Stress tensor for a nematic liquid crystal

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STRESS TENSOR FOR A NEMATIC LIQUID CRYSTAL

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Résumé. — Des équations constitutives pour les cristaux liquides nématiques ont été établies par Ericksen et Leslie. Dans ces équations apparaît un tenseur des contraintes faisant intervenir six coefficients de viscosité α_i ($i = 1...6$). On montre ici que l'application des relations d'Onsager conduit à la relation

$$\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5 .$$

Il ne subsiste donc que cinq coefficients de viscosité indépendants.

Abstract. — Constitutive equations for nematic liquid crystals were first established by Ericksen and Leslie. They used a stress tensor with six viscosity coefficients α_i ($i = 1...6$). It is shown in this paper that the Onsager reciprocal relations lead to the relation

$$\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5 .$$

Hence there are only five independent viscosity coefficients for a nematic liquid crystal.

I. **Introduction.** — Ericksen [1] and Leslie [2] [3] have written constitutive equations for anisotropic fluids. For incompressible isothermal nematic liquid crystals, the dissipative part of the stress tensor takes the form [1]

$$\overline{\overline{\sigma}} = \alpha_1(\mathbf{nn} : \overline{\overline{A}})\,\mathbf{nn} + \alpha_2\,\mathbf{nN} + \alpha_3\,\mathbf{Nn} + \alpha_4\,\overline{\overline{A}} + \alpha_5\,\mathbf{nn}.\overline{\overline{A}} + \alpha_6\,\overline{\overline{A}}.\mathbf{nn} \tag{1.1}$$

where $\mathbf{n}$ is the director ($\mathbf{n}^2 = 1$), $\overline{\overline{A}}$ the strain rate tensor :

$$A_{ij} = \frac{1}{2}\left(\frac{\partial v_j}{\partial x_i} + \frac{\partial v_i}{\partial x_j}\right) \tag{1.2}$$

and $\mathbf{N}$ the velocity of the director relative to the fluid :

$$\mathbf{N} = \dot{\mathbf{n}} - \boldsymbol{\omega} \times \mathbf{n} = (\boldsymbol{\Omega} - \boldsymbol{\omega}) \times \mathbf{n} \tag{1.3}$$

where $\boldsymbol{\Omega}$ and $\boldsymbol{\omega}$ are the angular velocities of the director and of the fluid :

$$\boldsymbol{\omega} = \tfrac{1}{2}\,\mathrm{curl}\,\mathbf{v} . \tag{1.4}$$

The torque exerted by the director on the fluid is

$$\boldsymbol{\Gamma}_n = \mathbf{n} \times (\gamma_1\,\mathbf{N} + \gamma_2\,\overline{\overline{A}}.\mathbf{n}) \tag{1.5}$$

where γ_1 and γ_2 are given by

$$\gamma_1 = \alpha_3 - \alpha_2 \,; \quad \gamma_2 = \alpha_6 - \alpha_5 . \tag{1.6}$$

When deriving these equations, Ericksen and Leslie have taken into account the spatial symmetry properties of the medium, and the equality of action and reaction. It is shown in this paper that the Onsager reciprocal relations, that reflect the time-reversal invariance of the equations of motion of the individual particles, lead to the relation

$$\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5 . \tag{1.7}$$

2. **Thermodynamic fluxes and forces.** — In order to use the Onsager [4] reciprocal relations, one has to express the entropy production as a product of thermodynamic fluxes and forces [5] [6]. The entropy production is given by

$$T\dot{S} = \overline{\overline{\sigma}} : \mathbf{grad}\,\mathbf{v} + \boldsymbol{\Gamma}_n.\boldsymbol{\Omega} . \tag{2.1}$$

Let $\overline{\overline{\Omega}}$ be the antisymmetric tensor

$$(\overline{\overline{\Omega}})_{\alpha\beta} = -(\overline{\overline{\Omega}})_{\beta\alpha} = (\boldsymbol{\Omega})_\gamma \tag{2.2}$$

where (α, β, γ) is a cyclic permutation, and $\overline{\overline{\Gamma}}_n$ the tensor

