Hydrodynamics of cholesteric liquid crystals in the coarse-grained limit

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Abstract. The hydrodynamical behaviour of cholesteric liquid crystals has been considered in the limit of low amplitude and low frequency distortions and motions. It is shown that there are interesting analogies with superfluid-hydrodynamics, such as the fountain effect, thermal superconductivity and temperature wave propagation. In certain situations, there is an unusual formation of a boundary layer at low velocities, and in certain others the properties resemble those of percolation in porous media. Results concerning some special phenomena peculiar to cholesteric liquid crystals are also presented. Finally it is pointed out that there should be two types of second sound in chiral smectic C.

Keywords. Cholesteric liquid crystals; thermomechanical effect; superfluid hydrodynamics; boundary layer phenomena; percolation in porous media; thermal convection; chiral smectic C.

1. Introduction

The dynamical description of liquid crystals includes not only the fluid particle motions but also the time variations of the parameters describing the ordered state. Leslie (1969) has proposed a continuum theory of cholesteric liquid crystals by extending the hydrodynamics of nematics as first developed by Ericksen (1961) and Leslie (1968). Here the fluid velocity \mathbf{v} and the director \mathbf{n} are the primary variables.

The hydrodynamics of cholesterics has two interesting regimes. In one limit the velocities and the director variations are of high amplitude and frequency. This situation is, in general, very complicated and often one has to resort to numerical techniques for solving the problem. In the other limit, velocities and director distortions vary slowly and smoothly over many pitches. This is often referred to as the coarse-grained or the supercontinuum limit. Leslie's theory gets considerably simplified in this limit, and in many ways the governing equations resemble those for smectic A; in other words, the medium behaves as though it is layered. Lubensky (1972) has also developed an hydrodynamical theory of cholesterics from general thermodynamic considerations. In this paper we employ Leslie's theory.

2. Theory

With each fluid particle, as in normal fluid dynamics, we associate a velocity field v. In addition, in order to incorporate the order prevailing in the medium, we have at each point a dimensionless unit vector n called the director. Many of the interesting and distinctive flow phenomena exhibited by liquid crystals are due to an intimate coupling between v and n. The governing equations are obtained from the conservation laws.

Mass conservation leads to the familiar continuity equation:

$$\frac{1}{\rho}\frac{\mathrm{d}\rho}{\mathrm{d}t} + v_{i,i} = 0,\tag{1}$$

where

$$v_{i,i} = \frac{\partial v_i}{\partial x_i} \qquad i = 1, 2, 3.$$

Conservation of linear momentum gives

$$\rho \frac{\mathrm{d}v_i}{\mathrm{d}t} = F_i - P_{,i} - \left(\frac{\partial W}{\partial n_{k,i}} n_{k,i}\right)_{,i} + t'_{ji,j},\tag{2}$$

Here F_i = external body force per unit volume, P = pressure, W = elastic energy density and t'_{ij} = hydrodynamic stress.

Conservation of angular momentum yields

$$\sigma \frac{\mathrm{d}n_i}{\mathrm{d}t} = G_i + \gamma n_i - \frac{\partial W}{\partial n_i} + \left(\frac{\partial W}{\partial n_{i,j}}\right)_{i,j} + g_i',\tag{3}$$

with σ = the moment of inertia of the director, G_i = the external director body force, γ = an arbitrary scalar and g'_i = the hydrodynamic director body force.

The explicit expressions for W, t'_{il} and g'_i are given by

$$W = \frac{1}{2} [K_{11}(\nabla \cdot \mathbf{n})^2 + K_{22}(\mathbf{n} \cdot \nabla \times \mathbf{n} + q_0)^2 + K_{33}(\mathbf{n} \times \nabla \times \mathbf{n})^2], \tag{4}$$

$$t'_{ji} = \alpha_1 n_k n_p A_{kp} n_i n_j + \alpha_2 N_i n_j + \alpha_3 N_j n_i + \alpha_4 A_{ij} + \alpha_5 A_{ik} n_k n_j + \alpha_6 A_{ik} n_k n_i + \alpha_7 e_{ipq} n_p T_{e} n_j + \alpha_8 e_{ipq} n_p T_{e} n_i,$$
 (5)

$$g_i' = \lambda_1 N_i + \lambda_2 A_{ik} n_k + \lambda_3 e_{ijk} T_k n_i, \tag{6}$$

with

$$\lambda_1 = \alpha_2 - \alpha_3;$$
 $\lambda_2 = \alpha_5 - \alpha_6;$
 $\lambda_3 = \alpha_7 - \alpha_8;$
 $A_{ij} = \frac{1}{2}(v_{i,j} + v_{j,i});$
 $M_{ij} = \frac{1}{2}(v_{i,j} - v_{j,i});$
 $M_i = dn_i/dt - \omega_{ik}n_k;$
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Finally the conservation of energy is described by

$$\dot{Q} = -q_{i,i} + t'_{ii}A_{ii} - g'_{i}N_{i},\tag{7}$$

with

$$q_i = K_1 T_{,i} + K_2 n_j T_{,j} n_i + K_3 e_{ijk} n_j N_k + K_4 e_{ijk} n_j A_{kp} n_p.$$
 (8)

