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On a Material Coefficient in Cholesteric Liquid Crystals

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Abstract—We propose and analyze an experiment which may determine the existence of a material coefficient in the continuum theory of cholesteric liquid crystals.

For static, isothermal states of an incompressible cholesteric liquid crystal we may reduce the local expressions of the balance of moment of momentum and momentum in the continuum theory proposed on Ericksen[1] and Leslie[2] to

$$\left(\frac{\partial F}{\partial d_{i,k}}\right)_k - \frac{\partial F}{\partial d_i} + \gamma d_i = 0 \tag{1}$$

and $p = p_0 - F$ respectively. Here the director $d_i(x_k)$ is a unit vector field describing the orientation of the molecular axis, $F = W + \Theta$ is the density of total energy, $W(d_i, d_{i,k})$ is the Helmholtz free energy density, $\Theta(d_i, x_i)$ is the potential energy density of external influences, γ and the pressure p are arbitrary scalar fields associated with the constraints of d_i to fixed magnitude and incompressibility respectively, and p_0 is an arbitrary constant.

In the absence of surface actions, there are associated with these field equations the boundary conditions

$$L_i = \epsilon_{ijk} d_j \left(\frac{\partial W}{\partial d_{k,l}} + \alpha \epsilon_{klp} d_p\right) n_l \tag{2}$$

and

$$T_i = \left[-p\delta_{ij} - \frac{\partial W}{\partial d_{k,j}} d_{k,i} + \alpha \epsilon_{jkp} (d_p d_i)_{,k}\right] n_j, \tag{3}$$

where L_i and T_i are, respectively, the couple and the traction applied to the surface element with unit outward normal n_i. The coefficient α is introduced in a constitutive relation for the entropy flux in the general theory[2] and remains as a constant in this static, isothermal specialization. The physical significance of this coefficient is poorly understood and the necessity of retaining it in the theory has not been established. In this note we indicate the rather dramatic effect that its presence may have upon the director orientation in a particularly simple experimental configuration.

For this purpose, we utilize the form of the Helmholtz free energy density proposed by Frank,[3]

$$2W = \alpha_1(d_{i,i})^2 + \alpha_2(\tau + d_i\epsilon_{ijk}d_{k,j})^2 + \alpha_3 d_i d_j d_{k,i} d_{k,j} + \\ + (\alpha_2+\alpha_4)[d_{i,j}d_{j,i} - (d_{i,i})^2], \tag{4}$$

where, in isothermal states, the coefficients are constants. Leslie's[4] arguments lead to the restrictions $\alpha_1 > 0$, $\alpha_2 > 0$, $\alpha_3 > 0$. We assume that the external influences are an applied magnetic field H_i and gravity. In this case[1] $\Theta = \chi - \frac{1}{2}[\nu(H_k d_k)^2 + \mu H_k H_k]$, where χ is the gravitational potential and ν and μ are positive constants.

We consider a sample of a cholesteric liquid crystal bounded below by a plane solid boundary and above by an isotropic liquid. We assume that this free surface is plane, horizontal and parallel to the solid surface and that a magnetic field of constant magnitude H is applied normal to these planes. We choose a coordinate system in which the free and solid surfaces are located at $z = l$ and $z = 0$ respectively. With this simple geometry and applied field, it seems reasonable to examine director configurations of the form

$$d_x = \cos\theta\cos\phi, \quad d_y = \cos\theta\sin\phi, \quad d_z = \sin\theta, \tag{5}$$

where $\theta = \theta(z)$ and $\phi = \phi(z)$. In static situations, the balance of moment of momentum requires that these functions be continuously differentiable. For these energy densities and fields, we obtain from Eq. (1), after some manipulation to eliminate the scalar γ, the two equations

$$2f\theta'' + f_{,\theta}(\theta')^2 - g_{,\theta}(\phi')^2 + (\nu H^2 - 2\alpha_2\tau\phi')\sin 2\theta = 0, \tag{6}$$

