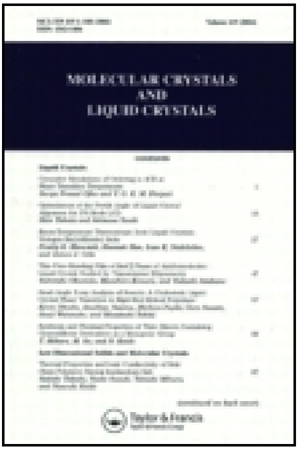

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Hydrodynamics of Biaxial Nematic Liquid Crystals

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Hydrodynamics of Biaxial Nematic Liquid Crystals

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The linearized hydrodynamic equations and the elastic part of the free energy density for a biaxial nematic liquid crystal are derived. The equations are then used to predict the attenuation of low frequency sound waves.

I INTRODUCTION

Recently, the existence of a biaxial nematic liquid crystal was experimentally verified by Yu and Saupe[1] in potassium laurate-1-decanol water mixtures. This is the first time such a phase has been observed in nature.

From the theoretical point of view, it has been predicted by mean field type calculations[2–4] that molecules which differ appreciably from a "rod-like" shape by being "rectangular" or "ellipsoidal" in shape can exhibit this biaxial nematic phase as an expression of additional orientational order. While the uniaxial nematic phase is characterized by long range orientational order in one of the spatial directions,[5] the biaxial nematic phase is characterized by long range orientational order in two (hence all three) of the spatial directions. A biaxial nematic has rotational symmetry broken around three distinct axes. The discovery of such a phase provides the motivation for our hydrodynamic study.

In Section II we discuss the broken symmetry and its relation to the hydrodynamic variables, then derive the hydrodynamic equations appropriate for this phase. In Section III we derive the attenuation of low frequency sound waves in terms of the dissipative parameters introduced in Section II. Finally in Section IV we summarize our results and compare them with the independent work of Brand and Pleiner.[6]

II HYDRODYNAMICS

The hydrodynamic variables associated with a biaxial nematic liquid crystal are those due to microscopic conservation laws and those arising from any continuous symmetries broken in the phase.[7] The densities of the conserved quantities are the mass density ρ, the momentum density g_i, and the energy density ϵ. In the fixed laboratory frame these satisfy the conservation equations

$$\dot{\rho} + \nabla_i g_i = 0, \tag{1}$$

$$\dot{g}_i + \nabla_j \sigma_{ij} = 0, \tag{2}$$

$$\dot{\epsilon} + \nabla_i j^{\epsilon}_i = 0, \tag{3}$$

where σ_{ij} is the symmetric stress tensor† and j^{ϵ}_i is the energy current density. The cartesian index i runs from 1 to 3 and a summation convention for repeated indices is used.

Variables corresponding to the continuous broken symmetries can be deduced by considering fluctuations of the symmetric, traceless, local order parameter for liquid crystals. The order parameter is proportional to the quadrupolar term in the mass density[9]

$$R_{ij} = \sum_{\alpha k} m^{\alpha k} \left[(r^{\alpha k} - r^{\alpha})_i (r^{\alpha k} - r^{\alpha})_j - \tfrac{1}{3} \delta_{ij} (\mathbf{r}^{\alpha k} - \mathbf{r}^{\alpha})^2\right] \delta(\mathbf{r} - \mathbf{r}^{\alpha}), \tag{4}$$

where $r_i^{\alpha k}$ and $m^{\alpha k}$ are the coordinate and mass of the k^{th} particle in the α^{th} molecule, and r^{α}_i is the location of the molecular center of mass. Assuming biaxial symmetry and averaging over molecular orientations we find

$$\langle R_{ij}(\mathbf{r})\rangle = Q_{ij}(\mathbf{r}) = S(\mathbf{r})[n_i(\mathbf{r})\, n_j(\mathbf{r}) - \tfrac{1}{3}\delta_{ij}] + \xi(\mathbf{r})[m_i(\mathbf{r})\, m_j(\mathbf{r}) - l_i(\mathbf{r})\, l_j(\mathbf{r})], \tag{5}$$

