_r$. There are altogether thirty-six different state vectors and below

we introduce a classification of these, also giving an account of how the corresponding compounds will order in the presence of the fields.

TYPE A: The twentyfour state vectors for which the axis with the largest dielectric permittivity does not coincide with the axis with the largest magnetic susceptibility, i.e. the state vectors of the type ψ_{pqr}^{ijk} where $i \neq p$. Compounds belonging to this type will always adopt the solution S_{ip} irrespective of the magnitudes of the applied fields.

TYPE B: The six state vectors for which the ordering of the three dielectric permittivities is the same as that of the three magnetic susceptibilities, i.e. the state vectors of the type ψ_{ijk}^{ijk} . Compounds belonging to this type will adopt the solution S_{ij} if the electric field is dominating ($\epsilon_0 \epsilon_{ij} E^2 > \mu_0^{-1} \chi_{ij} B^2$) while the solution S_{ji} is adopted if the inequality given above is reversed.

TYPE C: The six state vectors for which the axis with the largest dielectric permittivity coincides with the axis with the largest magnetic susceptibility but for which the ordering of the two remaining dielectric permittivities is opposite to that of the magnetic susceptibilities, i.e. the state vectors of the type ψ_{ikj}^{ijk} . Compounds belonging to this type will adopt the solution S_{ik} if the electric field is dominating ($\epsilon_0 \epsilon_{ij} E^2 > \mu_0^{-1} \chi_{ij} B^2$), while it will adopt the solution S_{ji} if the magnetic field is dominating ($\mu_0^{-1} \chi_{ik} B^2 > \epsilon_0 \epsilon_{ik} E^2$). For intermediate field strengths ($\mu_0^{-1} B^2 \chi_{ik} / \epsilon_{ik} < \epsilon_0 E^2 < \mu_0^{-1} B^2 \chi_{ij} / \epsilon_{ij}$) it turns out⁷ that both the solutions S_{ik} and S_{ji} are stable and thus the system for these values of the field strengths exhibits bistability.

From the above discussion it is clear that some caution has to be taken when trying to orient a biaxial nematic sample by the means of electric and magnetic fields, because depending upon which type the compound belongs to, different orientations will be achieved. Especially, when dealing with a compound belonging to TYPE C, we will find that for intermediate field strengths, the final orientation obtained depends upon the configuration of the system prior to the application of the fields as well as upon the order in which the fields are turned on. A general treatment of the behaviour of biaxial nematics in the presence of electric and magnetic fields and of the classification scheme introduced above has been given by us elsewhere⁷.

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