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(1) The dyadic $\mathbf{ab}$ has components $(\mathbf{ab})_{\alpha\beta} = a_\alpha\, b_\beta$.
The products $\overline{\overline{A}}.\overline{\overline{B}}$ and $\overline{\overline{A}} : \overline{\overline{B}}$ are defined by

$$(\overline{\overline{A}}.\overline{\overline{B}})_{\alpha\beta} = \sum_\gamma A_{\alpha\gamma}\, B_{\gamma\beta} \,; \qquad \overline{\overline{A}} : \overline{\overline{B}} = \sum_{\alpha,\beta} A_{\alpha\beta}\, B_{\alpha\beta} .$$

The vectors $\overline{\overline{A}}.\mathbf{b}$ and $\mathbf{b}.\overline{\overline{A}}$ have components

$$(\overline{\overline{A}}.\mathbf{b})_\alpha = \sum_\beta A_{\alpha\beta}\, b_\beta \,; \qquad (\mathbf{b}.\overline{\overline{A}})_\alpha = \sum_\beta b_\beta\, A_{\beta\alpha} .$$

Hence $ab : \overline{\overline{A}} = a.\overline{\overline{A}}.b = \sum_{\alpha\beta} A_\alpha\, b_\beta\, A_{\alpha\beta}$ is a scalar.

The dyadics $\mathbf{ab}.A$ and $A.\mathbf{ab}$ have components

$$(\mathbf{ab}.\overline{\overline{A}})_{\alpha\beta} = \sum_\gamma a_\gamma\, b_\gamma\, A_{\gamma\beta} \,; \qquad (\overline{\overline{A}}.\mathbf{ab})_{\alpha\beta} = \sum_\gamma A_{\alpha_\beta}\; a_\alpha\; b_\beta .$$

$$\overline{\overline{\Gamma}}_n = \frac{\gamma_1}{2}(\mathbf{nN} - \mathbf{Nn}) + \frac{\gamma_2}{2}(\mathbf{nn}.\overline{\overline{A}} - \overline{\overline{A}}.\mathbf{nn}) . \quad (2.3)$$

Using eqs. (1.1) and (1.5), one easily shows that the antisymmetric part of $\overline{\overline{\sigma}}$ is $-\overline{\overline{\Gamma}}_n$, and that

$$(\overline{\overline{\Gamma}}_n)_{\alpha\beta} = -(\overline{\overline{\Gamma}}_n)_{\beta\alpha} = \tfrac{1}{2}(\mathbf{\Gamma}_n)_\gamma . \quad (2.4)$$

Let $\overline{\overline{\Pi}}$ be the symmetric part of the stress tensor $\overline{\overline{\sigma}}$,

$$\overline{\overline{\Pi}} = \alpha_1(\mathbf{nn} : \overline{\overline{A}})\,\mathbf{nn} + \frac{\alpha_2 + \alpha_3}{2}(\mathbf{nN} + \mathbf{Nn}) + \alpha_4 \overline{\overline{A}} + \frac{\alpha_5 + \alpha_6}{2}(\mathbf{nn}.\overline{\overline{A}} + \overline{\overline{A}}.\mathbf{nn}) . \quad (2.5)$$

The **grad v** tensor can be split into a symmetric part $\overline{\overline{A}}$, and an antisymmetric part $\overline{\overline{\omega}}$,

$$(\overline{\overline{\omega}})_{\alpha\beta} = \frac{1}{2}\left(\frac{\partial v_\beta}{\partial x_\alpha} - \frac{\partial v_\alpha}{\partial x_\beta}\right) = -(\overline{\overline{\omega}})_{\beta\alpha} = (\boldsymbol{\omega})_\gamma . \quad (2.6)$$

Eq. (2.1) can be now written

$$T\dot{S} = \overline{\overline{\Pi}} : \overline{\overline{A}} + \overline{\overline{\Gamma}}_n : (\overline{\overline{\Omega}} - \overline{\overline{\omega}}) . \quad (2.7)$$

In eq. (2.7), two independent thermodynamic forces appear, $\overline{\overline{A}}$ and $(\overline{\overline{\Omega}} - \overline{\overline{\omega}})$. The conjugated fluxes are $\overline{\overline{\Pi}}$ and $\overline{\overline{\Gamma}}_n$. These are the symmetric and antisymmetric viscous parts of the momentum density flux. We have now to express these fluxes as linear forms on the forces.