where $S(\mathbf{r})$ is a measure of the unixiality and $\xi(\mathbf{r})$ is a measure of the biaxiality in $Q_{ij}(\mathbf{r})$.‡ The "uniaxial director" $n_i(\mathbf{r})$ and the "biaxial director" $m_i(\mathbf{r})$ are orthogonal unit vectors defining the local symmetry axes and $l_i = \epsilon_{ijk} m_j n_k$ where ϵ_{ijk} is the totally antisymmetric tensor. For the purposes of this paper we define n_i^o, the equilibrium direction of n_i, to be in the direction of the 3 axis and m_i^o to be in the direction of the 2 axis.

In the uniaxial nematic, long-wavelength fluctuations in two components of the order parameter, δQ_{13} and δQ_{23}, have been shown to exhibit hydrodynamic behavior.[10] Subjecting the biaxial order parameter (5) to an identical analysis yields three low-frequency hydrodynamic variables for the biaxial

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†It is always possible to chose a stress tensor such that $\sigma_{ij} = \sigma_{ji}$.[8]

‡ $n_i Q_{ij} n_j = (\tfrac{2}{3})S(\mathbf{r})$; $m_i Q_{ij} m_j = \xi(\mathbf{r}) - (\tfrac{1}{3})S(\mathbf{r})$.

nematic phase: δQ_{13}, δQ_{23}, and a new variable δQ_{12}, which is associated with the additional broken rotational symmetry about the 3 axis. These can be rigorously identified with fluctuations in the directors $n_i(\mathbf{r})$ and $m_i(\mathbf{r})$:

$$\delta n_1 = \delta Q_{13}/(S+\xi),\ \delta n_2 = \delta Q_{23}/(S-\xi),\ \delta m_1 = \delta Q_{12}/2\xi. \tag{6}$$

The equations of motion for the symmetry breaking variables can be written

$$\dot{n}_i + \nabla_j X_{ij}^{(n)} = 0, \tag{7}$$

$$\dot{m}_i + \nabla_j X_{ij}^{(m)} = 0, \tag{8}$$

where $X_{ij}^{(n)}$ and $X_{ij}^{(m)}$ are the director currents. The two equations for the directors contain only three independent components due to the orthonormality condition. In our convention; $\dot{n}_3 = 0, \dot{m}_2 = 0$, and $\dot{m}_3 = -\dot{n}_2$. The explict form of the linearized hydrodynamic equations has been calculated using the microscopic approach of Forster *et al.*,[10] and Martin, Parodi, and Pershan.[8]

The existence of an equilibrium state with order parameter Q_{ij} implies we can construct a thermodynamic identity, valid in the laboratory frame, of the form

$$Td(\rho s) = d\epsilon - \mu d\rho - v_i dg_i - \phi_{ij}^{(n)}\, d(\nabla_i n_j) - \phi_{ij}^{(m)}\, d(\nabla_i m_j), \tag{9}$$

where T is the temperature, s is the entropy density, μ is the chemical potential, v_i is the velocity of the rest frame with respect to the lab frame, and the last term expresses the fact that spatial variations of $n_i(\mathbf{r})$ and $m_i(\mathbf{r})$ must relax with a frequency that goes to zero as the wave number of the disturbance goes to zero. The variables conjugate to $\nabla_i n_j$ and $\nabla_i m_j$ contain the elastic contributions to the free energy and can be written

$$\phi_{ij}^{(n)} = K_{ijkr}^{(n)}\, \nabla_r n_k + K_{ijkr}^{(n,m)}\, \nabla_r m_k, \tag{10}$$

$$\phi_{ij}^{(m)} = K_{ijkr}^{(m)}\, \nabla_r m_k + K_{ijkr}^{(n,m)}\, \nabla_r n_k, \tag{11}$$

