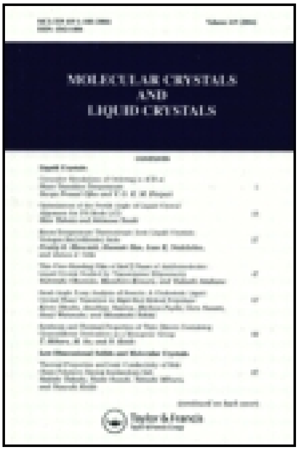

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Continuum and Molecular Theories of Biaxial Nematics: Calculation of the 2-Director Viscosity Coefficients

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Continuum and Molecular Theories of Biaxial Nematics: Calculation of the 2-Director Viscosity Coefficients

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We show the equivalence of the 2-director and the 3-director continuum theories of biaxial nematics, thereby extending the 2-director theory to the compressible case. We calculate the viscosity coefficients in terms of order parameters within the framework of the 2-director continuum theory. Finally, we extend the Doi molecular approach to the biaxial case and calculate the 2-director viscosity coefficients in terms of the order parameters, diffusion coefficients and temperature.

Keywords: biaxial liquid crystals, nematic liquid crystals, theory of biaxial order

1. INTRODUCTION

Since the experimental discovery of the biaxial nematic phase in the early eighties in a 3-component mixture,[1] many theoretical advances have been made in the description of these very interesting mesophases. In fact, theoretical work in this context preceded the experimental discovery by about a decade, when Freiser,[2] Alben[3] and Straley[4] published their independent phenomenological theories in the early seventies. The work of Alben extended the Landau theory of uniaxial nematics, while both Freiser and Straley generalized a microscopic statistical mechanical mean field theory first applied to the uniaxial nematics by Maier and Saupe.[5] Continuum theories for the biaxial nematics belong to two categories: the 2-director and 3-director models. Both use the dissipation function approach, as first put forward by Lam.[6] However, the 3-director continuum theory uses the three director: **n**, **l**, and **m** on equal footing, and a symmetric dissipation function is thereby obtained. In the following, our goal is to establish the equivalence of the 2-director and the 3-director continuum theories, in the sense that each viscosity coefficient in one model is expressible as a linear combination of the viscosity coefficients of the other model.

Also, Chauré[7] proposed the 2-director theory for an incompressible fluid. We have extended the 2-director theory to include compressible biaxial nematics also.

Some inequalities implied by the positive definiteness of the dissipation function have been recorded in Section 2. In Section 3, we calculate the viscosity coefficients of the two director model in terms of the order parameters, within the framework of the continuum theories. Following Ericksen,[8] these expressions may be called the consistency conditions of the dissipation function. Finally in Section 4, we have used Doi-type molecular theoretical approach to calculate the viscosity coefficients for the 2-director biaxial case, in the same spirit as Marrucci's[9] calculation of the Leslie viscosity coefficients for the uniaxial case.

2. EQUIVALENCE OF THE 2- AND 3-DIRECTOR MODELS OF BIAXIAL NEMATICS

Relative merits in brief: the 3-director model can be readily related to the Harvard[10] formulation; the 2-director model is more compatible with the Ericksen-Leslie-Parodi[7] (ELP) approach. Note that crossed electromagnetic fields[11] are needed for experiments in order to orient the biaxial nematic sample.

Chauré generalized the Ericksen-Leslie theory,[12,13] using the isotropic representation theorems of Wang.[14] The mechanical part of the dissipation function for biaxial nematics, thus obtained, contains 12 invariants, as against only 5 in the uniaxial theory.[8] The invariants are written in terms of two directors **n** and **m** only. This is the 2-director continuum theory of biaxial nematics. The 3-director theory[15] starts with the premise that macroscopically, a velocity field $\mathbf{v}(\mathbf{r}, t)$ or a tensor velocity gradient field **K**, an inertial tensor field $\mathbf{I}(\mathbf{r}, t)$ and a tensor order parameter field $\mathbf{S}(\mathbf{r}, t)$ are required for the description of the anisotropic fluid. The elements of second-rank tensors **I** and **Q** read:

$$I_{\alpha\beta} = I_1 l_\alpha l_\beta + I_2 m_\alpha m_\beta + I_3 n_\alpha n_\beta \tag{2.1}$$

and

$$Q_{\alpha\beta} \equiv -\frac{1}{3}(S - B)l_\alpha l_\beta - \frac{1}{3}(S + B)m_\alpha m_\beta + \frac{2}{3} S n_\alpha n_\beta \tag{2.2}$$

where S and B are two macroscopic order parameters, and I_1, I_2, and I_3 denote moments of inertia about **l**, **m**, **n**, respectively. The forms for the directors **l**, **m**, and **n** in terms of the Euler angles θ, ϕ, and χ are given in Appendix C. Lagrange's equations of motion of the Euler angles are used to describe the time-dependent behavior of the orientational field. Using the definitions:

$$A_{\alpha\beta} \equiv \frac{1}{2}(\partial_\alpha v_\beta + \partial_\beta v_\alpha) \equiv \frac{1}{2}(K_{\alpha\beta} + K^{\mathrm{T}}_{\alpha\beta}), \tag{2.3a}$$

and

$$\Omega_{\alpha\beta} \equiv \frac{1}{2}(\partial_\alpha v_\beta - \partial_\beta v_\alpha) = \frac{1}{2}(K_{\alpha\beta} - K^{\mathrm{T}}_{\alpha\beta}) \tag{2.3b}$$

for the symmetric (**A**) and the skew-symmetric (Ω) parts of the velocity gradient tensor and the definitions:

$$L_\alpha \equiv \dot{l}_\alpha - \Omega_{\alpha\beta} l_\beta, \tag{2.4a}$$

$$M_\alpha \equiv \dot{m}_\alpha - \Omega_{\alpha\beta} m_\beta, \tag{2.4b}$$

and

$$N_\alpha \equiv \dot{n}_\alpha - \Omega_{\alpha\beta} n_\beta \tag{2.4c}$$

for the corotational time derivatives of the directors **l**, **m**, and **n**, respectively, Govers and Vertogen[15] obtained the Rayleigh dissipation function for a compressible biaxial nematic in the 3-director formalism as:

$$\begin{aligned} D = &\Big(\frac{1}{2}\eta_1 l_\alpha l_\beta l_\gamma l_\delta + \frac{1}{2}\eta_2 m_\alpha m_\beta m_\gamma m_\delta + \frac{1}{2}\eta_3 n_\alpha n_\beta n_\gamma n_\delta + \eta_4 l_\alpha l_\beta m_\gamma m_\delta \\ &+ \eta_5 m_\alpha m_\beta n_\gamma n_\delta + \eta_6 n_\alpha n_\beta l_\gamma l_\delta + \eta_7 l_\alpha m_\beta l_\gamma m_\delta + \eta_8 m_\alpha n_\beta m_\gamma n_\delta \\ &+ \eta_9 n_\alpha l_\beta n_\gamma l_\delta\Big) A_{\alpha\beta} A_{\gamma\delta} + \frac{1}{2}\eta_{10}(m_\gamma L_\gamma)^2 + \frac{1}{2}\eta_{11}(n_\gamma M_\gamma)^2 \\ &+ \frac{1}{2}\eta_{12}(l_\gamma N_\gamma)^2 + (\eta_{13} l_\alpha m_\beta m_\gamma L_\gamma + \eta_{14} m_\alpha n_\beta n_\gamma M_\gamma \\ &+ \eta_{15} n_\alpha l_\beta l_\gamma N_\gamma) A_{\alpha\beta}. \end{aligned} \tag{2.5}$$

where the η_j are viscous coefficients that depend on order parameters, as shown in Sections 3 and 4. For the incompressible case,

$$\begin{aligned} 0 &= \operatorname{tr} \mathbf{A} = \mathbf{m}\cdot\mathbf{A}\mathbf{m} + \mathbf{n}\cdot\mathbf{A}\mathbf{n} + \mathbf{l}\cdot\mathbf{A}\mathbf{l} \\ l_\alpha l_\beta l_\gamma l_\delta A_{\alpha\beta} A_{\gamma\delta} &= (l_\alpha l_\beta m_\gamma m_\delta + n_\alpha n_\beta l_\gamma l_\delta) A_{\alpha\beta} A_{\gamma\delta} \end{aligned} \tag{2.6}$$

Similarly,

$$\begin{aligned} m_\alpha m_\beta m_\gamma m_\delta A_{\alpha\beta} A_{\gamma\delta} &= (m_\alpha m_\beta n_\gamma n_\delta + l_\alpha l_\beta m_\gamma m_\delta) A_{\alpha\beta} A_{\gamma\delta}, \\ n_\alpha n_\beta n_\gamma n_\delta A_{\alpha\beta} A_{\gamma\delta} &= (n_\alpha n_\beta l_\gamma l_\delta + m_\alpha m_\beta n_\gamma n_\delta) A_{\alpha\beta} A_{\gamma\delta}. \end{aligned} \tag{2.7}$$

Thus, D has only 12 terms for the incompressible case.

Note the constraints:

$$l_\alpha l_\alpha = m_\alpha m_\alpha = n_\alpha n_\alpha = 1. \tag{2.8}$$

Also,

$$l_\alpha = \varepsilon_{\alpha\beta\gamma} n_\beta m_\gamma \quad \text{or} \quad l_\alpha l_\beta + m_\alpha m_\beta + n_\alpha n_\beta = \delta_{\alpha\beta}. \tag{2.9}$$

Further,

$$\begin{aligned} \mathbf{m}\cdot\mathbf{N} + \mathbf{n}\cdot\mathbf{M} = 0, &\qquad \mathbf{l}\cdot\mathbf{M} + \mathbf{m}\cdot\mathbf{L} = 0, \\ \mathbf{n}\cdot\mathbf{L} + \mathbf{l}\cdot\mathbf{N} = 0 \quad \text{and} &\quad \mathbf{m}\cdot\mathbf{M} = \mathbf{n}\cdot\mathbf{N} = \mathbf{l}\cdot\mathbf{L} = 0. \end{aligned} \tag{2.10}$$

Therefore, using (2.5) and (2.6), obtain:

$$\begin{aligned} \eta_{10}(m_\gamma L_\gamma)^2 + \eta_{12}(l_\gamma N_\gamma)^2 &= \eta_{10}(l_\gamma M_\gamma)^2 + \eta_{12}(l_\gamma N_\gamma)^2 \\ &= \eta_{10} l_\gamma l_\delta M_\gamma M_\delta + \eta_{12} l_\gamma l_\delta N_\gamma N_\delta \\ &= \eta_{10}|\mathbf{M}|^2 - \eta_{10} m_\gamma M_\gamma m_\delta M_\delta - \eta_{10} n_\gamma n_\delta M_\gamma M_\delta \\ &\quad + \eta_{12}|\mathbf{N}|^2 - \eta_{12} m_\gamma m_\delta N_\gamma N_\delta - \eta_{12} n_\gamma n_\delta N_\gamma N_\delta \\ &= \eta_{10}|\mathbf{M}|^2 + \eta_{12}|\mathbf{N}|^2 - (\eta_{10} + \eta_{12})(\mathbf{n}\cdot\mathbf{M})^2. \end{aligned} \tag{2.11}$$

Now,

$$\begin{aligned} \eta_{13} l_\alpha m_\beta m_\gamma L_\gamma A_{\alpha\beta} &= -\eta_{13} l_\alpha m_\beta l_\gamma M_\gamma A_{\alpha\beta} \\ &= \eta_{13} m_\beta M_\gamma A_{\alpha\beta}(\delta_{\alpha\gamma} - m_\alpha m_\gamma - n_\alpha n_\gamma) \\ &= \eta_{13}(m_\beta M_\gamma A_{\gamma\beta} - m_\alpha m_\beta m_\gamma M_\gamma A_{\alpha\beta} + n_\alpha m_\beta n_\gamma M_\gamma A_{\alpha\beta}) \\ &= \eta_{13}(m_\beta M_\gamma A_{\gamma\beta} + n_\alpha m_\beta A_{\alpha\beta} n_\gamma M_\gamma). \end{aligned} \tag{2.12}$$