$$\mathbf{N} = (\mathbf{\Omega} - \boldsymbol{\omega}) \times \mathbf{n} = \mathbf{n}.(\overline{\overline{\Omega}} - \overline{\overline{\omega}}) = -(\overline{\overline{\Omega}} - \overline{\overline{\omega}}).\mathbf{n} . \quad (2.8)$$

Eqs (2.3) and (2.5) can be written

$$\overline{\overline{\Pi}} = \alpha_1(\mathbf{n}.\overline{\overline{A}}.\mathbf{n})\,\mathbf{nn} + \left(\frac{\alpha_5 + \alpha_6}{2}\right)(\mathbf{nn}.\overline{\overline{A}} + \overline{\overline{A}}.\mathbf{nn}) + \alpha_4 \overline{\overline{A}} + \frac{\alpha_2 + \alpha_3}{2}\{\mathbf{nn}.(\overline{\overline{\Omega}} - \overline{\overline{\omega}}) - (\overline{\overline{\Omega}} - \overline{\overline{\omega}}).\mathbf{nn}\} \quad (2.9)$$

$$\overline{\overline{\Gamma}}_n = \frac{\gamma_2}{2}(\mathbf{nn}.\overline{\overline{A}} - \overline{\overline{A}}.\mathbf{nn}) + \frac{\gamma_1}{2}\{\mathbf{nn}.(\overline{\overline{\Omega}} - \overline{\overline{\omega}}) + (\overline{\overline{\Omega}} - \overline{\overline{\omega}}).\mathbf{nn}\} . \quad (2.10)$$

3. **Onsager reciprocal relations.** — Eqs (2.9) and (2.10) are linear relations between fluxes $\overline{\overline{J}}^i$ and forces $\overline{\overline{X}}^i$:

$$J^i_{\alpha\beta} = \sum_{\gamma\delta j} L^{ij}_{\alpha\beta,\gamma\delta} X^j_{\gamma\delta} . \quad (3.1)$$

Onsager reciprocal relations state that

$$L^{ij}_{\alpha\beta,\gamma\delta} = L^{ji}_{\alpha\beta,\gamma\delta} . \quad (3.2)$$

Let $\overline{\overline{\Pi}}$, $\overline{\overline{\Gamma}}_n$, $\overline{\overline{A}}$ and $(\overline{\overline{\Omega}} - \overline{\overline{\omega}})$ be $\overline{\overline{J}}^1$, $\overline{\overline{J}}^2$, $\overline{\overline{X}}^1$, $\overline{\overline{X}}^2$.

The matrices L^{12} and L^{21} have elements

$$L^{12}_{\alpha\beta,\gamma\delta} = \frac{\alpha_2 + \alpha_3}{2}(n_\alpha n_\gamma \delta_{\beta\delta} - \delta_{\alpha\gamma}\, n_\beta\, n_\delta)$$
$$L^{21}_{\alpha\beta,\gamma\delta} = \frac{\gamma_2}{2}(n_\alpha\, n_\gamma\, \delta_{\beta\delta} - \delta_{\alpha\gamma}\, n_\beta\, n_\delta) . \quad (3.3)$$

Relations (3.2) implies that

$$\alpha_2 + \alpha_3 = \gamma_2 = \alpha_6 - \alpha_5 . \quad (3.4)$$

There are now five independent viscosity coefficients, α_1, α_4, γ_1, γ_2 and $\beta = \alpha_5 + \alpha_6$. The symmetric part of the stress tensor is

$$\overline{\overline{\Pi}} = \alpha_1(\mathbf{n}.\overline{\overline{A}}.\mathbf{n})\,\mathbf{nn} + \alpha_4 \overline{\overline{A}} + \frac{\beta}{2}(\mathbf{nn}.\overline{\overline{A}} + \overline{\overline{A}}.\mathbf{nn}) + \frac{\gamma_2}{2}(\mathbf{nN} + \mathbf{Nn}) . \quad (3.5)$$

