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Parodi's Relation as a Stability Condition for Nematics

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It is shown that the requirement that the basic uniformly-oriented state of equilibrium of a nematic liquid crystal be stable to small disturbances leads to Parodi's relation between the viscosity coefficients, provided the material aligns in shear flow. This relation was first derived from Onsager reciprocal relations.

INTRODUCTION

In the continuum theory for nematic liquid crystals proposed by Leslie[1], six viscosity coefficients $\mu_1 \ldots \mu_6$ appear. As shown by Leslie, the viscous dissipation is positive if and only if these viscosity coefficients satisfy the following inequalities:

$$\mu_4 > 0,\ 2\mu_1 + 3\mu_4 + 2\mu_5 + 2\mu_6 \geqslant 0$$
$$2\mu_4 + \mu_5 + \mu_6 \geqslant 0,\ \mu_3 - \mu_2 \geqslant 0 \tag{1}$$
$$4(\mu_3 - \mu_2)(2\mu_4 + \mu_5 + \mu_6) \geqslant (\mu_2 + \mu_3 + \mu_6 - \mu_5)^2.$$

Since no further restrictions are placed on these coefficients, there would appear to be six independent viscosities, constrained by (1).

However, on the basis of the Onsager reciprocal relations, Parodi[2] has proposed the condition

$$\mu_2 + \mu_3 + \mu_5 - \mu_6 = 0 \tag{2}$$

This relation reduces the number of independent viscosity coefficients to five.

Parodi's relation has been adopted by the Orsay Liquid Crystal Group[3] when analysing their experiments on wave propagation in nematic liquid crystals. Thus the values for the viscosity coefficients derived from these experiments depend on the assumption that (2) is valid. However, the fundamental thermodynamic basis for the Onsager reciprocal relations has been severely criticised recently, in particular in an article by Truesdell[4]. After a thorough examination, Truesdell concludes that the principles and details of Onsagerist thermodynamics are at present unsatisfactory but suggests that it is possible that for particular materials 'counterparts of Onsager relations can be found and given a specific meaning, ... and that meaning will express on the phenomenological scale a *special symmetry* or *stability*'.

The purpose of this note is to confirm this interpretation of the Onsager relations when the material is a nematic liquid crystal. A possible experimental test of Parodi's relation has already been proposed[5]. Here we show that Parodi's relation is a consequence of the requirement that the basic uniformly-oriented state of equilibrium of a nematic liquid crystal be *stable* to small disturbances, provided the material adopts a constant alignment in uniform shear flow.

STABILITY ANALYSIS

The governing equations of the continuum theory for nematic liquid crystals[1] are linearized about a basic state in which the material is at rest and the orientation is everywhere parallel to the unit vector **d**. We let **u** be the perturbation to the director specifying the orientation in the material and **v** be the velocity vector, and seek plane wave solutions of the linearized equations of the form

$$\begin{aligned} \mathbf{u} &= \mathbf{a} \exp\, i\,(m\nu.\,\mathrm{x} - \omega t)\,, \\ \mathbf{v} &= \mathbf{b} \exp\, i\,(m\nu.\,\mathrm{x} - \omega t)\,. \end{aligned} \tag{3}$$

where ω is the real frequency, m the complex wave number and ν a unit vector specifying the direction of propagation of the wave.

As expected from the symmetry of the physical situation, there are two modes: the in-plane mode for which **u** and **v** lie in the plane defined by the basic orientation **d** and the direction of propagation ν; and the out-of-plane mode in which both **u** and **v** are normal to this plane. For the in-plane mode, we find from the linearized equations that[5, 6, 7]

$$\begin{aligned} \mathbf{a} \;=\; & A\{\nu - \mathbf{d}\}(\mathrm{v.\,d})\ ,\ \mathrm{b} = B\{\mathbf{d} - \nu\,(\mathbf{d}\,\nu)\}, \\ & 2\omega\, mp\,(\theta)\,A + m^2\{g\,(\theta) - 2i\rho\,\omega\,B\} = 0, \\ & 2\{m^2 f(\theta) + \lambda_1\, \mathrm{i}\omega\}A - imq\,(\theta)\,B = 0. \end{aligned} \tag{4}$$