In addition, we have the following equations when the constraints are suitably used:

$$\begin{aligned} \eta_{15} n_\alpha l_\beta l_\gamma N_\gamma A_{\alpha\beta} &= \eta_{15} n_\alpha N_\gamma A_{\alpha\gamma} - \eta_{15}(n_\alpha m_\beta m_\gamma N_\gamma - n_\alpha n_\beta n_\gamma N_\gamma) A_{\alpha\beta} \\ &= \eta_{15}(n_\alpha N_\gamma A_{\alpha\gamma} + n_\alpha m_\beta A_{\alpha\beta} n_\gamma M_\gamma), \end{aligned} \tag{2.13}$$

$$\begin{aligned} (\eta_7 l_\alpha m_\beta l_\gamma m_\delta + \eta_9 n_\alpha l_\beta n_\gamma l_\delta) A_{\alpha\beta} A_{\gamma\delta} &= \eta_7 m_\alpha m_\gamma A_{\alpha\beta} A_{\beta\gamma} + \eta_9 n_\alpha n_\gamma A_{\alpha\beta} A_{\beta\gamma} \\ - (\eta_7 m_\alpha m_\beta m_\gamma m_\delta + \eta_9 n_\alpha n_\beta n_\gamma n_\delta) A_{\alpha\beta} A_{\gamma\delta} &- (\eta_7 + \eta_9) m_\alpha n_\beta m_\gamma n_\delta A_{\alpha\beta} A_{\gamma\delta}, \end{aligned} \tag{2.14}$$

$$\begin{aligned} (\eta_4 l_\alpha l_\beta m_\gamma m_\delta + \eta_6 n_\alpha n_\beta l_\gamma l_\delta) A_{\alpha\beta} A_{\gamma\delta} &= [-\eta_4 m_\alpha m_\beta m_\gamma m_\delta - \eta_6 n_\alpha n_\beta n_\gamma n_\delta \\ - (\eta_4 + \eta_6) m_\alpha m_\beta n_\gamma n_\delta] A_{\alpha\beta} A_{\gamma\delta} &+ \eta_4 m_\gamma m_\delta A_{\gamma\delta} A_{\alpha\alpha} + \eta_6 n_\alpha n_\beta A_{\alpha\beta} A_{\gamma\gamma}, \end{aligned} \tag{2.15}$$

and

$$
\begin{aligned}
(\mathbf{l}\cdot\mathbf{A}\mathbf{l})^2 &= (\mathrm{tr}\,\mathbf{A} - \mathbf{m}\cdot\mathbf{A}\mathbf{m} - \mathbf{n}\cdot\mathbf{A}\mathbf{n})^2 \\
&= (\mathrm{tr}\,\mathbf{A})^2 + (\mathbf{m}\cdot\mathbf{A}\mathbf{m})^2 + (\mathbf{n}\cdot\mathbf{A}\mathbf{n})^2 - 2(\mathrm{tr}\,\mathbf{A})(\mathbf{m}\cdot\mathbf{A}\mathbf{m}) \qquad (2.16) \\
&\quad - 2(\mathrm{tr}\mathbf{A})(\mathbf{n}\cdot\mathbf{A}\mathbf{n}) + 2(\mathbf{m}\cdot\mathbf{A}\mathbf{m})(\mathbf{n}\cdot\mathbf{A}\mathbf{n}).
\end{aligned}
$$

Therefore, the dissipation function D (2.5) reduces to:

$$
\begin{aligned}
D = {} & \frac{1}{2}\eta_1 A_{\alpha\alpha}A_{\gamma\gamma} + \Bigg[\left(\frac{1}{2}\eta_1 - \eta_7 + \frac{1}{2}\eta_2 - \eta_4\right) m_\alpha m_\beta m_\gamma m_\delta \\
& + \left(\frac{1}{2}\eta_1 - \eta_9 - \eta_6 + \frac{1}{2}\eta_3\right) n_\alpha n_\beta n_\gamma n_\delta + (\eta_1 - \eta_4 - \eta_6 + \eta_5) \\
& \cdot m_\alpha m_\beta n_\gamma n_\delta + (-\eta_7 - \eta_9 + \eta_8) m_\alpha n_\beta m_\gamma n_\delta\Bigg] A_{\alpha\beta}A_{\gamma\delta} + \frac{1}{2}\eta_{10}M_\gamma^2 \\
& + \frac{1}{2}\eta_{12}N_\gamma^2 + \frac{1}{2}(\eta_{11} - \eta_{10} - \eta_{12})(n_\gamma M_\gamma)^2 + \eta_{13} m_\beta M_\gamma A_{\gamma\beta} \qquad (2.17) \\
& + \eta_{15} n_\alpha N_\gamma A_{\gamma\alpha} + (\eta_{13} + \eta_{14} + \eta_{15}) n_\alpha m_\beta A_{\alpha\beta} n_\gamma M_\gamma \\
& + \eta_7 m_\alpha m_\gamma A_{\alpha\beta}A_{\beta\gamma} + \eta_9 n_\alpha n_\gamma A_{\alpha\beta}A_{\beta\gamma} + (\eta_4 - \eta_1) m_\gamma m_\delta A_{\gamma\delta}A_{\alpha\alpha} \\
& + (\eta_6 - \eta_1) n_\alpha n_\beta A_{\alpha\beta}A_{\gamma\gamma}.
\end{aligned}
$$

Now, we use the identity:

$$
\begin{aligned}
& \mathbf{n}\cdot\mathbf{A}^2\mathbf{n} + \mathbf{m}\cdot\mathbf{A}^2\mathbf{m} + (\mathbf{n}\cdot\mathbf{A}\mathbf{n})(\mathbf{m}\cdot\mathbf{A}\mathbf{m}) - (\mathbf{n}\cdot\mathbf{A}\mathbf{m})^2 + (\mathrm{tr}\,\mathbf{A})^2 \\
& \qquad - 2(\mathrm{tr}\,\mathbf{A})(\mathbf{m}\cdot\mathbf{A}\mathbf{m}) - 2(\mathrm{tr}\,\mathbf{A})(\mathbf{n}\cdot\mathbf{A}\mathbf{n}) = \frac{1}{2}\mathrm{tr}\,\mathbf{A}^2. \qquad (2.18)
\end{aligned}
$$

(The proof is given in Appendix A). Thus, one arrives at the 2-director dissipation function valid for compressible biaxial nematics:

$$
D = \frac{1}{2}(\eta_4 + \eta_6 - \eta_5)(\mathrm{tr}\,\mathbf{A})^2 + \left(\frac{1}{2}\eta_1 - \eta_4 - \eta_7 + \frac{1}{2}\eta_2\right)(\mathbf{m}\cdot\mathbf{A}\mathbf{m})^2
$$

$$+ \left(\frac{1}{2}\eta_1 - \eta_6 - \eta_9 + \frac{1}{2}\eta_3\right)(\mathbf{n}\cdot\mathbf{An})^2 + (\eta_1 - \eta_4 - \eta_6 - \eta_7$$

$$- \eta_9 + \eta_5 + \eta_8)(\mathbf{m}\cdot\mathbf{An})^2 + \frac{1}{2}\eta_{10}|\mathbf{M}|^2 + \frac{1}{2}\eta_{11}|\mathbf{N}|^2$$

$$+ \frac{1}{2}(\eta_{11} - \eta_{10} - \eta_{12})(\mathbf{n}\cdot\mathbf{M})^2 + \eta_{13}\mathbf{m}\cdot\mathbf{AM} + \eta_{15}\mathbf{n}\cdot\mathbf{AN}$$

$$+ (\eta_{13} + \eta_{14} + \eta_{15})(\mathbf{n}\cdot\mathbf{M})(\mathbf{n}\cdot\mathbf{Am}) + \frac{1}{2}(\eta_1 + \eta_5 - \eta_4 - \eta_6)\operatorname{tr}\mathbf{A}^2$$

$$+ (\eta_7 + \eta_4 + \eta_6 - \eta_1 - \eta_5)\mathbf{m}\cdot\mathbf{A}^2\mathbf{m} + (\eta_9 + \eta_4$$

$$+ \eta_6 - \eta_1 - \eta_5)\mathbf{n}\cdot\mathbf{A}^2\mathbf{n} + (\eta_1 - \eta_4 - 2\eta_6 + 2\eta_5)(\operatorname{tr}\mathbf{A})(\mathbf{m}\cdot\mathbf{Am})$$

$$+ (\eta_1 - \eta_6 - 2\eta_4 + 2\eta_5)(\operatorname{tr}\mathbf{A})(\mathbf{n}\cdot\mathbf{An}). \tag{2.19}$$

The first and the last two terms of (2.19) are absent for the incompressible case where tr $\mathbf{A} = 0$. The incompressible equation can be compared to the Chauré dissipation function[7]:

$$D = \chi_1 \operatorname{tr}\mathbf{A}^2 + \chi_2(\mathbf{n}\cdot\mathbf{An})^2 + \chi_3(\mathbf{m}\cdot\mathbf{Am})^2 + \chi_4\mathbf{n}\cdot\mathbf{A}^2\mathbf{n} + \chi_5\mathbf{m}\cdot\mathbf{A}^2\mathbf{m}$$

$$+ \chi_6(\mathbf{n}\cdot\mathbf{Am})^2 + \chi_7\mathbf{n}\cdot\mathbf{AN} + \chi_8\mathbf{m}\cdot\mathbf{AM} + \chi_9(\mathbf{m}\cdot\mathbf{N})(\mathbf{n}\cdot\mathbf{Am}) \tag{2.20}$$

$$+ \chi_{10}(\mathbf{m}\cdot\mathbf{N})^2 + \chi_{11}|\mathbf{N}|^2 + \chi_{12}|\mathbf{M}|^2.$$

The viscous coefficients for the 3-director model and the 2-director model are related as:

$$\begin{bmatrix} \chi_1 = \frac{1}{2}(\eta_1 + \eta_5 - \eta_4 - \eta_6) & \chi_2 = \frac{1}{2}\eta_1 - \eta_6 - \eta_9 + \frac{1}{2}\eta_3 \\ \chi_3 = \frac{1}{2}\eta_1 - \eta_4 - \eta_7 + \frac{1}{2}\eta_2 & \chi_4 = \eta_9 + \eta_4 + \eta_6 - \eta_1 - \eta_5 \\ \chi_5 = \eta_7 + \eta_4 + \eta_6 - \eta_1 - \eta_5 & \chi_6 = \eta_1 - \eta_4 - \eta_6 - \eta_7 - \eta_9 + \eta_5 + \eta_8 \\ \chi_7 = \eta_{15} & \chi_8 = \eta_{13} \\ \chi_9 = -(\eta_{13} + \eta_{14} + \eta_{15}) & \chi_{10} = \frac{1}{2}(\eta_{11} - \eta_{10} - \eta_{12}) \\ \chi_{11} = \frac{1}{2}\eta_{11} & \chi_{12} = \frac{1}{2}\eta_{10} \end{bmatrix} \tag{2.21}$$

Therefore, the Chauré dissipation function (compressible) has 3 more terms, i.e.,

$$\chi_{13}(\text{tr }\mathbf{A})^2 + \chi_{14}(\mathbf{m}\cdot\mathbf{Am})(\text{tr }\mathbf{A}) + \chi_{15}(\mathbf{n}\cdot\mathbf{An})(\text{tr }\mathbf{A}). \tag{2.22}$$