With these notations, the stress tensor is

$$\overline{\overline{\sigma}} = \alpha_1(\mathbf{n}.\overline{\overline{A}}.\mathbf{n})\,\mathbf{nn} + \frac{\gamma_2 - \gamma_1}{2}\mathbf{nN} + \frac{\gamma_2 + \gamma_1}{2}\mathbf{Nn} + \alpha_4 \overline{\overline{A}} + \frac{\beta - \gamma_2}{2}\mathbf{nn}.\overline{\overline{A}} + \frac{\beta + \gamma_2}{2}\overline{\overline{A}}.\mathbf{nn} .$$

And the entropy production (2.7) can be written as

$$T\dot{S} = \alpha_4 \overline{\overline{A}}^2 + \beta(\mathbf{n}.\overline{\overline{A}})^2 + \alpha_1(\mathbf{n}.\overline{\overline{A}}.\mathbf{n})^2 + 2\gamma_2\, \mathbf{n}.\overline{\overline{A}}.\mathbf{N} + \gamma_1 \mathbf{N}^2 . \quad (3.6)$$

Let us take local axes with $\mathbf{n}$ along the z-axis. Eq. (3.6) is now

$$T\dot{S} = \sum_{i,j} \alpha_4 A_{ij}^2 + \sum_i \{(2\alpha_4 + \beta)A_{zi}^2 + 2\gamma_2 A_{zi} N_i + \gamma_1 N_i^2\} + (\alpha_1 + \beta + \alpha_4)A_{zz}^2 \qquad (i, j = x, y) . \quad (3.7)$$

This expression must be positive definite. This implies that

$$\alpha_4 > 0 ; \quad \gamma_1 > 0 ; \quad 2\alpha_4 + \beta > 0$$
$$\alpha_1 + \beta + \alpha_4 > 0 ; \quad \gamma_1(2\alpha_4 + \beta) - \gamma_2^2 > 0 . \quad (3.8)$$

4. **Some applications.** — *a*) STRESS EXERTED BY A ROTATION OF THE DIRECTOR. — Let us first consider the fluid at rest, and let us rotate $\mathbf{n}$ with angular velocity $\mathbf{\Omega}$. Take $\mathbf{n}$ along Z-axis and $\mathbf{\Omega}$ along X-axis (Fig. 1). Using (3.5) and (1.5), one finds

$$\Gamma_{nX} = \gamma_1\, \Omega$$

$$\Pi_{YZ} = \Pi_{ZY} = -\frac{\gamma_2}{2}\Omega .$$

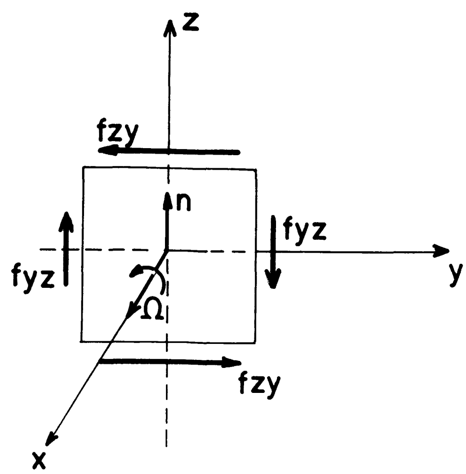

FIG. 1. — The fluid is at rest, and the director **n** spins with angular velocity **Ω** (**Ω** ⊥ **n**). f_{zy}, f_{yz} are the surface forces exerted by an elementary volume including **n** on the external fluid

$(\gamma_2 < -\gamma_1)$.

The dissipative stress tensor, $\bar{\bar{\sigma}}$, has non-zero components

$$\sigma_{YZ} = -\left(\frac{\gamma_1 + \gamma_2}{2}\right)\Omega$$

$$\sigma_{ZY} = \frac{\gamma_1 - \gamma_2}{2}\Omega .$$

The forces acting on the surface of an elementary volume in the fluid are shown on figure 1 :

$$f_{YZ} = -\sigma_{YZ} = \frac{\gamma_1 + \gamma_2}{2}\Omega$$
$$f_{ZY} = -\sigma_{ZY} = -\frac{\gamma_1 - \gamma_2}{2}\Omega . \qquad (4.1)$$

b) SHEAR FLOW. — α) Let us now assume **n** at rest ($\dot{\mathbf{n}} = 0$), and a shear flow in *YZ*-plane parallel to **n** (Fig. 2*a*) :

$$\frac{\partial v_Z}{\partial Y} = -W .$$

Using eqs (3.7) and (1.5), one finds for the torque per unit volume exerted by the flow on the nematic molecule $\mathbf{\Gamma}_f$ ($\mathbf{\Gamma}_f = -\mathbf{\Gamma}_n$) :

$$\Gamma'_{fX} = -\frac{\gamma_1 + \gamma_2}{2} W$$
$$\Gamma'_{fY} = \Gamma'_{fZ} = 0 . \qquad (4.2a)$$