where $K_{ijkr}^{(n)}$, $K_{ijkr}^{(m)}$, and $K_{ijkr}^{(n,m)}$ are the Frank elastic constants for the biaxial nematic phase.† Biaxial symmetry dictates that there be twelve invariants in the elastic part of the free energy density. They can be summarized in the following expressions,

$$\begin{aligned} K_{ijkr}^{(n)} = {} & (K_1 l_j^o l_r^0 + K_2 m_j^o m_r^o + K_3 n_j^o n_r^o)\, l_i^o l_k^o \\ & + (K_4 l_j^o l_r^{o0} + K_5 m_j^o m_r^o + K_6 n_j^o n_r^o)\, m_i^o m_k^o + K_{10} l_i^o l_j^o m_k^o m_r^o \end{aligned} \tag{12}$$

$$K_{ijkr}^{(m)} = (K_7 l_j^o l_r^{o0} + K_8 m_j^o m_r^o + K_9 n_j^o n_r^o)\, l_i^o l_k^o \tag{13}$$

$$K_{ijkr}^{(n,m)} = K_{11} l_i^o m_j^o l_k^o n_r^o + K_{12} m_i^o l_j^o l_k^o n_r^o \tag{14}$$

†See the appendix for a derivation of the elastic constants.

The reactive terms in hydrodynamic equations are those relating currents to densities possessing the same time-reversal symmetry. Using Galilean invariance and the isotropy of the liquid pressure, p, the linearized reactive parts of the mass current and the energy current are

$$(g_i)^R = \rho v_i \tag{15}$$

and,

$$(j_i^\epsilon)^R = (\epsilon + p)v_i, \tag{16}$$

respectively. Symmetry considerations similarly indicate that the reactive part of the director current is coupled to the momentum density.

Evaluation of the Poisson bracket yields[11]

$$\begin{aligned}(X_{ij}^{(n)})^R = &-(\tfrac{1}{2})[(\lambda_1+1)\, n_j^o l_r^o + (\lambda_1 - 1)\, n_r^o l_j^o]\, v_r l_i^o \\ &-(\tfrac{1}{2})[(\lambda_2+1)\, n_j^o m_r^o + (\lambda_2 - 1)\, m_j^o n_r^o]\, v_r m_i^o,\end{aligned} \tag{17}$$

$$(X_{ij}^{(m)})^R = -(\tfrac{1}{2})[(\lambda_3+1)\, m_j^o l_r^o + (\lambda_3 - 1)\, l_j^o m_r^o]\, v_r l_i^o \tag{18}$$

where λ_1, λ_2, and λ_3 are reactive parameters analogous to the λ introduced in Forster *et al.*[10] The microscopic result above is in accord with the requirement that $\dot{n}_i + \epsilon_{ijk} n_j^o \omega_k = 0$ and $\dot{m}_i + \epsilon_{ijk} m_j^o \omega_k = 0$, where $\omega_i = (\tfrac{1}{2})\epsilon_{ijk}\partial_j v_k$, for solid body rotation. The reactive part of the stress tensor can be calculated via the thermodynamic identities

$$-\frac{\partial \dot{n}_i}{\partial v_k} = \frac{\partial \dot{g}_k}{\partial[\nabla_j \phi_{ij}^{(n)}]} \tag{19}$$

$$-\frac{\partial \dot{m}_i}{\partial v_k} = \frac{\partial \dot{g}_k}{\partial[\nabla_j \phi_{ij}^{(m)}]} \tag{20}$$