Also,

$$\chi_{13} = \frac{1}{2}(\eta_4 + \eta_6 - \eta_5), \qquad \chi_{14} = \eta_1 - \eta_4 - 2\eta_6 + 2\eta_5, \quad \text{and}$$

$$\chi_{15} = \eta_1 - \eta_6 - 2\eta_4 + 2\eta_5. \tag{2.23}$$

For the symmetric part of the mechanical stress tensor (compressible), we have:

$$\begin{aligned}\sigma_s^{ij} = \frac{\partial D}{\partial A_{ij}} &= 2\chi_1 A^{ij} + 2\chi_2(\mathbf{n}\cdot\mathbf{An})n^i n^j + 2\chi_3(\mathbf{m}\cdot\mathbf{Am})m^i m^j + \chi_4(A^{ip}n_p n^j \\ &+ A^{jp}n_p n^i) + \chi_5(A^{ip}m_p m^j + A^{jp}m_p m^i) + \chi_6(\mathbf{n}\cdot\mathbf{Am})(n^i m^j + m^i n^j) \\ &+ \frac{\chi_7}{2}(n^i N^j + n^j N^i) + \frac{\chi_8}{2}(m^i M^j + m^j M^i) + \frac{\chi_9}{2}(\mathbf{m}\cdot\mathbf{N}) \\ &\cdot(n^i m^j + m^i n^j) + 2\chi_{13}(\text{tr }\mathbf{A})\delta^{ij} + \chi_{14}(\text{tr }\mathbf{A})m^i m^j \\ &+ \chi_{14}(\mathbf{m}\cdot\mathbf{Am})\delta^{ij} + \chi_{15}(\text{tr }\mathbf{A})n^i n^j + \chi_{15}(\mathbf{n}\cdot\mathbf{An})\delta^{ij}.\end{aligned} \tag{2.24}$$

The antisymmetric part of the mechanical stress tensor is given by:

$$\sigma_a^{ij} = h^{[i}n^{j]} + k^{[i}m^{j]}, \tag{2.25}$$

where the director forces are given by:

$$\begin{aligned} h^i &= -\frac{\partial D}{\partial N_i} = -2\chi_{11}N^i - \chi_7 A^{ip}n_p - [2\chi_{10}(\mathbf{m}\cdot\mathbf{N}) + \chi_9(\mathbf{n}\cdot\mathbf{Am})]m^i \\ k^i &= -\frac{\partial D}{\partial M_i} = -2\chi_{12}M^i - \chi_8 A^{ip}m_p. \end{aligned} \tag{2.26}$$

Also,

$$h^{[i}n^{j]} \equiv \frac{1}{2}(h^i n^j - h^j n^i)$$

$$= \chi_{11}(N^j n^i - N^i n^j) + \frac{\chi_7}{2}(A^{jp}n_p n^i - A^{ip}n_p n^j)$$

$$+ \left[\chi_{10}(\mathbf{m}\cdot\mathbf{N}) + \frac{\chi_9}{2}(\mathbf{n}\cdot\mathbf{Am})\right](n^i m^j - n^j m^i)$$

$$k^{[i}m^{j]} \equiv \frac{1}{2}(k^i m^j - k^j m^i)$$

$$= \chi_{12}(M^j m^i - M^i m^j) + \frac{\chi_8}{2}(A^{jp}m_p m^i - A^{ip}m_p m^j). \quad (2.27)$$

The total mechanical stress tensor (compressible) is thus given by:

$$\tau^{ij} = \sigma_s^{ij} + h^{[i}n^{j]} + k^{[i}m^{j]}$$

$$= 2\chi_1 A^{ij} + 2\chi_2(\mathbf{n}\cdot\mathbf{An})n^i n^j + 2\chi_3(\mathbf{m}\cdot\mathbf{Am})m^i m^j$$

$$+ \left(\chi_6 + \frac{\chi_9}{2}\right)(\mathbf{n}\cdot\mathbf{Am})n^i m^j + \left(\chi_6 - \frac{\chi_9}{2}\right)(\mathbf{n}\cdot\mathbf{Am})n^j m^i$$

$$+ \left(\chi_4 - \frac{\chi_7}{2}\right)A^{ip}n_p n^j + \left(\chi_4 + \frac{\chi_7}{2}\right)A^{jp}n_p n^i + \left(\chi_5 - \frac{\chi_8}{2}\right)A^{ip}m_p m^j$$

$$+ \left(\chi_5 + \frac{\chi_8}{2}\right)A^{jp}m_p m^i + \left(\frac{\chi_9}{2} + \chi_{10}\right)(\mathbf{m}\cdot\mathbf{N})n^i m^j$$

$$+ \left(\frac{\chi_9}{2} - \chi_{10}\right)(\mathbf{m}\cdot\mathbf{N})n^j m^i + \left(\frac{\chi_7}{2} + \chi_{11}\right)N^j n^i + \left(\frac{\chi_7}{2} - \chi_{11}\right)N^i n^j$$

$$+ \left(\frac{\chi_8}{2} - \chi_{12}\right)M^i m^j + \left(\frac{\chi_8}{2} + \chi_{12}\right)M^j m^i + 2\chi_{13}(\mathrm{tr}\,\mathbf{A})\delta^{ij}$$

$$+ \chi_{14}(\mathrm{tr}\,\mathbf{A})m^i m^j + \chi_{14}(\mathbf{m}\cdot\mathbf{Am})\delta^{ij} + \chi_{15}(\mathrm{tr}\,\mathbf{A})n^i n^j + \chi_{15}(\mathbf{n}\cdot\mathbf{An})\delta^{ij} \quad (2.28)$$

Thus,

$$\tau = \nu_1\mathbf{A} + \nu_2(\mathbf{n}\cdot\mathbf{An})\mathbf{n}\otimes\mathbf{n} + \nu_3(\mathbf{m}\cdot\mathbf{Am})\mathbf{m}\otimes\mathbf{m} + \nu_4(\mathbf{n}\cdot\mathbf{Am})\mathbf{n}\otimes\mathbf{m}$$
$$+ \nu_5(\mathbf{n}\cdot\mathbf{Am})\mathbf{m}\otimes\mathbf{n} + \nu_6\mathbf{An}\otimes\mathbf{n} + \nu_7\mathbf{n}\otimes\mathbf{An}$$
$$+ \nu_8\mathbf{Am}\otimes\mathbf{m} + \nu_9\mathbf{m}\otimes\mathbf{Am} + \nu_{10}(\mathbf{m}\cdot\mathbf{N})\mathbf{n}\otimes\mathbf{m}$$
$$+ \nu_{11}(\mathbf{m}\cdot\mathbf{N})\mathbf{m}\otimes\mathbf{n} + \nu_{12}\mathbf{n}\otimes\mathbf{N} + \nu_{13}\mathbf{N}\otimes\mathbf{n} + \nu_{14}\mathbf{m}\otimes\mathbf{M}$$
$$\nu_{15}\mathbf{M}\otimes\mathbf{m} + \nu_{16}(\mathrm{tr}\,\mathbf{A})\mathbf{1} + \nu_{17}[(\mathrm{tr}\,\mathbf{A})\mathbf{m}\otimes\mathbf{m} + (\mathbf{m}\cdot\mathbf{Am})\mathbf{1}]$$
$$+ \nu_{18}[(\mathrm{tr}\,\mathbf{A})\mathbf{n}\otimes\mathbf{n} + (\mathbf{n}\cdot\mathbf{An})\mathbf{1}]. \qquad (2.29)$$

with

$$\begin{array}{lll}
\nu_1 = 2\chi_1 & \nu_2 = 2\chi_2 & \nu_3 = 2\chi_3 \\
\nu_4 = \chi_6 + \frac{\chi_9}{2} & \nu_5 = \chi_6 - \frac{\chi_9}{2} & \nu_6 = \chi_4 - \frac{\chi_7}{2} \\
\nu_7 = \chi_4 + \frac{\chi_7}{2} & \nu_8 = \chi_5 - \frac{\chi_8}{2} & \nu_9 = \chi_5 + \frac{\chi_8}{2} \\
\nu_{10} = \frac{\chi_9}{2} + \chi_{10} & \nu_{11} = \frac{\chi_9}{2} - \chi_{10} & \nu_{12} = \frac{\chi_7}{2} + \chi_{11} \\
\nu_{13} = \frac{\chi_7}{2} - \chi_{11} & \nu_{14} = \frac{\chi_8}{2} + \chi_{12} & \nu_{15} = \frac{\chi_8}{2} - \chi_{12} \\
\nu_{16} = 2\chi_{13} & \nu_{17} = \chi_{14} & \nu_{18} = \chi_{15}.
\end{array} \qquad (2.30)$$

Not all of these 18 ν's are independent. Note the three Onsager relations:

$$\nu_{12} + \nu_{13} = \nu_7 - \nu_6,\ \nu_{14} + \nu_{15} = \nu_9 - \nu_8,\ \text{and}\ \nu_{10} + \nu_{11} = \nu_4 - \nu_5. \qquad (2.31)$$

which can be obtained from (2.30). The first of these relations is associated with the director **n**, the second with the director **m** and the last one involves both the directors **n** and **m**. Therefore, for the compressible case, the number of independent viscosity coefficients is $18 - 3 = 15$, which agrees with the number Govers and Vertogen[15] got for the 3-director model. For the incompressible case, terms containing ν_{16}, ν_{17}, ν_{18} are absent. Thus, there are 12 independent viscosity coefficients for the incompressible case, in perfect agreement with Govers and Vertogen.[15]

We now express D as the sum of independent quadratics:

$$
\begin{aligned}
D = &\left(\chi_1 + \chi_4 - \frac{\chi_7^2}{4\chi_{11}} + \chi_2\right)(\mathbf{n}\cdot\mathbf{An})^2 + \left(\chi_1 + \chi_5 - \frac{\chi_8^2}{4\chi_{12}} + \chi_3\right)(\mathbf{m}\cdot\mathbf{Am})^2 \\
&+ \chi_1(\mathbf{l}\cdot\mathbf{Al})^2 + \left(2\chi_1 + \chi_4 - \frac{\chi_7^2}{4\chi_{11}}\right)(\mathbf{l}\cdot\mathbf{An})^2 + \left(2\chi_1 + \chi_5 - \frac{\chi_8^2}{4\chi_{12}}\right)(\mathbf{l}\cdot\mathbf{Am})^2 \\
&+ \left(2\chi_1 + \chi_4 - \frac{\chi_7^2}{4\chi_{11}} + \chi_5 - \frac{\chi_8^2}{4\chi_{12}} + \chi_6 - \frac{\chi_9^2}{4\chi_{10}}\right)(\mathbf{n}\cdot\mathbf{Am})^2 \\
&+ \chi_{11}\left(\mathbf{N} + \frac{\chi_7}{2\chi_{11}}\mathbf{An}\right)^2 + \chi_{12}\left(\mathbf{M} + \frac{\chi_8}{2\chi_{12}}\mathbf{Am}\right)^2 \\
&+ \chi_{10}\left(\mathbf{m}\cdot\mathbf{N} + \frac{\chi_9}{2\chi_{10}}(\mathbf{n}\cdot\mathbf{Am})\right)^2
\end{aligned} \tag{2.32}
$$

Positive definiteness of D then requires:

$$
\begin{aligned}
&\chi_1 + \chi_4 - \frac{\chi_7^2}{4\chi_{11}} + \chi_2 > 0 \rightarrow (\nu_1 + \nu_2 + \nu_6 + \nu_7)(\nu_{12} - \nu_{13}) > (\nu_7 - \nu_6)^2 \\
&\chi_1 + \chi_5 - \frac{\chi_8^2}{4\chi_{12}} + \chi_3 > 0 \rightarrow (\nu_1 + \nu_3 + \nu_8 + \nu_9)(\nu_{14} - \nu_{15}) > (\nu_9 - \nu_8)^2 \\
&\chi_1 > 0 \rightarrow \nu_1 > 0 \\
&2\chi_1 + \chi_4 - \frac{\chi_7^2}{4\chi_{11}} > 0 \rightarrow (2\nu_1 + \nu_6 + \nu_7)(\nu_{12} - \nu_{13}) > (\nu_7 - \nu_6)^2 \\
&2\chi_1 + \chi_5 - \frac{\chi_8^2}{4\chi_{12}} > 0 \rightarrow (2\nu_1 + \nu_8 + \nu_9)(\nu_{14} - \nu_{15}) > (\nu_9 - \nu_8)^2 \\
&\chi_{10} > 0 \rightarrow \nu_{10} > \nu_{11} \\
&\chi_{11} > 0 \rightarrow \nu_{12} > \nu_{13} \\
&\chi_{12} > 0 \rightarrow \nu_{14} > \nu_{15}
\end{aligned} \tag{2.33}
$$

and

$$2\chi_1 + \chi_4 - \frac{\chi_7^2}{4\chi_{11}} + \chi_5 - \frac{\chi_8^2}{4\chi_{12}} + \chi_6 - \frac{\chi_9^2}{4\chi_{10}}$$

$$> 0 \rightarrow (2\nu_1 + \nu_6 + \nu_7) - \frac{(\nu_7 - \nu_6)^2}{(\nu_{12} - \nu_{13})} + (\nu_8 + \nu_9) - \frac{(\nu_9 - \nu_8)^2}{(\nu_{14} - \nu_{15})}$$

$$+ (\nu_4 + \nu_5) - \frac{(\nu_4 - \nu_5)^2}{(\nu_{10} - \nu_{11})} \tag{2.34}$$

$$> 0.$$

3. CONSISTENCY CONDITIONS OF THE DISSIPATION FUNCTION

The viscosity coefficients for biaxial nematics are functions of two order parameters and temperature, just as the Ericksen-Leslie viscosity coefficients for a uniaxial nematics are functions of a single order parameter and temperature. The general order tensor theory of Macmillan[16] permits expressing these viscosity coefficients in terms of a power series of macroscopic parameters, such as the order parameters, with the coefficients now being functions of temperature alone. Doi's molecular theory[17] gives similar insights into the viscosity coefficients (see Marrucci,[9] Kuzzu and Doi,[18,19] and Berry[20]). One advantage of this approach is to obtain the uniaxial theory by setting the second biaxial order parameter equal to zero.