$$g\phi'' + g_{,\theta}\theta'\phi' + \alpha_2\tau\sin 2\theta\theta' = 0, \tag{7}$$

where a prime denotes differentiation with respect to z, $f = f(\theta) \equiv (\alpha_1\cos^2\theta + \alpha_2\sin^2\theta)$ and $g = g(\theta) \equiv (\alpha_2\cos^2\theta + \alpha_3\sin^2\theta)\cos^2\theta$. We may immediately integrate (7) to obtain

$$g\phi' - \alpha_2\tau\cos^2\theta = k. \tag{8}$$

The integration constant k may be evaluated at either boundary.

At the free surface, it is natural to assume that the isotropic liquid exerts a normal pressure upon but applies no couple to the liquid crystal. In this case, we may reduce the couple conditions (2) to the requirements that

$$\theta' = 0 \quad \text{and} \quad g\phi' + (\alpha - \alpha_2\tau)\cos^2\theta = 0 \tag{9}$$

at $z = l$. The traction condition (3) is satisfied if the pressure in the isotropic fluid is uniform at the free surface.

The determination of the physically appropriate boundary condition at the solid surface is more difficult. We might plausibly assume that the nature or the prior treatment of the solid surface encourages the director to align parallel to the surface and that the normal component of the applied couple vanishes. In this event, we have, from (5) and the relevant component of (2),

$$\theta = 0 \quad \text{and} \quad \alpha_2\phi' + (\alpha - \alpha_2\tau) = 0 \tag{10}$$

at $z = 0$. Calculations based upon these boundary conditions have been carried out by Leslie[2,4,5] and appear to be consistent with observations. Adopting the boundary conditions (9) and (10), we proceed to determine the integration constant k in (8).

Evaluating (8) at $z = l$ and using the boundary condition (9), we obtain $k = -\alpha\cos^2\theta(l)$. However, evaluating (8) at $z = 0$ and using the boundary condition (10), we find $k = -\alpha$. If α is nonzero, the two determinations are compatible only if $\theta(l) = 0$. Now with the smoothness assumptions already made, it is a relatively easy matter to show that the only solution of (6) and (8) with $\theta = 0$ and $\theta' = 0$ at $z = l$ is

$$\theta \equiv 0, \quad \phi = \left(\tau - \frac{\alpha}{\alpha_2}\right)z + \phi_0 . \tag{11}$$

On the other hand, if α is zero, the boundary conditions (8) and (9) require that k be zero. In this case, Leslie[5] shows that if the magnitude of the applied magnetic field is below a critical value $H_C = \alpha_1(\tau/2l) + \alpha_3\tau^2$, the uniform twist (11) with α zero is, again, the only possible solution. However, if the magnitude of the applied field exceeds H_C, there exists, in addition, a distorted configuration which, if $\alpha_2 > \alpha_3$, involves less total energy than the uniform twist. Because, in the absence of applied forces, a local twist is a characteristic of director configurations observed in cholesteric liquid crystals, we expect that this inequality is satisfied. Consequently, if α is zero, we anticipate that as the magnitude of the applied field exceeds H_C the uniform twist will be deformed. If, however, α is nonzero, the only possible configuration of the form (5) which satisfies our interpretations (9) and (10) of the boundary condition (2) is the uniform twist (11) no matter how large the magnitude at the applied field. Thus, our analysis suggests an experiment to determine the presence of the coefficient α.

Surface effects at a plane solid boundary have been considered by Rapini and Papoular[6] and, at a free surface, by Jenkins and Barratt.[7] Incorporating their generalizations of the boundary conditions (2) and (3) into the analysis does not alter our results for nonzero α, but modifies Leslie's critical field, deformed solution, and energy arguments. Also Jenkins[8] and Nehring and Saupe[9] have proposed more general forms for the free energy density for cholesteric liquid crystals which we do not consider here.

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