which follow from (9) in the absence of dissipation. It can be written

$$\begin{aligned}(\sigma_{ij})^R = &(\tfrac{1}{2}) n_i^o \partial_k \phi_{rk}^{(n)}\, (\lambda_1 l_j^o l_r^o + \lambda_2 m_j^o m_r^o) \\ &+ (\tfrac{1}{2}) \partial_k \phi_{rk}^{(m)}\, (\lambda_3 m_i^o l_j^o l_r^o + \lambda_2 n_i^o m_j^o n_r^o) \\ &+ (\tfrac{1}{2}) l_r^o \partial_k \phi_{rk}^{(n)} (n_k^o l_i^o - n_i^o l_k^o) \\ &+ (\tfrac{1}{2}) m_r^o \partial_k \phi_{rj}^{(n)} (n_k^o m_i^o - n_i^o m_k^o) \\ &+ (\tfrac{1}{2}) l_r^o \partial_k \phi_{rj}^{(m)} (m_k^o l_i^o - m_i^o l_k^o) \\ &+ (\tfrac{1}{2}) n_r^o \partial_k \phi_{rj}^{(m)} (n_k^o m_i^o - n_i^o m_k^o) \\ &+ [ij \to ji] + \delta_{ij} p.\end{aligned} \tag{21}$$

Irreversible effects enter the equations of motion through the dissipative parts of the currents. They are

$$(g_i)^D = 0, \tag{22}$$

$$(j_i^{\epsilon})^D = -\kappa_{11} l_i^o l_j^o \partial_j(\delta T) - \kappa_{22} m_i^o m_j^o \partial_j(\delta T) - \kappa_{33} n_i^o n_j^o \partial_j(\delta T), \tag{23}$$

$$(X_{ij}^{(n)})^D = \Gamma_1 l_i^o l_k^o \phi_{kj}^{(n)} + \Gamma_2 m_i^o m_k^o \phi_{kj}^{(n)}, \tag{24}$$

$$(X_{ij}^{(m)})^D = \Gamma_3 l_i^o l_k^o \phi_{kj}^{(m)} \tag{25}$$

$$\begin{aligned}(\sigma_{ij})^D = &-l_i^o l_j^o (l_k^o l_r^o \eta_1 + m_k^o m_r^o \eta_2 + n_k^o n_r^o \eta_3) A_{kr} \\ &- m_i^o m_j^o (l_k^o l_r^o \eta_2 + m_k^o m_r^o \eta_4 + n_k^o n_r^o \eta_5) A_{kr} \\ &- n_i^o n_j^o (l_k^o l_r^o \eta_3 + m_k^o m_r^o \eta_5 + n_k^o n_r^o \eta_6) A_{kr} \\ &- 2(l_i^o m_j^o + l_j^o m_i^o) l_k^o m_r^o \eta_7 A_{kr} \\ &- 2(l_i^o n_j^o + l_j^o n_i^o) l_k^o n_r^o \eta_8 A_{kr} \\ &- 2(m_i^o n_j^o + m_j^o n_i^o) m_k^o n_r^o \eta_9 A_{kr},\end{aligned} \tag{26}$$

where $A_{kr} = (\frac{1}{2})(\partial_k v_r + \partial_r v_k)$. The invariants of the dissipative part of the stress tensor given above are equivalent to the viscosities of a biaxial crystal.[12] This completes our derivation of the hydrodynamics, a summary and discussion are given in Section IV.

III ACOUSTIC ATTENUATION

The attenuation of the longitudinal part of the propagating sound mode has been calculated in terms of the dissipative parameters derived in Section II. The attenuation constant is found to depend on the angle ϕ between the propagation direction and the "uniaxial director" n_i, and the angle θ between the propagation direction and the "biaxial director" m_i in the following way:

$$\begin{aligned}\alpha = \frac{\omega^2}{c^3 \rho_o} &[\eta_1 \sin^2\theta + \eta_4 \cos^2\theta + (\eta_6 - \eta_1)\cos^2\phi \\ &+ 2(\eta_1 - \eta_2 - 2\eta_7 - \eta_3 - 2\eta_8 + \eta_5 + 2\eta_4)\cos^2\theta \cos^2\theta \\ &+ (2\eta_3 - \eta_1 - \eta_6 + 4\eta_8)\cos^2\phi \sin^2\phi \\ &+ (2\eta_2 - \eta_1 - \eta_4 + 4\eta_7)\cos^2\theta \sin^2\theta] \\ &+ \left(\frac{1}{c_v} - \frac{1}{c_p}\right)[\kappa_{11}\sin^2\theta + \kappa_{22}\cos^2\theta + (\kappa_{33} - \kappa_{11})\cos^2\phi],\end{aligned} \tag{27}$$

where the η's are the viscosities introduced in Section II, c is the speed of sound, ρ_o is the equilibrium mass density, and c_v and c_p are the constant volume and pressure specific heats.