The general order tensor can be written as:

$$Q_{\alpha\beta} = S\left(n_\alpha n_\beta - \frac{1}{3}\delta_{\alpha\beta}\right) + \frac{B}{2}(m_\alpha m_\beta - l_\alpha l_\beta), \tag{3.1}$$

(This equation is the same as (2.2) except for a rescaling of B) where S and B are respectively the uniaxial and biaxial order parameters, and we have used (2.9). So, we have:

$$\begin{aligned}\mathbf{Q} &= S\left(\mathbf{n}\otimes\mathbf{n} - \frac{1}{3}\mathbf{1}\right) + \frac{B}{2}[\mathbf{m}\otimes\mathbf{m} - (\mathbf{1} - \mathbf{m}\otimes\mathbf{m} - \mathbf{n}\otimes\mathbf{n})] \\ &= \left(S + \frac{B}{2}\right)\mathbf{n}\otimes\mathbf{n} + B\mathbf{m}\otimes\mathbf{m} - \left(\frac{S}{3} + \frac{B}{2}\right)\mathbf{1},\end{aligned} \tag{3.2}$$

or

$$\mathbf{Q} = a\mathbf{n}\otimes\mathbf{n} + b\mathbf{m}\otimes\mathbf{m} - c\mathbf{1} \tag{3.3}$$

with

$$a \equiv \left(S + \frac{B}{2}\right), \quad b \equiv B, \quad \text{and} \quad c \equiv \left(\frac{S}{3} + \frac{B}{2}\right). \tag{3.4}$$

The eigenvalues of $\mathbf{Q}$ are: $2/3\ S$ (along $\mathbf{n}$), $-S/3 + B/2$ (along $\mathbf{m}$) and $-S/3 - B/2$ (along $\mathbf{l}$). Ericksen[8] obtained all the invariants of the MacMillan model[16] using the representation theorems of Smith[21] and Wang.[14] An objective dissipation function is reducible to a quadratic function of two symmetric traceless tensors. These were taken by Ericksen to be $\mathbf{A}$ and

$$\hat{\mathbf{Q}} = \dot{\mathbf{Q}} - \boldsymbol{\Omega}\cdot\mathbf{Q} + \mathbf{Q}\cdot\boldsymbol{\Omega} \tag{3.5}$$

which is the corotational time derivative used by MacMillan.[16] In his biaxial nematic theory, Chaure[7] assumed the dissipation function to be quadratic in $\mathbf{A}$ and $\mathbf{N}$ (or $\mathbf{M}$), the corotational time derivative of the director $\mathbf{n}$ (or $\mathbf{m}$). Using polynomials quadratic in $\mathbf{Q}$, Ericksen[8] obtained all the invariants of the ELP theory when a uniaxial order tensor is substituted for $\mathbf{Q}$.

When a general biaxial order tensor such as Equation (3.3) is substituted for $\mathbf{Q}$, we obtain the relevant terms as:

$$\mathrm{tr}\,\mathbf{A}^2$$

$$\begin{aligned}
\mathrm{tr}\,\mathbf{A}^2\mathbf{Q} &= \mathrm{tr}(a\mathbf{A}^2\mathbf{n}\otimes\mathbf{n} + b\mathbf{A}^2\mathbf{m}\otimes\mathbf{m} - c\mathbf{A}^2)\\
&= a\mathbf{n}\cdot\mathbf{A}^2\mathbf{n} + b\mathbf{m}\cdot\mathbf{A}^2\mathbf{m} - c\,\mathrm{tr}\,\mathbf{A}^2
\end{aligned}$$

$$\begin{aligned}
(\mathrm{tr}\,\mathbf{Q}^2)\mathrm{tr}\,\mathbf{A}^2 &= \mathrm{tr}(a^2\mathbf{n}\otimes\mathbf{n}\cdot\mathbf{n}\otimes\mathbf{n} + b^2\mathbf{m}\otimes\mathbf{m}\cdot\mathbf{m}\otimes\mathbf{m} + c^2\mathbf{1} - 2ac\mathbf{n}\otimes\mathbf{n}\\
&\quad - 2bc\mathbf{m}\otimes\mathbf{m})\mathrm{tr}\,\mathbf{A}^2\\
&= (a^2 + b^2 + 3c^2 - 2ac - 2bc)\mathrm{tr}\,\mathbf{A}^2
\end{aligned}$$

$$\begin{aligned}
\mathrm{tr}(\mathbf{QA})^2 &= \mathrm{tr}(a\mathbf{A}\cdot\mathbf{n}\otimes\mathbf{n} + b\mathbf{A}\mathbf{m}\otimes\mathbf{m} - c\mathbf{A})^2\\
&= a^2(\mathbf{n}\cdot\mathbf{An})^2 + b^2(\mathbf{m}\cdot\mathbf{Am})^2 + c^2\,\mathrm{tr}\,\mathbf{A}^2 + 2ab(\mathbf{n}\cdot\mathbf{An})(\mathbf{m}\cdot\mathbf{Am})\\
&\quad - 2ac\mathbf{n}\cdot\mathbf{A}^2\mathbf{n} - 2bc\mathbf{m}\cdot\mathbf{A}^2\mathbf{m}\\
&= a^2(\mathbf{n}\cdot\mathbf{An})^2 + b^2(\mathbf{m}\cdot\mathbf{Am})^2 + (c^2 + ab)\mathrm{tr}\,\mathbf{A}^2 + 2ab(\mathbf{n}\cdot\mathbf{Am})^2\\
&\quad - 2b(a + c)\,\mathbf{m}\cdot\mathbf{A}^2\mathbf{m} - 2a(b + c)\mathbf{n}\cdot\mathbf{A}^2\mathbf{n}
\end{aligned}$$

which was obtained by using the identity (A.5), given by:

$$\mathbf{n}\cdot\mathbf{A}^2\mathbf{n} + \mathbf{m}\cdot\mathbf{A}^2\mathbf{m} + (\mathbf{n}\cdot\mathbf{An})(\mathbf{m}\cdot\mathbf{Am}) - (\mathbf{n}\cdot\mathbf{Am})^2 = \frac{1}{2}\,\mathrm{tr}\,\mathbf{A}^2.$$

Also,

$$\begin{aligned}
\mathrm{tr}(\mathbf{Q}^2\mathbf{A}^2) &= a^2\,\mathrm{tr}(\mathbf{n}\otimes\mathbf{n}\mathbf{A}^2) + b^2\,\mathrm{tr}(\mathbf{m}\otimes\mathbf{m}\mathbf{A}^2) + c^2\,\mathrm{tr}\,\mathbf{A}^2 - 2ac\mathbf{n}\cdot\mathbf{A}^2\mathbf{n}\\
&\quad - 2bc\mathbf{m}\cdot\mathbf{A}^2\mathbf{m}\\
&= (a^2 - 2ac)\mathbf{n}\cdot\mathbf{A}^2\mathbf{n} + (b^2 - 2bc)\mathbf{m}\cdot\mathbf{A}^2\mathbf{m} + c^2\,\mathrm{tr}\,\mathbf{A}^2
\end{aligned}$$

$$\begin{aligned}
\mathrm{tr}(\hat{\mathbf{Q}}^2) &= \mathrm{tr}[a(\mathbf{N}\otimes\mathbf{n} + \mathbf{n}\otimes\mathbf{N}) + b(\mathbf{m}\otimes\mathbf{M} + \mathbf{M}\otimes\mathbf{m})]^2 \qquad \text{(cf. (B.1))}\\
&= 2a^2|\mathbf{N}|^2 + 2b^2|\mathbf{M}|^2 + ab(\mathbf{N}\otimes\mathbf{n}:\mathbf{M}\otimes\mathbf{m} + \mathbf{n}\otimes\mathbf{N}:\mathbf{m}\otimes\mathbf{M}\\
&\quad + \mathbf{M}\otimes\mathbf{m}:\mathbf{N}\otimes\mathbf{n} + \mathbf{m}\otimes\mathbf{M}:\mathbf{n}\otimes\mathbf{N})\\
&= 2a^2|\mathbf{N}|^2 + 2b^2|\mathbf{M}|^2 - 4ab(\mathbf{n}\cdot\mathbf{M})^2 \qquad \text{(cf. (2.6))}
\end{aligned}$$

$$\mathrm{tr}(\hat{\mathbf{Q}}^2\mathbf{Q}) = (a^3 - 2a^2c)|\mathbf{N}|^2 + (b^3 - 2b^2c)|\mathbf{M}|^2 + (2abc - a^2b - ab^2)(\mathbf{n}\cdot\mathbf{M})^2$$

Since

$$\hat{\mathbf{Q}}^2\mathbf{Q} = \{a^2(\mathbf{n}\otimes\mathbf{N}\cdot\mathbf{N}\otimes\mathbf{n} + \mathbf{N}\otimes\mathbf{N}) + b^2(\mathbf{m}\otimes\mathbf{M}\cdot\mathbf{M}\otimes\mathbf{m} + \mathbf{M}\otimes\mathbf{M}) + ab(\mathbf{N}\otimes\mathbf{n}\cdot\mathbf{M}\otimes\mathbf{m} + \mathbf{n}\otimes\mathbf{N}\cdot\mathbf{m}\otimes\mathbf{M} + \mathbf{M}\otimes\mathbf{m}\cdot\mathbf{N}\otimes\mathbf{n} + \mathbf{m}\otimes\mathbf{M}\cdot\mathbf{n}\otimes\mathbf{N})\}(a\mathbf{n}\otimes\mathbf{n} + b\mathbf{m}\otimes\mathbf{m} - c\mathbf{1}),$$

$$\operatorname{tr}\mathbf{Q}^2\operatorname{tr}\hat{\mathbf{Q}}^2 = (a^2 + b^2 + 3c^2 - 2ac - 2bc)(2a^2|\mathbf{N}|^2 + 2b^2|\mathbf{M}|^2 - 4ab(\mathbf{n}\cdot\mathbf{M})^2)$$

$$\operatorname{tr}(\mathbf{Q}\hat{\mathbf{Q}} - \hat{\mathbf{Q}}\mathbf{Q})^2 = -2a^4|\mathbf{N}|^2 - 2b^4|\mathbf{M}|^2 + 4ab(a^2 + b^2 - 3ab)(\mathbf{n}\cdot\mathbf{M})^2$$

$$\operatorname{tr}(\mathbf{A}\hat{\mathbf{Q}}) = 2a\mathbf{n}\cdot\mathbf{AN} + 2b\mathbf{m}\cdot\mathbf{AM}$$

$$\operatorname{tr}(\mathbf{AQ}\hat{\mathbf{Q}}) = (a^2 - 2ac)\mathbf{n}\cdot\mathbf{AN} + (b^2 - 2bc)\mathbf{m}\cdot\mathbf{AM}$$

$$\operatorname{tr}\mathbf{Q}^2\operatorname{tr}(\mathbf{A}\hat{\mathbf{Q}}) = (a^2 + b^2 + 3c^2 - 2ac - 2bc)(2a\mathbf{n}\cdot\mathbf{AN} + 2b\mathbf{m}\cdot\mathbf{AM})$$

$$\operatorname{tr}(\mathbf{Q}^2\mathbf{A}\hat{\mathbf{Q}}) = a(a^2 - 2ac + 2c^2)\mathbf{n}\cdot\mathbf{AN} + b(b^2 - 2bc + 2c^2)\mathbf{m}\cdot\mathbf{AM} - ab(a - b)(\mathbf{n}\cdot\mathbf{M})(\mathbf{m}\cdot\mathbf{An}) \tag{3.6}$$

These invariants reduce to those derived by Ericksen if one sets the biaxial order parameter (B) equal to zero, i.e., $a = S$, $b = 0$, and $c = S/3$.