Here ρ is the density and θ is defined by $\sin\theta = \nu.\mathbf{d}$. The functions g, p, q, f and the constant λ_1 are given in terms of the free energy coefficients α_1, α_3 and the viscosities $\mu_1 \ldots \mu_6$ by

$$g(\theta) = 2\mu_1 \sin^2\theta \cos^2\theta + (\mu_3 + \mu_6)\cos^2\theta + (\mu_5 - \mu_2)\sin^2\theta + \mu_4,$$
$$p(\theta) = \mu_2 \sin^2\theta - \mu_3 \cos^2\theta,\ \lambda_1 = \mu_2 - \mu_3, \tag{5}$$
$$q(\theta) = (\mu_2 - \mu_3 + \mu_5 - \mu_6)\cos^2\theta + (\mu_2 - \mu_3 - \mu_5 + \mu_6)\sin^2\theta,$$
$$f(\theta) = \alpha_1 \cos^2\theta + \alpha_3 \sin^2\theta.$$

It is clear from (4) that these modes are diffusive. They were considered first in detail by Martinoty and Candau[7]. Similar equations to (4) and (5) hold for the out-of-plane mode, but these have no significance in the present discussion.

Consider the angles θ_0 and θ_1 defined by

$$\mathrm{q}(\theta_0) = 0,\ 0 \leqslant \theta_0 \leqslant \pi/2, \qquad \mathrm{p}(\theta_1) = 0,\ 0 \leqslant \theta_1 \leqslant \pi/2. \tag{6}$$

It follows from (5) and the inequalities (1) that θ_0 is real unless

$$\mu_2 - \mu_3 < \mu_5 - \mu_6 < \mu_3 - \mu_2, \tag{7}$$

and that θ_1 is real unless

$$\mu_2 < 0 < \mu_3. \tag{8}$$

If θ_1 is real, then along the cone of directions ν for which $\nu.\mathbf{d} = \sin\theta_1$ a purely orientational mode is possible, without any associated velocity disturbance. For (4) is satisfied with $B = 0$, $\mathrm{m}^2 = -\lambda_1 i\omega/f(\theta_1)$. Similarly if θ_0 is real, there is a cone of directions along which purely acoustic modes are possible, with no orientational disturbance, Eqs. (4) being satisfied with $A = 0$, $\mathrm{m}^2 = 2i\rho\omega/\mathrm{g}(\theta_0)$.

The existence of these special directions along which pure modes can propagate has the following consequence, pointed out to me by F. M. Leslie[8]. Along a direction v for which θ is close to θ_1, a small velocity disturbance can produce a large disturbance in the orientation. For as $\theta \to \theta_1$ in (4), the ratio A/B becomes either zero or infinite depending on the choice of m, provided that $q(\theta_1) \neq 0$. Similarly, along directions with θ close to θ_0, a small orientational disturbance can produce a large velocity disturbance, if $p(\theta_0) \neq 0$.

We can take this observation by Leslie a stage further and conclude that if either θ_0 is real and $p(\theta_0) \neq 0$ or θ_1 is real and $q(\theta_1) \neq 0$, the basic uniformly oriented equilibrium state is *unstable* in the sense that small disturbances in the orientation or velocity can give rise to unbounded disturbances in the velocity or orientation respectively. This instability is avoided only if $\theta_1 = \theta_0$. In this case the two sets of special directions coincide and along these directions Eqs. (4) are uncoupled. From (5) and (6) it follows that $\theta_0 = \theta_1$ only if Parodi's relation (2) holds, provided that

$$\mu_2 \neq \mu_3. \tag{9}$$

Thus we have shown that if the viscosity coefficients satisfy (9) and not more than one of (7) and (8), the uniformly oriented state is unstable unless Parodi's relation (2) holds. It is found experimentally that nematics usually adopt a constant orientation in uniform shear flow, with exceptional materials reported only by Gähwiller[9]. The analysis given by Leslie[1] shows that for a material that does align in shear flow the viscosities must satisfy

$$|\mu_5 - \mu_6| \geqslant |\mu_2 - \mu_3| \tag{10}$$

Moreover, unless the alignment is perpendicular to the flow, $\mu_2 \neq \mu_3$. For these materials we conclude that (9) is satisfied and (7) cannot hold. Thus Parodi's relation must hold to avoid instability.

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