 K_1 and K_2 are coefficients of thermal conductivity. K_3 and K_4 , like α_7 and α_8 , represent thermomechanical effects, all of which change sign under a change from right-handed to left-handed axes.

Equations (1) to (8) describe completely the mechanics of cholesteric liquid crystals, and were first proposed by Leslie. In his theory another material constant α enters the

dynamical behaviour, but since its existence is still unsettled we ignore it throughout the present discussions.

3. The coarse-grained approximation

In any given problem we have to solve the above equations, and, as stated in the introduction, we invariably end up with numerical solutions to the governing differential equations. However, in certain situations the equations are greatly simplified. Suppose that the variables that describe static distortions and dynamical features are of small amplitude, and that they vary very slowly and smoothly over many pitches. Under these circumstances we can ignore the director inertia term and average out all the high frequency components. Let us also assume that cholesteric distortions will not tip the local director away from the plane of the local layer. Then the governing differential equations simplify to

$$\frac{1}{\rho} \frac{\partial \rho}{\partial t} + v_{i, i} = 0, \tag{9}$$

$$\rho \frac{\partial v_i}{\partial t} = -P_{,i} + \delta_{i3}g + t'_{ij,j}, \qquad (10)$$

$$\dot{u} - v_3 = (-1/\lambda_1 q_0^2)g + (-\lambda_3/\lambda_1 q_0)T_{,3}, \tag{11}$$

$$Q = K_{\parallel} T_{,33} + K_{\perp} (T_{,11} + T_{,22}) + K_3 q_0 (\dot{u} - v_3),_3$$
 (12)

$$g = K_{22} q_0^2 \frac{\partial^2 u}{\partial z^2}. \tag{13}$$

Here uq_0 represents change in the local orientation of the director in its own plane. The stresses t'_{ij} also get considerably simplified. If we impose Onsager's principle for the dissipative process, we find (Parodi 1970, Prost 1972)

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

and

$$K_3 = -\lambda_3$$
.

Equations (9) to (13) are very similar to the ones proposed by de Gennes (1969) for smectic A. Lubensky (1972) and Martin *et al* (1972), from general thermodynamical considerations, established similar equations for the dynamics of layered media. Also \dot{u} can in general arise from angular as well as linear motions of the cholesteric layers. We shall consider in the next section a few implications of this model.

4. Results

4.1 The superfluid analogy

(i) Uniform flow: If no special care is taken to anchor the director at the walls, then we may look for solutions corresponding to the extreme case of $v_3 = \dot{u}$, which means that permeation is absent. In the non-dissipative limit for a steady flow along the twist axis we get (Ranganath 1983)

$$P_{.3} = -\lambda_3 q_0 T_{.3}$$
 or $\partial P/\partial T = -\lambda_3 q_0$. (14)

This is a thermomechanical effect wherein a temperature gradient sets up a pressure gradient. For negative λ_3 it is the analogue of the London equation for superfluid He II, which explains the fountain effect (Landau and Lifshitz 1966).

Such a flow carries heat with it which can be evaluated by incorporating the small viscous terms in t'_{ij} . The average velocity v_3 in a flow between plates 'h' apart is then given by

$$\overline{v} = \left(\frac{-\lambda_3 q_0}{\eta} (h^2/12)\right) T_{.3} + \left(\frac{-h^2}{12\eta}\right) P_{.3}$$

and the heat carried is given by

$$Q = \rho \overline{v} ST = \left(\frac{-\lambda_3 q_0 \rho ST \beta}{\eta}\right) T_{.3} + \left(\frac{-\rho ST \beta}{\eta}\right) P_{.3}, \tag{15}$$

 $\beta = +h^2/12$ and in general depends on the channel geometry. We find that the medium behaves as though it has an extra thermal conductivity given by (Ranganath 1983)

$$K^{e} = -\lambda_{3} q_{0} \rho ST \beta / \eta.$$

This is the analogue of the thermal superconductivity found in superfluids (Putterman 1974). It depends not only on T but also on the geometry of the channel.