We multiply these 13 invariants by A_1 through A_{13}, add and obtain:

$$\frac{\nu_1}{2} = \chi_1 = A_1 - A_2c + A_3(a^2 + b^2 + 3c^2 - 2ac - 2bc) + A_4(c^2 + ab) + A_5c^2$$

$$\frac{\nu_2}{2} = \chi_2 = A_4a^2$$

$$\frac{\nu_3}{2} = \chi_3 = A_4b^2$$

$$\frac{(\nu_6 + \nu_7)}{2} = \chi_4 = A_2a - 2A_4a(b + c) - 2A_5ac$$

$$\frac{(\nu_8 + \nu_9)}{2} = \chi_5 = A_2b - 2A_4b(a + c) - 2A_5bc$$

$$\frac{(\nu_4 + \nu_5)}{2} = \chi_6 = 2A_4ab$$

$$\nu_{12} + \nu_{13} = \nu_7 - \nu_6$$

$$\nu_7 - \nu_6 = \chi_7 = 2A_{10}a + A_{11}(a^2 - 2ac) + 2A_{12}a(a^2 + b^2 + 3c^2 - 2ac - 2bc) + A_{13}a(a^2 + 2c^2 - 2ac)$$

$$\nu_{14} + \nu_{15} = \nu_9 - \nu_8$$

$$\nu_9 - \nu_8 = \chi_8 = 2A_{10}b + A_{11}(b^2 - 2bc) + 2A_{12}b(a^2 + b^2 + 3c^2 - 2ac - 2bc) + A_{13}b(b^2 - 2bc + 2c^2)$$

$$\nu_{10} + \nu_{11} = \nu_4 - \nu_5$$
$$\nu_4 - \nu_5 = \chi_9 = -A_{13}ab(a - b)$$
$$\frac{(\nu_{10} - \nu_{11})}{2} = \chi_{10} = -4A_6ab + A_7ab(2c - a - b) - 4A_8ab(a^2 + b^2 + 3c^2 - 2ac - 2bc) + 4A_9ab(a^2 + b^2 - 3ab)$$
$$\frac{\nu_{12} - \nu_{13}}{2} = \chi_{11} = 2A_6a^2 + A_7(a^3 - 2a^2c) + 2A_8a^2(a^2 + b^2 + 3c^2 - 2ac - 2bc) - 2A_9a^4$$
$$\frac{\nu_{14} - \nu_{15}}{2} = \chi_{12} = 2A_6b^2 + A_7(b^3 - 2b^2c) + 2A_8b^2(a^2 + b^2 + 3c^2 - 2ac - 2bc) - 2A_9b^4 \quad (3.7)$$

Thus we get:

$$\nu_1 = 2A_1 - \frac{2}{3}A_2S + \frac{2}{3}\left(2A_3 + \frac{1}{3}A_4 + \frac{1}{3}A_5\right)S^2 - A_2B + \frac{1}{2}(2A_3 + 3A_4 + A_5)B^2 + \frac{2}{3}(4A_4 + A_5)SB$$
$$\nu_2 = 2A_4\left(S + \frac{B}{2}\right)^2$$
$$\nu_3 = 2A_4B^2$$
$$\nu_4 = 2A_4B\left(S + \frac{B}{2}\right) - \frac{A_{13}}{2}B\left(S^2 - \frac{B^2}{4}\right)$$
$$\nu_5 = 2A_4B\left(S + \frac{B}{2}\right) + \frac{A_{13}}{2}B\left(S^2 - \frac{B^2}{4}\right)$$
$$\nu_6 = (A_2 - A_{10})\left(S + \frac{B}{2}\right) - \frac{1}{3}\left(2A_4 + 2A_5 + \frac{A_{11}}{2}\right)S^2 - \frac{1}{2}\left(3A_4 + A_5 - \frac{A_{11}}{4}\right)B^2 - \frac{1}{3}\left(10A_4 + 4A_5 - \frac{A_{11}}{2}\right)BS - \left(S + \frac{B}{2}\right)\left[\frac{1}{3}\left(2A_{12} + \frac{5}{6}A_{13}\right)S^2 + \frac{1}{2}\left(A_{12} + \frac{A_{13}}{4}\right)B^2 + \frac{A_{13}}{6}BS\right]$$
$$\nu_7 = (A_2 + A_{10})\left(S + \frac{B}{2}\right) - \frac{1}{3}\left(2A_4 + 2A_5 - \frac{A_{11}}{2}\right)S^2 - \frac{1}{2}\left(3A_4 + A_5 - \frac{3}{4}A_{11}\right)B^2 - \frac{1}{3}\left(10A_4 + 4A_5 + \frac{A_{11}}{2}\right)BS + \left(S + \frac{B}{2}\right)\left[\frac{1}{3}\left(2A_{12} + \frac{5}{6}A_{13}\right)S^2 + \frac{1}{2}\left(A_{12} + \frac{A_{13}}{4}\right)B^2 + \frac{A_{13}}{6}BS\right]$$

$$\nu_8 = (A_2 - A_{10})B - (2A_4 + A_5)B^2 - \frac{1}{3}(8A_4 + 2A_5 - A_{11})BS - \frac{1}{3}\left(2A_{12} + \frac{A_{13}}{3}\right)BS^2 - \frac{1}{4}(2A_{12} + A_{13})B^3$$

$$\nu_9 = (A_2 + A_{10})B - (2A_4 + A_5)B^2 - \frac{1}{3}(8A_4 + 2A_5 + A_{11})BS + \frac{1}{3}\left(2A_{12} + \frac{A_{13}}{3}\right)BS^2 + \frac{1}{4}(2A_{12} + A_{13})B^3$$

$$\nu_{10} = -4A_6B\left(S + \frac{B}{2}\right) - B\left(S + \frac{B}{2}\right)\left[\left(\frac{A_7}{3} + \frac{A_{13}}{2}\right)S + \left(A_7 - \frac{A_{13}}{2}\right)\frac{B}{2}\right] - 4B\left(S + \frac{B}{2}\right)\left[\frac{1}{4}(2A_8 + A_9)B^2 + \left(\frac{2}{3}A_8 - A_9\right)S^2 + 2A_9BS\right]$$

$$\nu_{11} = 4A_6B\left(S + \frac{B}{2}\right) + B\left(S + \frac{B}{2}\right)\left[\left(\frac{A_7}{3} - \frac{A_{13}}{2}\right)S + \left(A_7 + \frac{A_{13}}{2}\right)\frac{B}{2}\right] + 4B\left(S + \frac{B}{2}\right)\left[\frac{1}{4}(2A_8 + A_9)B^2 + \left(\frac{2}{3}A_8 - A_9\right)S^2 + 2A_9SB\right]$$

$$\begin{aligned}\nu_{12} = {} & A_{10}\left(S + \frac{B}{2}\right) + \left(\frac{A_{11}}{6} + 2A_6\right)S^2 + \frac{1}{2}\left(A_6 + \frac{A_{11}}{4}\right)B^2 \\ & - \frac{1}{2}\left(\frac{A_{11}}{3} - 4A_6\right)BS + \left(S + \frac{B}{2}\right)\left(\frac{5}{6}A_{13} + 2A_{12} + A_7\right)\frac{S^2}{3} \\ & + \frac{1}{4}\left(\frac{A_{13}}{2} + 2A_{12} - A_7\right)B^2\left(S + \frac{B}{2}\right) \\ & + \frac{1}{6}(A_{13} - 2A_7)BS\left(S + \frac{B}{2}\right) + \left(-2A_9 + \frac{4}{3}A_8\right)S^2\left(S + \frac{B}{2}\right)^2 \\ & + \frac{1}{2}(-A_9 + 2A_8)B^2\left(S + \frac{B}{2}\right)^2 - 2A_9BS\left(S + \frac{B}{2}\right)^2\end{aligned}$$

$$\begin{aligned}\nu_{13} = {} & A_{10}\left(S + \frac{B}{2}\right) + \left(\frac{A_{11}}{6} - 2A_6\right)S^2 - \frac{1}{2}\left(A_6 + \frac{A_{11}}{4}\right)B^2 \\ & - \left(2A_6 + \frac{A_{11}}{6}\right)BS + \frac{1}{3}\left(\frac{5}{6}A_{13} + 2A_{12} - A_7\right)S^2\left(S + \frac{B}{2}\right) \\ & + \frac{1}{4}\left(\frac{A_{13}}{2} + 2A_{12} + A_7\right)B^2\left(S + \frac{B}{2}\right) \\ & + \frac{1}{6}(A_{13} + 2A_7)BS\left(S + \frac{B}{2}\right) + \left(-\frac{4}{3}A_8 + 2A_9\right)S^2\left(S + \frac{B}{2}\right)^2 \\ & + \frac{1}{2}(-2A_8 + A_9)B^2\left(S + \frac{B}{2}\right)^2 + 2A_9BS\left(S + \frac{B}{2}\right)^2\end{aligned}$$

$$\nu_{14} = A_{10}B + 2A_6B^2 - \frac{1}{3}A_{11}BS - \frac{2}{3}A_7B^2S + \frac{1}{4}(2A_{12} + A_{13})B^3$$
$$+ \frac{1}{6}\left(2A_{12} + \frac{A_{13}}{3}\right)BS^2 + (A_8 - 2A_9)B^4 + \frac{4}{3}A_8B^2S^2$$
$$\nu_{15} = A_{10}B - 2A_6B^2 - \frac{1}{3}A_{11}BS + \frac{2}{3}A_7B^2S + \frac{1}{4}(2A_{12} + A_{13})B^3$$
$$+ \frac{1}{6}\left(2A_{12} + \frac{A_{13}}{3}\right)BS^2 + (2A_9 - A_8)B^4 - \frac{4}{3}A_8B^2S^2. \tag{3.8}$$

The ν-coefficients satisfy the Onsager relations indicated in (2.31). When we set the biaxial order parameter (B) equal to zero, we obtain:

$$\nu_3 = \nu_4 = \nu_5 = \nu_8 = \nu_9 = \nu_{10} = \nu_{11} = \nu_{14} = \nu_{15} = 0. \tag{3.9}$$

Two of the Onsager relations (those associated with **m**, and the mixed type associated with both **m** and **n**) become non-entities. Thus, we are left with one Onsager relation and six viscosity coefficients as follows:

$$\nu_1 = 2A_1 - \frac{2}{3}A_2S + \left(\frac{4}{3}A_3 + \frac{2}{9}A_4 + \frac{2}{9}A_5\right)S^2$$
$$\nu_2 = 2A_4S^2$$
$$\nu_6 = (A_2 - A_{10})S - \left(\frac{2}{3}A_4 + \frac{2}{3}A_5 + \frac{A_{11}}{6}\right)S^2 - \frac{1}{3}\left(2A_{12} + \frac{5}{6}A_{13}\right)S^3$$
$$\nu_7 = (A_2 + A_{10})S - \left(\frac{2}{3}A_4 + \frac{2}{3}A_5 - \frac{A_{11}}{6}\right)S^2 + \frac{1}{3}\left(2A_{12} + \frac{5}{6}A_{13}\right)S^3 \tag{3.10}$$
$$\nu_{12} = A_{10}S + \left(\frac{A_{11}}{6} + 2A_6\right)S^2 + \left(\frac{5}{6}A_{13} + 2A_{12} + A_7\right)\cdot\frac{S^3}{3} + 2\left(-A_9 + \frac{2}{3}A_8\right)S^4$$
$$\nu_{13} = A_{10}S + \left(\frac{A_{11}}{6} - 2A_6\right)S^2 + \left(\frac{5}{6}A_{13} + 2A_{12} - A_7\right)\cdot\frac{S^3}{3} + 2\left(-\frac{2}{3}A_8 + A_9\right)S^4$$

It can be checked that the Onsager relation for the uniaxial case:

$$\nu_7 - \nu_6 = \nu_{12} + \nu_{13} \tag{3.11}$$

is satisfied. This is the uniaxial Leslie-Ericksen theory.