β) Let us now assume a shear flow perpendicular to **n**, and parallel to the *Z*-axis (Fig. 2*b*) :

$$\frac{\partial v_Y}{\partial Z} = -W .$$

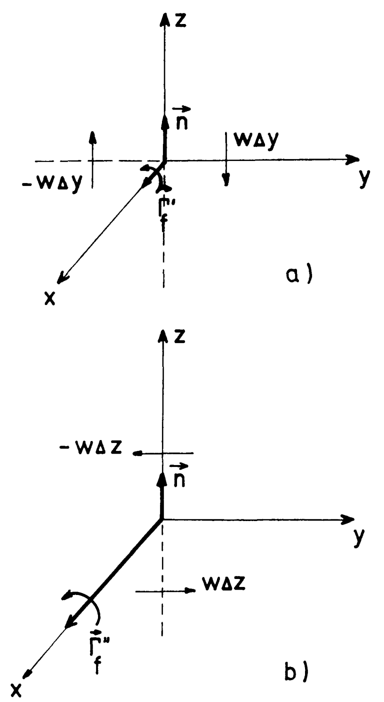

FIG. 2. — The director **n** is at rest in a shear flow in the *yz* plane (*a*) shear flow parallel to **n** ; *b*) shear flow perpendicular to **n**). Γ_f and Γ''_f are the torques exerted by the fluid on the director

$(\gamma_2 < -\gamma_1)$.

Eq. (3.7) give

$$\Gamma''_{fX} = -\frac{\gamma_1 - \gamma_2}{3}(-W)$$
$$\Gamma''_{fY} = \Gamma''_{fZ} = 0 . \qquad (4.2b)$$

The coefficients are the same in eqs (4.1) and (4.2). This results from eq. (1.7).

In case α) and β), the two flows have opposite angular velocities

$$\omega''_x = -\omega'_x = \frac{W}{2}, \quad \omega_Y = \omega_Z = 0 .$$

The difference in the torques Γ'_{fX} and Γ''_{fX} is due to the anisotropic shape of the nematic molecule. This effect is very well known for macroscopic bodies. In this case, $\Gamma''_{fX} \gg \Gamma'_{fX}$, which implies that γ_2 is negative, and $|\gamma_1 - \gamma_2| \ll \gamma_1$.

Helfrich [7] has derived this last result for nematic molecules from a model in which the molecules are assumed to be equally and rigidly oriented ellipsoids of revolution, colliding with each other like the molecules of a gas. He finds $\gamma_2 < -\gamma_1$. This result implies that, in case α), the torque exerted on the nematic molecule is negative.

Helfrich's model is not very convincing : it neglects the correlations between molecules and the exchange of angular momentum. Moreover, in his model, the sign of $(\gamma_1 + \gamma_2)$ depends strongly on the shape of the molecule (2).

The behaviour of a nematic liquid crystal in a shear flow depends strongly on the sign of $(\gamma_1 + \gamma_2)$. Leslie [3] has shown that, if $(\gamma_1 + \gamma_2) < 0$, the nematic molecules are oriented in a shear flow, and that the angle θ, between the flow and the director is given by

$$\cos 2\,\theta = -\frac{\gamma_1}{\gamma_2}.$$

If $(\gamma_1 + \gamma_2) > 0$, there is no such orientation.

The experiments of Marinin and Tsvetkov [8] seem to confirm the orientation of *p*-azoxyanisole in a rectangular capillary. It then seems reasonable to think that $\gamma_2 < -\gamma_1$.

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(2) *Note added in proof.* In a more recent paper, to be published in the Journal of Chemical Physics, Helfrich uses a new model, with a two-body ellipsoidal interaction potential. From this model, which is more convincing, he derives the same result $(\gamma_1 + \gamma_2) > 0$.