(ii) Wave propagation: We shall next consider wave propagation through cholesterics. Suppose the wave is travelling in the x-z plane

$$\begin{split} v_1 &= v_1^0 \exp[i(\mathbf{k} \cdot \mathbf{r} - wt)]. \\ v_3 &= v_3^0 \exp[i(\mathbf{k} \cdot \mathbf{r} - wt)], \\ \mathbf{k} &= (k_1, O, k_3). \end{split}$$

In the non-dissipative limit and in the absence of permeation, we find from (9) and (10)

$$v_3/v_1 = -(Ak_1^2 - w^2)/Ak_1k_3, (16)$$

and

$$w^4 - w^2 \left[A(k_1^2 + k_3^2) + (B/\rho)k_3^2 \right] + (AB/\rho)k_1^2 k_3^2 = 0$$
 (17)

Here $A = \partial P/\partial \rho$ a measure of the volume compressibility, $B = K_{22}q_0^2$. This gives us two modes with amplitudes and velocities given by

Mode 1
$$v_3/v_1 = k_3/k_1$$
, $c_1^2 \approx A$, (18)

Mode 2
$$v_3/v_1 = -k_1/k_3$$
, $c_2^2 \approx \frac{B}{\rho} \frac{k_1^2 k_3^2}{k^4}$. (19)

Mode 1 is a pure longitudinal wave with accompanying density fluctuations, which exists even in normal fluids. Mode 2, however, is quite different. It is a transverse wave associated with the layer displacements u (or, equivalently, fluctuations in \mathbf{n} or the phase of the layer), but with no density changes to a very good approximation. It is, in this sense, akin to second sound in superfluid He II. In terms of the angle θ that k makes with Z axis

$$c_2 = (B/\rho)^{1/2} \cos \theta \sin \theta$$

which vanishes along and normal to the twist axis. Historically it was in smectic A liquid crystals that such a wave was predicted to exist by de Gennes (1969). Later Lubensky (1972) made a similar prediction in the case of cholesteric liquid crystals, but he ignored the thermomechanical coupling. However, as is evident from (11), with no permeation (i.e., $\dot{u} = v_3$) the phase fluctuations are accompanied by temperature fluctuations as well. In this sense there is a closer analogy with superfluid He II (Ranganath 1983). For positive λ_3 the temperature wave is a travelling wave. The ratio of the amplitude of the temperature wave to that of the phase wave is given by

$$f = -K_{22}q_0/\lambda_3.$$

For negative λ_3 the temperature wave is non-propagating.

It may be relevant to mention here a somewhat similar situation in normal fluid dynamics. In the non-dissipative limit it is an ideal fluid. In a gravitation field, pressure necessarily varies with height resulting in an *inhomogeneity*. In such a case we find (Landau and Lifshitz 1966) a propagating *transverse wave* whose frequency is given by

$$w = (T/c_p)^{1/2} (g/\rho)(\partial \rho/\partial T)_p \sin \theta,$$

where θ is the angle between the vertical and the direction of propagation. We see that this wave cannot travel vertically (i.e. $\theta = 0$) and has a frequency which is angle dependent. Also this has temperature and density fluctuations associated with it.

4.2 Permeation effects

(1) Plug flow: Cholesteric liquid crystals exhibit enormous viscosities in capillary flow (Porter et al 1966). This may be interpreted to mean that the viscosity coefficients α_i 's are large. However, as was first suggested by Helfrich (1969; see also Kini et al 1975) in spite of small α_i 's one can have large apparent viscosities due to a permeation process.

In the absence of temperature gradients and for an incompressible liquid with a blocked cholesteric texture, i.e., u = 0 (which is obtained by firm anchoring at the boundaries) we find

$$v_{i,i} = 0, (20)$$

$$\rho \frac{\partial v_i}{\partial t} = -P_{,i} + \delta_{i3}(\lambda_1 q_0^2) v_3 + t'_{ij,j}, \tag{21}$$

 $t'_{ij,j}$ may be ignored relative to the second term. Thus we find a steady flow along the twist axis with a velocity

$$v_3 = \frac{1}{\lambda_1 q_0^2} P_{.3}. \tag{22}$$

This describes a plug flow. This flat velocity profile is incompatible with the boundary condition that v_3 should vanish at the walls. To achieve this we include t'_{ij} terms and solve (21). We give below the solution for two different situations:

(1) Flow between plates at $z = \pm h/2$

$$v_3 = \frac{P_{z}}{\lambda_1 q_0^2} \left(1 - \frac{\cosh kx}{\cosh kh/2} \right), \quad \eta_a \approx -\frac{\lambda_1 (q_0 h)^2}{12}.$$
 (23A)

(2) Plate at z = h moves relative to the plate at z = 0 with a constant velocity V

$$v_3 = V \frac{\sinh kx}{\sinh kh}, \quad \eta_a = \overline{\eta} (q_0 h), \tag{23B}$$

with $k = (-\lambda_1 q_0^2/\overline{\eta})^{1/2}$ $\overline{\eta}$ = average viscosity and the apparent viscosity η_a has been calculated for $q_0 h \gg 1$.