4. MOLECULAR APPROACH

In this section, the aim is to calculate the viscosity coefficients appearing in the Chauré continuum theory[7] for biaxial nematic liquid crystals in terms of the average diffusion coefficient $\bar{D}_r$, concentration c, temperature T, and the uniaxial and biaxial order parameters: S and B respectively. We note here that Marrucci[9] used Doi's molecular theoretical approach[17] to calculate the Leslie viscosity coefficients[12,13] for the uniaxial nematic system. The latter is observed in rod-like molecular assemblies. Solutions of biopolymers such as TMV, PBLG, actin and tubulin are uniaxial nematics. However, polymer molecules are often rigid and lath-like. Copolyesters of hydroxy benzoic and hydroxy naphthoic acids are known to be biaxial nematics,[21] although biaxiality in biopolymers is yet to be evidenced. The polymer backbone confers a great deal of viscoelasticity to the system such that molecular rotational motion about the long axes is curbed. The effect is the development of correlation of molecular rotation about the long axes over a distance that is considerable relative to a single molecular diameter, leading to the biaxial characteristics of these systems.

Here we adopt the Marrucci approach and extend it to the biaxial case. We model the biaxial moelcule as a rectangular plate, as depicted in Figure 1. The vector **u** connects two ends of the molecule, e.g., a body diagonal. As shown by Doi[22], the kinetic equation for the **u** orientational distribution function $f(\mathbf{u}, t)$ is:

$$\frac{\partial f}{\partial t} = \bar{D}_r \nabla_{\mathbf{u}} \cdot \left[\nabla_{\mathbf{u}} f + \frac{f}{k_B T} \nabla_{\mathbf{u}} V_{\mathrm{tot}} \right] - \nabla_{\mathbf{u}} \cdot \dot{\mathbf{u}} f \tag{4.1}$$

where $V_{\mathrm{tot}}(\mathbf{u})$ is the total potential acting on the plate (due to the mean field and the external electromagnetic fields) and $\bar{D}_r$ is the effective rotational diffusion coefficient given by[17]:

$$\bar{D}_r \approx \nu_1 D_{r0} (cL^3)^{-2} \left(1 - \frac{3}{2}\mathbf{S}:\mathbf{S}\right)^{-2} \tag{4.2}$$

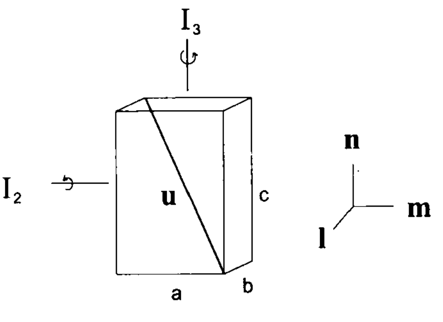

FIGURE 1 Model of a molecule forming the biaxial nematic phase. The object is a rectangular parallelepiped with the three sides unequal, $a \neq b \neq c$. The body diagonal **u** is shown by the straight bold line. Principal axes of inertia I_2 and I_3 are also shown in the diagram. The axis corresponding to the moment of inertia I_1 is normal to the plane of paper.

where L is the length of the body diagonal and D_{r0} is the diffusion constant of the body taken a rod about its body diagonal and is given by[17]:

$$D_{r0} = k_B T \ln(L/d)(3\pi\eta_s L^3)^{-1} \quad (4.3)$$

and η_s is the solvent viscosity. Also, $\dot{\mathbf{u}}$ is the rate of change due to the velocity gradient $\mathbf{K}$ and

$$\dot{\mathbf{u}} = \mathbf{K}\cdot\mathbf{u} - (\mathbf{u}\cdot\mathbf{K}\cdot\mathbf{u})\mathbf{u}. \quad (4.4)$$

The form for $\nabla_{\mathbf{u}}$ and pertinent details are given in Appendix C. The order tensor is defined by:

$$\mathbf{S} \equiv \left\langle \mathbf{u}\otimes\mathbf{u} - \frac{1}{3}\mathbf{1}\right\rangle. \quad (4.5)$$

In the same spirit as the uniaxial case, we assume:

$$\langle \mathbf{u}\otimes\mathbf{u}\rangle_{eq} \equiv \frac{1}{3}\mathbf{1} + S\left(\mathbf{n}\otimes\mathbf{n} - \frac{1}{3}\mathbf{1}\right) + B\left(\mathbf{m}\otimes\mathbf{m} - \frac{1}{3}\mathbf{1}\right) + \sqrt{BS}(\mathbf{n}\otimes\mathbf{m} + \mathbf{m}\otimes\mathbf{n}), \quad (4.6)$$

where

$$S \equiv \int d^2\mathbf{u} f(\mathbf{u}) \frac{1}{2}(3(\mathbf{u}\cdot\mathbf{n})^2 - 1), \quad (4.7)$$

and

$$B \equiv \int d^2\mathbf{u} f(\mathbf{u}) \frac{1}{2}(3(\mathbf{u}\cdot\mathbf{m})^2 - 1). \quad (4.8)$$

The definition of B differs from that of the B used in Section 3. The latter is given by:

$$B \equiv \int d^2\mathbf{u} f(\mathbf{u})\left((\mathbf{u}\cdot\mathbf{m})^2 - (\mathbf{u}\cdot\mathbf{l})^2\right)$$

This applies to the B used in (4.9).

Equation (4.6) satisfies the condition: tr $\mathbf{S}_{eq} = 0$; and as B goes to zero, $\mathbf{S}_{eq} = S(\mathbf{n}\otimes\mathbf{n} - 1/3\,\mathbf{1})$, which is valid for the uniaxial case. The form:

$$\mathbf{S} = S\left(\mathbf{n}\otimes\mathbf{n} - \frac{1}{3}\mathbf{1}\right) + B\left(\mathbf{m}\otimes\mathbf{m} - \frac{1}{3}\mathbf{1}\right) - B(\mathbf{n}\otimes\mathbf{m} + \mathbf{m}\otimes\mathbf{n}), \quad (4.9)$$

which is widely used in the literature,[15] will be studied in a later paper. As pointed out by Dzyaloshinskii,[23] any form such as:

$$S\left(\mathbf{n}\otimes\mathbf{n} - \frac{1}{3}\mathbf{1}\right) + C\left(\mathbf{m}\otimes\mathbf{m} - \frac{1}{3}\mathbf{1}\right) + D(\mathbf{n}\otimes\mathbf{m} + \mathbf{m}\otimes\mathbf{n}) \quad (4.10)$$

is suitable as a representation for the biaxial order tensor, provided it satisfies the above conditions. The form (4.6) is chosen here to match (4.12) for the biaxial molecular field, so that a reasonable equilibrium condition (4.19) can be found. Also,

$$V_{\text{tot}} = -\frac{3}{2}k_BTU\,\langle\mathbf{u}\otimes\mathbf{u}\rangle:\mathbf{u}\otimes\mathbf{u} - \frac{1}{2}\mathbf{u}\otimes\mathbf{u}:\mathbf{x}\otimes\mathbf{x}, \quad (4.11)$$

where

$$\mathbf{x} \equiv \sqrt{\chi_a}\,\mathbf{H} + \sqrt{\varepsilon_a}\mathbf{E} \quad (4.12)$$

is the resultant of the crossed electric (**E**) and magnetic (**H**) fields, each scaled with the appropriate susceptibilities. Further,

$$U = \nu c d L^2 \quad (4.13)$$

stands for the strength of the molecular interaction potential, as in the theories of Doi[17] and Zwanzig,[24] with c representing the number density, L the length of the body diagonal, d the maximum width perpendicular to the body diagonal, and ν is a numerical factor. Here U is taken as a parameter which is a function of S and B. Also, χ_a is the anisotropy in the magnetic susceptibility, i.e. $\chi_a = \chi_\parallel - \chi_\perp$. $\chi_\parallel$ is the magnetic susceptibility along the director **n**, and $\chi_\perp$ is the average susceptibility perpendicular to **n**. Similarly, $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$, where $\varepsilon_\parallel$ and $\varepsilon_\perp$ (average) are the dielectric constants along and perpendicular to the director **m**. Experimentally, if only a magnetic field is applied, the body will be oriented along and rotating about **n**. One can then measure $\chi_\parallel$ and $\chi_\perp$. Similarly, when an electric field is applied, the body will be oriented along and rotating about **m**. One thus measures $\varepsilon_\parallel$ and $\varepsilon_\perp$.

Using the Marrucci technique, we obtain:

$$\frac{d\mathbf{S}}{dt} = \mathbf{F}(\mathbf{S}) + \mathbf{M}(\mathbf{S}) + \mathbf{G}(\mathbf{S}), \quad (4.14)$$

where

$$\mathbf{F}(\mathbf{S}) \equiv -6\bar{D}_r\left\{\left(1 - \frac{U}{3}\right)\mathbf{S} - U\left[\mathbf{S}\cdot\mathbf{S} - \frac{1}{3}(\mathbf{S}:\mathbf{S})\mathbf{1}\right] + U(\mathbf{S}:\mathbf{S})\mathbf{S}\right\}, \quad (4.15)$$

$$\mathbf{G}(\mathbf{S}) \equiv \frac{1}{3}(\mathbf{K} + \mathbf{K}^T) + (\mathbf{K}\cdot\mathbf{S} + \mathbf{S}\cdot\mathbf{K}^T) - \frac{2}{3}(\mathbf{K}:\mathbf{S})\mathbf{1} - 2(\mathbf{K}:\mathbf{S})\mathbf{S}, \quad (4.16)$$

and

$$\mathbf{M}(\mathbf{S}) \equiv \frac{\bar{\mathbf{D}}_r}{k_B T}\,[\langle \mathbf{u}\otimes\mathbf{u}\rangle\cdot\mathbf{x}\otimes\mathbf{x} + \mathbf{x}\otimes\mathbf{x} \cdot\langle \mathbf{u}\otimes\mathbf{u}\rangle - 2(\langle \mathbf{u}\otimes\mathbf{u}\rangle:\mathbf{x}\otimes\mathbf{x})\langle \mathbf{u}\otimes\mathbf{u}\rangle]. \tag{4.17}$$

In the absence of a velocity gradient, the equilibrium condition is given by:

$$\mathbf{F}(\mathbf{S}_{\mathrm{eq}}) + \mathbf{M}(\mathbf{S}_{\mathrm{eq}}) = 0 \tag{4.18}$$

This leads to

$$\left(1 - \frac{U}{3}\right) - \frac{1}{3}\,U(S + B) + \frac{2}{3}\,U(S + B)^2 = \frac{\chi_a H^2}{9k_B TS}\,\{1 + (S + B) - 2(S + B)^2\} \tag{4.19}$$

Also,

$$\frac{\varepsilon_a E^2}{\chi_a H^2} = \frac{B}{S}. \tag{4.20}$$

Therefore, the stress tensor becomes:

$$\sigma = -\frac{ck_B T}{2\bar{D}_r}\left\{\mathbf{F}(\mathbf{S}) + \mathbf{M}(\mathbf{S}) - \frac{\bar{D}_r\chi_a}{k_B T}\,(\mathbf{x}\otimes\mathbf{x}\cdot\mathbf{S} - \mathbf{S}\cdot\mathbf{x}\otimes\mathbf{x})\right\} \tag{4.21}$$