IV CONCLUSION

We have shown that rotational symmetry breaking results in three independent hydrodynamic variables for a biaxial nematic liquid crystal. The linearized hydrodynamic equations were then derived using the microscopic order parameter for this biaxial phase. Our results are in agreement with a preprint of work done by Brand and Pleiner[6] on the nonlinear aspects of biaxial liquid crystals. While they report 15 different invariants in the free energy density for a biaxial nematic liquid crystal, in the linear limit, three of their invariants can be eliminated via surface terms due to the commutativity of infinitesimal rotations. The twelve remaining invariants are equivalent to the set we report.

The attenuation of low frequency sound waves was then calculated in terms of the dissipative parameters of our hydrodynamic equations. It was found that the attenuation constant depends on both the angle ϕ between the direction of propagation and the uniaxial director n_i, and the angle θ between the direction of propagation and the biaxial director m_i.

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References

1. L. J. Yu and A. Saupe, *Phys. Rev. Lett.*, **45**, 1000 (1980).
2. M. J. Freiser, *Phys. Rev. Lett.*, **24**, 1081 (1970).
3. J. P. Straley, *Phys. Rev. A*, **10**, 1881 (1974).
4. R. Alben, *J. Chem. Phys.*, **59**, 4299 (1973).
5. P. G. de Gennes, *The Physics of Liquid Crystals* (Oxford University Press, London, 1974), p. 23.
6. H. Brand and H. Pleiner, submitted for publication.
7. D. Forster, *Hydrodynamic Fluctuations, Broken Symmetries and Correlation Functions* (Benjamin, Reading, 1975), p. 255.
8. P. C. Martin, O. Parodi and P. S. Pershan, *Phys. Rev. A*, **6**, 2401 (1972).
9. T. C. Lubensky, *Phys. Rev. A*, **2**, 2497 (1970).
10. D. Forster, et al., *Phys. Rev. Lett.*, **26**, 1016 (1971).
11. D. Forster, *Phys. Rev. Lett.*, **32**, 1161 (1974).
12. L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Pergamon Press, Oxford, 1970), p. 37.

APPENDIX

The elastic part of the free energy density can, in general, be written as a quadratic function of the spatial derivatives of the order parameter:[10]

$$F_{el} = \tfrac{1}{2} \int d^3x \; E_{ijkrst} \nabla_i Q_{jk} \nabla_r Q_{st}. \tag{28}$$

General symmetry considerations imply that E_{ijkrst} be invariant under the following operations

1) $j \leftrightarrow k$ or $s \leftrightarrow t$,

2) $ijk \leftrightarrow rst$,

3) $i \leftrightarrow r$.

Biaxiality imposes the additional restriction that the free energy density be invariant under

4) $x \leftrightarrow -x$,

5) $y \leftrightarrow -y$,

6) $z \leftrightarrow -z$.

The resulting twelve invariants can been written in terms of the spatial derivatives of $n_i(\mathbf{r})$ and $m_i(\mathbf{r})$;

$$F_{el} = \tfrac{1}{2}\int d^3x \, \{K^{(n)}_{ijkr} \nabla_j n_i \nabla_r n_k + K^{(m)}_{ijkr} \nabla_j m_i \nabla_r m_k + K^{(n,m)}_{ijkr} (\nabla_j n_i \nabla_r m_k + \nabla_j m_i \nabla_r n_k)\}. \quad (29)$$