We notice two important features. Firstly the velocity is practically flat over most of the volume but for a thin boundary layer. Interestingly one gets exactly similar velocity profiles at high frequencies for a Newtonian viscous fluid subjected to an oscillation of pressure gradient or shear, a problem first investigated by Richardson and Tyler (1929).

Secondly the apparent viscosity η_a is very different for the two geometries. Generally $h \sim 300~\mu$ and $P \sim 1~\mu$ giving an apparent viscosity of $10^6~\overline{\eta}$ for flow between plates. Interestingly this is reduced to a value of $10^3~\overline{\eta}$ in a simple shear flow. But in either case it is enormously large compared to normal fluid viscosities. Physically this large viscosity is not due to neighbouring fluid regions moving with different velocities but due to friction between individual molecules and the blocked cholesteric textures.

Before concluding this section we point out the other important feature of permeation. Equation (22) bears a close resemblance to Darcy's equation of percolation through porous media. Here a normal fluid is forced by a pressure gradient through a porous medium like sand or soil. In this case we find (Batchelor 1981)

$$v_i = -\frac{K}{\eta} P_{,i}, \tag{24}$$

K is a constant called permeability. It is proportional to the square of the linear dimensions of the interstices for a given shape. This relationship, first obtained by Darcey in 1856, has a long history of use in soil mechanics. Many practical problems like seepage from dams, movement of groundwater near a coast due to tidal pressure variations have been solved with the help of (24).

(ii) Boundary layer formation: Let us consider a slow two-dimensional flow in the zx plane with the twist axis along z. Again ignoring temperature gradients, we find to a good approximation

$$P_{.3} = (-\lambda_1 q_0^2) v_3, \tag{25}$$

$$P_{,1} = t'_{1j,j} \approx (\gamma_3 v_{1,33} + \gamma_2 v_{1,11}), \tag{26}$$

together with

$$v_{1,1} + v_{3,3} = 0, (27)$$

 γ_3 and γ_2 are linear combinations of α_i 's. Eliminating P between (25) and (26) gives

$$v_{3,1} = (-1/\lambda_1 q_0^2) (\gamma_3 v_{1,333} + \gamma_2 v_{1,113}).$$

To a very good approximation this can be written as

$$v_{3,1} = 0. (28)$$

Thus in this limit v_3 depends only on z. If we now have an obstacle in the form of a flat plate placed normal to the twist axis, and if at large distances from the plate the flow is along the x-axis then far away from the plate $v_3 = 0$. This together with (27) means that

 $v_1 = V$, the main flow velocity, throughout. But this stituation of $v_1 = V$ and $v_3 = 0$ everywhere is incompatible with the boundary condition $v_1 = 0$ on the flat plate. The fluid arriving in the vicinity of the plate must decelerate, *i.e.* P is a function of x. This localized pressure causes an upward fluid flow along the twist axis. This secondary flow is dominated by permeation as shown in the previous example. Thus near the plate we have a region dominated by permeation. The thickness δ of this permeation layer can easily be worked out and turns out to be

$$\delta \sim (x/q_0)^{1/2}$$
 or $\frac{\delta}{x} \sim (xq_0)^{-1/2}$. (29)

We find the permeation layer to be thin. Beyond this region, we have throughout $v_1 = V$ and $v_3 = 0$.

This phenomenon, first noticed by de Gennes (1974), is reminiscent of a boundary layer formation in classical hydrodynamics. But there are some important differences. While boundary layer formation usually takes place at very high main velocities, in the permeation boundary layer formation the main velocity is very small. Secondly the thickness of the classical boundary layer decreases with a main velocity as $V^{-1/2}$, whereas in the present problem δ is independent of the main velocity. Under these circumstances, the medium behaves as though it has a viscosity $\eta \sim V/q_0$ which is extremely small $\sim 10^{-4}-10^{-5}$ g/cm² i.e., behaves somewhat like a superfluid.