For a small velocity gradient (slow flow),

$$\mathbf{S} = \mathbf{S}_{\mathrm{eq}} + \mathbf{S}' \tag{4.22}$$

where $\mathbf{S}'$ is the deviation of the order tensor from its equilibrium value, $\mathbf{S}_{\mathrm{eq}}$, due to the slow flow velocity gradient $\mathbf{K}$. Also, from (4.14), when $\mathbf{K}$ varies slowly along trajectories and memory effects are neglected,

$$\mathbf{F}(\mathbf{S}) + \mathbf{M}(\mathbf{S}) + \mathbf{G}(\mathbf{S}) = 0, \tag{4.23}$$

and

$$\sigma = \frac{ck_BT}{2\bar{D}_r}\left\{\mathbf{G}(\mathbf{S}_{eq}) + \frac{\bar{D}_r\chi_a H^2}{k_BT}\left[(\mathbf{n}\otimes\mathbf{n}\cdot\mathbf{S}' - \mathbf{S}'\cdot\mathbf{n}\otimes\mathbf{n}) + \sqrt{\frac{B}{S}}(\mathbf{n}\otimes\mathbf{m}\cdot\mathbf{S}' - \mathbf{S}'\cdot\mathbf{n}\otimes\mathbf{m}) + \sqrt{\frac{B}{S}}(\mathbf{m}\otimes\mathbf{n}\cdot\mathbf{S}' - \mathbf{S}'\cdot\mathbf{m}\otimes\mathbf{n}) + \frac{B}{S}(\mathbf{m}\otimes\mathbf{m}\cdot\mathbf{S}' - \mathbf{S}'\cdot\mathbf{m}\otimes\mathbf{m})\right]\right\} \tag{4.24}$$

Subtracting (4.18) from (4.23), we obtain:

$$\mathbf{F}(\mathbf{S}) - \mathbf{F}(\mathbf{S}_{eq}) + \mathbf{M}(\mathbf{S}) - \mathbf{M}(\mathbf{S}_{eq}) = \mathbf{G}(\mathbf{S}) \approx -\mathbf{G}(\mathbf{S}_{eq}) \tag{4.25}$$

or

$$\begin{aligned}
&\left\{\left(1 - \frac{U}{3}\right) + \frac{2}{3}U(S+B) + \frac{2}{3}U(S+B)^2 + \frac{2}{3}\bar{\chi}\left[1 + \frac{B}{S} + \frac{2}{S}(S+B)^2\right]\right\}\mathbf{S}' \\
&- (US+\bar{\chi})(\mathbf{n}\otimes\mathbf{n}\cdot\mathbf{S}' + \mathbf{S}'\cdot\mathbf{n}\otimes\mathbf{n}) + 2S(US+\bar{\chi})(\mathbf{S}':\mathbf{n}\otimes\mathbf{n})\mathbf{n}\otimes\mathbf{n} \\
&- (US+\bar{\chi})\frac{B}{S}(\mathbf{m}\otimes\mathbf{m}\cdot\mathbf{S}' + \mathbf{S}'\cdot\mathbf{m}\otimes\mathbf{m}) + 2\frac{B^2}{S}(US+\bar{\chi}) \\
&\cdot(\mathbf{S}':\mathbf{m}\otimes\mathbf{m})\mathbf{m}\otimes\mathbf{m} + 2B(US+\bar{\chi})[(\mathbf{n}\otimes\mathbf{m}:\mathbf{S}')\mathbf{m}\otimes\mathbf{n} + (\mathbf{S}':\mathbf{n}\otimes\mathbf{m}) \\
&\mathbf{n}\otimes\mathbf{m}] - (US+\bar{\chi})\sqrt{BS}(\mathbf{n}\otimes\mathbf{m}\cdot\mathbf{S}' + \mathbf{S}'\cdot\mathbf{n}\otimes\mathbf{m} + \mathbf{m}\otimes\mathbf{n}\cdot\mathbf{S}' \\
&+ \mathbf{S}'\cdot\mathbf{m}\otimes\mathbf{n}) + \frac{2}{3}(1-S-B)(US+\bar{\chi})\left[\sqrt{\frac{B}{S}}[(\mathbf{S}':\mathbf{n}\otimes\mathbf{m}) \right. \\
&\left. + (\mathbf{n}\otimes\mathbf{m}:\mathbf{S}')] + (\mathbf{n}\otimes\mathbf{n}:\mathbf{S}') + \frac{B}{S}(\mathbf{m}\otimes\mathbf{m}:\mathbf{S}')\right]\mathbf{1} \\
&= \frac{1}{6\bar{D}_r}\{(1-S-B)(\mathbf{K}+\mathbf{K}^{\top}) + S(\mathbf{K}\cdot\mathbf{n}\otimes\mathbf{n} + \mathbf{n}\otimes\mathbf{n}\cdot\mathbf{K}^{\top}) \\
&+ \sqrt{BS}(\mathbf{K}\cdot\mathbf{n}\otimes\mathbf{m} + \mathbf{n}\otimes\mathbf{m}\cdot\mathbf{K}^{\top} + \mathbf{K}\cdot\mathbf{m}\otimes\mathbf{n} + \mathbf{m}\otimes\mathbf{n}\cdot\mathbf{K}^{\top}) \\
&+ B(\mathbf{K}\cdot\mathbf{m}\otimes\mathbf{m} + \mathbf{m}\otimes\mathbf{m}\cdot\mathbf{K}^{\top}) - 2S^2(\mathbf{K}:\mathbf{n}\otimes\mathbf{n})\mathbf{n}\otimes\mathbf{n} - 2B^2(\mathbf{K}:\mathbf{m}\otimes\mathbf{m})\mathbf{m}\otimes\mathbf{m} \\
&- 2BS[(\mathbf{K}:\mathbf{n}\otimes\mathbf{m})\mathbf{n}\otimes\mathbf{m} + (\mathbf{K}:\mathbf{m}\otimes\mathbf{n})\mathbf{m}\otimes\mathbf{n}] - \frac{2}{3}(1-S-B)[S(\mathbf{K}:\mathbf{n}\otimes\mathbf{n}) \\
&+ B(\mathbf{K}:\mathbf{m}\otimes\mathbf{m})]\mathbf{1} + \sqrt{BS}[\mathbf{K}:\mathbf{n}\otimes\mathbf{m} + (\mathbf{K}:\mathbf{m}\otimes\mathbf{n})]\mathbf{1}\}.
\end{aligned} \tag{4.26}$$

where $\tilde{\chi} \equiv \chi_a H^2/6k_BT$. Pre- and post-multiplying with $\mathbf{n}\otimes\mathbf{n} + (B/S)\mathbf{m}\otimes\mathbf{m} + (B/S)^{1/2}(\mathbf{n}\otimes\mathbf{m} + \mathbf{m}\otimes\mathbf{n})$ and taking the difference, obtain:

$$\frac{\bar{D}_r\chi_a H^2}{k_BT}\left\{(\mathbf{n}\otimes\mathbf{n}\cdot\mathbf{S}' - \mathbf{S}'\cdot\mathbf{n}\otimes\mathbf{n}) + \frac{B}{S}(\mathbf{m}\otimes\mathbf{m}\cdot\mathbf{S}' - \mathbf{S}'\cdot\mathbf{m}\otimes\mathbf{m})\right.$$
$$\left. + \sqrt{\frac{B}{S}}\,[(\mathbf{n}\otimes\mathbf{m}\cdot\mathbf{S}' - \mathbf{S}'\cdot\mathbf{n}\otimes\mathbf{m}) + (\mathbf{m}\otimes\mathbf{n}\cdot\mathbf{S}' - \mathbf{S}'\cdot\mathbf{m}\otimes\mathbf{n})]\right\}$$
$$= \frac{S}{(2+S+B)}\{(2+S-2B)(\mathbf{n}\otimes\mathbf{n}\cdot\mathbf{A} - \mathbf{A}\cdot\mathbf{n}\otimes\mathbf{n}) + 3S(\mathbf{N}\otimes\mathbf{n} - \mathbf{n}\otimes\mathbf{N})$$
$$+(2-2S+B)\frac{B}{S}(\mathbf{m}\otimes\mathbf{m}\cdot\mathbf{A} - \mathbf{A}\cdot\mathbf{m}\otimes\mathbf{m}) + 3\frac{B^2}{S}(\mathbf{M}\otimes\mathbf{m} - \mathbf{m}\otimes\mathbf{M})$$
$$+ 6B(B-S)(\mathbf{n}\cdot\mathbf{A}\cdot\mathbf{m})\mathbf{n}\otimes\mathbf{m} + 6B(S-B)(\mathbf{m}\cdot\mathbf{A}\cdot\mathbf{n})\mathbf{m}\otimes\mathbf{n}$$
$$- 6B(B-S-1)(\mathbf{n}\cdot\mathbf{M})\mathbf{n}\otimes\mathbf{m} + 6B(S-B+1)(\mathbf{n}\cdot\mathbf{M})\mathbf{m}\otimes\mathbf{n}\}, \qquad (4.27)$$

where we used the identities (2.3), and the equilibrium condition (4.19). Thus the stress tensor becomes:

$$\sigma = \frac{ck_BT}{2\bar{D}_r}\left\{\frac{2}{3}(1-S-B)\mathbf{A} + S\left(1+\frac{2+S-2B}{2+S+B}\right)\mathbf{n}\otimes\mathbf{n}\cdot\mathbf{A}\right.$$
$$+ \frac{3BS}{2+S+B}\mathbf{A}\cdot\mathbf{n}\otimes\mathbf{n} + B\left(1+\frac{2-2S+B}{2+S+B}\right)\mathbf{m}\otimes\mathbf{m}\cdot\mathbf{A}$$
$$+ B\left(1-\frac{2-2S+B}{2+S+B}\right)\mathbf{A}\cdot\mathbf{m}\otimes\mathbf{m} - S\left(1-\frac{3S}{2+S+B}\right)\mathbf{N}\otimes\mathbf{n}$$
$$- S\left(1+\frac{3S}{2+S+B}\right)\mathbf{n}\otimes\mathbf{N} - B\left(1-\frac{3B}{2+S+B}\right)\mathbf{M}\otimes\mathbf{m}$$
$$- B\left(1+\frac{3B}{2+S+B}\right)\mathbf{m}\otimes\mathbf{M} - 2S^2(\mathbf{A}:\mathbf{n}\otimes\mathbf{n})\mathbf{n}\otimes\mathbf{n}$$
$$- 2B^2(\mathbf{A}:\mathbf{m}\otimes\mathbf{m})\mathbf{m}\otimes\mathbf{m} - 2BS\left[1-\frac{3(B-S)}{2+S+B}\right)(\mathbf{A}:\mathbf{n}\otimes\mathbf{m})\mathbf{n}\otimes\mathbf{m}$$
$$- 2BS\left[1+\frac{3(B-S)}{2+S+B}\right](\mathbf{A}:\mathbf{m}\otimes\mathbf{n})\mathbf{m}\otimes\mathbf{n}$$
$$+ 2BS\left[1+\frac{3(S-B+1)}{2+S+B}\right](\mathbf{n}\cdot\mathbf{M})\mathbf{n}\otimes\mathbf{m}$$
$$\left. - 2BS\left[1-\frac{3(S-B+1)}{2+S+B}\right](\mathbf{n}\cdot\mathbf{M})\mathbf{m}\otimes\mathbf{n}\right\}. \qquad (4.28)$$

Comparing (4.26) to (2.29), the Chauré coefficients are (to within the common factor $ck_BT/2\bar{D}_r$):

$$\nu_1 = \frac{2}{3}(1 - S - B), \qquad \nu_2 = -2S^2, \qquad \nu_3 = -2B^2,$$

$$\nu_4 = -2BS\left[1 - \frac{3(B - S)}{2 + S + B}\right], \qquad \nu_5 = -2BS\left[1 + \frac{3(B - S)}{2 + S + B}\right],$$

$$\nu_6 = \frac{3BS}{2 + S + B}, \qquad \nu_7 = S\left[1 + \frac{2 + S - 2B}{2 + S + B}\right],$$

$$\nu_8 = B\left[1 - \frac{2 - 2S + B}{2 + S + B}\right], \qquad \nu_9 = B\left[1 + \frac{2 - 2S + B}{2 + S + B}\right],$$

$$\nu_{10} = -2BS\left[1 + \frac{3(S - B + 1)}{2 + S + B}\right], \qquad \nu_{11} = 2BS\left[1 - \frac{3(S - B + 1)}{2 + S + B}\right],$$

$$\nu_{12} = -S\left[1 + \frac{3S}{2 + S + B}\right], \qquad \nu_{13} = -S\left[1 - \frac{3S}{2 + S + B}\right],$$

$$\nu_{14} = -B\left[1 + \frac{3B}{2 + S + B}\right], \text{ and } \nu_{15} = -B\left[1 - \frac{3B}{2 + S + B}\right]. \tag{4.29}$$

With the biaxial order parameter $B = 0$, these viscosity coefficients reduce to those calculated by Marrucci.[9] However, these forms do not obey the Onsager relations indicated in (2.31). It is not clear yet whether the use of a single diffusion coefficient is too crude an approximation for the Onsager relations to appear. It may also be that the Onsager relations are not relevant in the molecular theory for biaxial nematics, although they seem to be indicated in the uniaxial molecular theory.[9]

5. CONCLUSIONS

In the molecular theory, the molecule is considered, dynamically, as a rod about its body diagonal. This permits the use of a single moment of inertia (or diffusion coefficient) in the rheological equations. Admittedly, the molecular theory is rather crude, as is probably reflected in the failure of the Onsager relations to appear. However, a fuller description involves the use of three moments of inertia: I_1, I_2, and I_3 which form the inertial tensor in (2.1). Currently, we are working on a variant of this molecular approach which allows the use of all three moments of inertia (or diffusion coefficients) and which shows definite promise of the appearance of the Onsager relations. Results will be published later.

In agreement with Ericksen's results[8] on uniaxial nematics, the agreement between the viscosity coefficients calculated from the continuum and molecular approaches, tends to be better, the smaller the values of the order parameters, S and B. There are some disconcerting features of this agreement as detailed below; 1) ν_2 (molecular) does not have any B in its expression, while ν_2 (continuum) does; and 2) ν_6 (molecular) becomes zero when $B = 0$, which agrees with Marrucci's results.[9] But ν_6 (continuum) does not. Overall, however, the agreement seems satisfactory.

Acknowledgment

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APPENDIX A

Some useful identities are considered here, and the derivation of (2.32) is given in some more detail:

$$\mathbf{An} = (\mathbf{n}\cdot\mathbf{An})\mathbf{n} + (\mathbf{n}\cdot\mathbf{Am})\mathbf{m} + (\mathbf{l}\cdot\mathbf{An})\mathbf{l}, \tag{A.1}$$

and

$$\mathbf{n}\cdot\mathbf{A}^2\mathbf{n} = |\mathbf{An}|^2 = (\mathbf{n}\cdot\mathbf{An})^2 + (\mathbf{n}\cdot\mathbf{Am})^2 + (\mathbf{n}\cdot\mathbf{Al})^2. \tag{A.2}$$

Similarly,

$$\mathbf{m}\cdot\mathbf{A}^2\mathbf{m} = |\mathbf{Am}|^2 = (\mathbf{m}\cdot\mathbf{Am})^2 + (\mathbf{n}\cdot\mathbf{Am})^2 + (\mathbf{m}\cdot\mathbf{Al})^2, \tag{A.3}$$

and

$$\mathbf{l}\cdot\mathbf{A}^2\mathbf{l} = |\mathbf{Al}|^2 = (\mathbf{l}\cdot\mathbf{Al})^2 + (\mathbf{m}\cdot\mathbf{Al})^2 + (\mathbf{n}\cdot\mathbf{Al})^2. \tag{A.4}$$

Now, by using (2.16), (A.2), (A.3) and (A.4), obtain:

$$\begin{aligned}\mathrm{tr}\,\mathbf{A}^2 &= \mathbf{n}\cdot\mathbf{A}^2\mathbf{n} + \mathbf{m}\cdot\mathbf{A}^2\mathbf{m} + \mathbf{l}\cdot\mathbf{A}^2\mathbf{l} \\ &= \mathbf{n}\cdot\mathbf{A}^2\mathbf{n} + \mathbf{m}\cdot\mathbf{A}^2\mathbf{m} + (\mathbf{l}\cdot\mathbf{Al})^2 + (\mathbf{n}\cdot\mathbf{Al})^2 + (\mathbf{m}\cdot\mathbf{Al})^2 \\ &= 2\mathbf{n}\cdot\mathbf{A}^2\mathbf{n} + 2\mathbf{m}\cdot\mathbf{A}^2\mathbf{m} + 2(\mathbf{n}\cdot\mathbf{An})(\mathbf{m}\cdot\mathbf{Am}) - 2(\mathbf{m}\cdot\mathbf{An})^2 \\ &\quad + (\mathrm{tr}\,\mathbf{A})^2 - 2\,(\mathrm{tr}\,\mathbf{A})(\mathbf{m}\cdot\mathbf{Am}) - 2\,(\mathrm{tr}\,\mathbf{A})(\mathbf{n}\cdot\mathbf{An}).\end{aligned} \tag{A.5}$$

For the incompressible case, $\mathrm{tr}\,\mathbf{A} = 0$, and the last three terms of (A.5) are absent.

The first step towards obtaining (2.32) is:

$$D = \chi_1 \operatorname{tr} \mathbf{A}^2 + \chi_{11}\left(\mathbf{N} + \frac{\chi_7}{2\chi_{11}}\mathbf{An}\right)^2 + \chi_3(\mathbf{m}\cdot\mathbf{Am})^2$$

$$+ \chi_{12}\left(\mathbf{M} + \frac{\chi_8}{2\chi_{12}}\mathbf{Am}\right)^2 + \chi_{10}\left(\mathbf{m}\cdot\mathbf{N} + \frac{\chi_9}{2\chi_{10}}(\mathbf{n}\cdot\mathbf{Am})\right)^2$$

$$+ \chi_2(\mathbf{n}\cdot\mathbf{An})^2 + \left(\chi_6 - \frac{\chi_9^2}{4\chi_{10}}\right)(\mathbf{n}\cdot\mathbf{Am})^2 + \left(\chi_4 - \frac{\chi_7^2}{4\chi_{11}}\right)\mathbf{n}\cdot\mathbf{A}^2\mathbf{n}$$

$$+ \left(\chi_5 - \frac{\chi_8^2}{4\chi_{12}}\right)\mathbf{m}\cdot\mathbf{A}^2\mathbf{m}. \qquad \text{(A.6)}$$

Use of (A.1)–(A.4) leads to (2.32).

APPENDIX B

Some useful results are considered here in connection with the evaluation of invariants given in (3.3):

$$\hat{\mathbf{Q}} = \dot{\mathbf{Q}} - \Omega\mathbf{Q} + \mathbf{Q}\Omega$$

$$= a(\mathbf{n}\otimes\dot{\mathbf{n}} + \dot{\mathbf{n}}\otimes\mathbf{n}) + b(\mathbf{m}\otimes\dot{\mathbf{m}} + \dot{\mathbf{m}}\otimes\mathbf{m}) - a\Omega\cdot\mathbf{n}\otimes\mathbf{n}$$

$$- a\mathbf{n}\otimes\Omega\cdot\mathbf{n} - b\Omega\cdot\mathbf{m}\otimes\mathbf{m} - b\mathbf{m}\otimes\Omega\cdot\mathbf{m}$$

$$= a(\mathbf{n}\otimes\mathbf{N} + \mathbf{N}\otimes\mathbf{n}) + b(\mathbf{m}\otimes\mathbf{M} + \mathbf{M}\otimes\mathbf{m}). \qquad \text{(B.1)}$$

APPENDIX C

The rotational energy for the linear rotor is:

$$E_J = \frac{\hbar^2}{8\pi^2 I} J(J+1) \qquad \text{(C.1)}$$

where I is the moment of inertia (For the linear rotor, only two moments of inertia are significant, and they are equal).

The rotational Hamiltonian corresponding to the energy in (C.1) is:

$$H = \frac{\hbar^2}{2\pi I}\nabla_{\mathbf{u}}^2 \qquad \text{(C.2)}$$

where

$$\nabla_u^2 \equiv \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\phi^2} \tag{C.3}$$

where (θ, ϕ) are the angles the rigid rotor makes with the laboratory axes.

For the symmetric top molecule, the energy is:[25]

$$E_{J,K} = \frac{1}{I_1}J(J+1) + \left(\frac{1}{I_3} - \frac{1}{I_1}\right)K^2 \tag{C.4}$$

where I_1 is the unique moment of inertia and $I_1 = I_2$. If $I_3 > I_1 = I_2$, the symmetrical top is oblate. An example is a circular disk. If $I_3 < I_1 = I_2$, the body is prolate. An example is a shuttle cock. K is a quantum number which ranges between $-J$ and J.

The Hamiltonian is:

$$H = \frac{\hbar^2}{2\pi I_1}\nabla_u^2 \tag{C.5}$$

where

$$\nabla_u^2 \equiv \frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\phi^2} - 2\cot\theta\frac{\partial^2}{\partial\theta\partial\chi} + \cot^2\theta\frac{\partial^2}{\partial\chi^2} + \left(\frac{I_1}{I_3} - 1\right)\frac{\partial^2}{\partial\chi^2} \tag{C.6}$$

and θ, ϕ, and χ are the Euler angles as defined in Reference 27.

The biaxial molecule (example: a planar rectangular plate) does not possess a 3-fold or more axis of symmetry. This means that one does not have closed-form (analytic) expressions as in the linear and symmetric top cases. This kind of asymmetric rotor is treated as a perturbation to the symmetric top. Following Wittmer,[26] the quantum mechanical energy expression is an infinite series:

$$E_{J,K} = \frac{1}{2}\left(\frac{1}{I_1} + \frac{1}{I_2}\right)J(J+1) + \left[\frac{1}{I_3} - \frac{1}{2}\left(\frac{1}{I_1} + \frac{1}{I_2}\right)\right]K^2 + f(J,K)\frac{\left(\frac{1}{I_1} - \frac{1}{I_2}\right)^2}{\frac{1}{I_3} - \frac{1}{2}\left(\frac{1}{I_1} + \frac{1}{I_2}\right)} + \dots \tag{C.7}$$

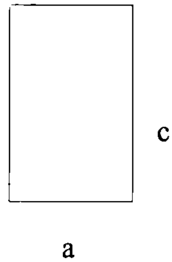

FIGURE 2 Variant of the model shown in Figure 1. Here $a \neq c \neq b \neq 0$. Such an object also forms the biaxial nematic phase.

where $f(J, K)$ is a function of J and K only (explicit form for $f(J, K)$ is given in Reference 23). For $I_1 = I_2$, we recover the symmetric top as in (C.3). However, for a planar plate (see Figure 2).

$$I_3 = I_1 - I_2 \tag{C.8}$$

and

$$I_3 << I_1, I_2 \tag{C.9}$$

Therefore the first perturbation term is $\approx I_3^3/I_1^2 I_2^2$, which is small. However, in our calculation, we have used only one moment of inertia (i.e., $I_1 = I_2 \neq I_3$ with $K = 0$). Moments of inertia are simply related to diffusion coefficients and the number of moments of inertia is equal to the number of diffusion coefficients.[28] This means that we use the same equations as Doi but introduce biaxiality through the order tensors only.

Transformation properties of the 3-D rotational group are given below, as it is useful in both Sections 2 and 4:

$$\begin{pmatrix} \mathbf{l} \\ \mathbf{m} \\ \mathbf{n} \end{pmatrix} = \begin{pmatrix} \cos\phi\cos\theta\cos\chi - \sin\phi\sin\chi & \sin\phi\cos\theta\cos\chi + \cos\phi\sin\chi & -\sin\theta\cos\chi \\ -\cos\phi\cos\theta\sin\chi - \sin\phi\cos\chi & \sin\phi\cos\theta\sin\chi + \cos\phi\cos\chi & \sin\theta\sin\chi \\ \cos\phi\sin\theta & \sin\phi\sin\theta & \cos\theta \end{pmatrix} \begin{pmatrix} \mathbf{e}_x \\ \mathbf{e}_y \\ \mathbf{e}_z \end{pmatrix} \tag{C.10}$$

where $\mathbf{e}_x$, $\mathbf{e}_y$, and $\mathbf{e}_z$ are unit vectors along the laboratory axis x, y, and z.

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