4.3 Special thermal effects

(i) The Lehmann rotation phenomenon: Let the cholesteric liquid crystal be confined between two parallel plates normal to the twist axis with a temperature gradient $T_{,3}$ along the plate normal, and further let $P_{,i} = 0$. In this geometry $v_3 = 0$. Then the dynamical equations yield

$$-q_0 u = (\lambda_3/\lambda_1)T_{,3}, \tag{30}$$

$$v_i = 0. (31)$$

As the layers cannot move along z-axis we immediately conclude that uq_0 represents an angular motion about the twist axis. The angular velocity with which the structure rotates about z-axis is

$$\Omega = (\lambda_3/\lambda_1)T_{-3}. \tag{32}$$

This is referred to as Lehmann's rotation (Lehmann 1900). The above relation was first arrived at by Leslie (1968b) from his detailed theory without resorting to the super continuum approximation.

(ii) Permeation: Let the cholesteric be between two plates with its axis parallel to the plates and let there be a temperature gradient along the twist axis. If the layers are blocked, i.e. $\dot{u} = 0$, we find in the absence of any pressure gradient, a steady flow along the twist axis described by more or less a flat velocity profie. The uniform velocity is given by

$$v_3 = (\lambda_3/\lambda_1 q_0) T_{,3}. \tag{33}$$

This flow can be either along or opposite to the imposed temperature gradient depending upon the sign of λ_3 (Jayaram et al 1983). Again one finds heat conduction mediated by this process. The extra thermal conductivity is given by (Ranganath 1983)

$$K^{e} = (\lambda_{3}/\lambda_{1}q_{0})\rho ST. \tag{34}$$

It must be remarked that thermal permeation as described by (33) is the opposite of what Prost (1972) has discussed. Under conditions of steady heat flow, with blocked layers, we get from (12)

$$T_{,3} = (K_3/K_{\parallel})v_3 q_0, \tag{35}$$

i.e., permeation v_3 induces a temperature gradient. This permeation itself can be brought about by an imposed pressure gradient.

(iii) Convection: As in normal fluids, in cholesterics also a roll instability can be initiated in a cholesteric film (with its twist axis normal to the boundary and heated from below) when the temperature gradient exceeds a certain critical value. However, in cholesterics the critical threshold is much smaller than what we find in a normal fluid (Dubois-Violette 1973). This is mainly due to the fact that layer undulation gets thermally coupled to temperature gradient through thermal conductivity anisotropy. In such a situation instability sets in the moment the buoyancy force overcomes the elastic forces of undulation which are much smaller than the viscous shear forces. Depending upon the sign of conductivity anisotropy one can have a roll instability even when the upper plate is hotter.

Dubois-Violette and de Gennes (1975) discussed a totally different form of thermal instability under gravity and an imposed temperature profile. Here the cholesteric layers are undistorted and convection is primarily controlled by permeation. Since a very similar instability is triggered by thermomechanical effect alone (i.e. even when buoyancy is absent) we discuss it here.

Let us confine ourselves to a two-dimensional flow in the xz plane, z direction coinciding with twist axis. Then the equations of motion can be simplified to

$$\psi_{,xx} = (-1/\lambda_1 q_0^2) [(\gamma_2 \psi_{,zzxx} + \gamma_3 \psi_{,zzzz}) + (f_{x,z} - f_{z,x}) + \lambda_3 q_0 T_{,xz}].$$
(36)

Here ψ is the stream function

$$f_x = \rho gT\beta \cos \alpha,$$

$$f_y = \rho T\beta \sin \alpha,$$

$$v_x = \psi_{,z}, \quad v_z = -\psi_{,x},$$

 β is the volume coefficient of expansion and α is the angle between the vertical and the z-axis.

To a good approximation the temperature distribution is given by

$$K_{\parallel}T_{,zz} + K_{\perp}T_{,xx} \approx 0, \tag{37}$$

i.e

$$T(z, x) = T_q(z)\cos qx \tag{38}$$

We shall impose a temperature profile with $qL \ll 1$, L being the sample thickness. Then

$$T_q(z) = A \cosh \overline{q}z + B \sinh \overline{q}z,$$

 $\overline{q} = q (K_{\parallel}/K_{\perp})^{1/2},$ (39)
 $A \neq 0$ $B = 0$ gives the even mode,
 $A = 0$ $B \neq 0$ gives the odd mode.

Since $qL \ll 1$, variations along x are slow and we can simplify (36) to

$$\psi_{,xx} - \frac{1}{K^2} \psi_{,zzzz} = (-1/\lambda_1 q_0^2) [(f_{x,z} - f_{z,x}) + \lambda_3 q_0 T_{,xz}], \tag{40}$$

with

$$K^2 = -(\lambda_1/\gamma_3)q_0^2.$$

It can be shown that for $Kq^2L \gg 1$ permeation effects dominate. Then in the body of the specimen we have

$$\psi = (1/\lambda_1 q^2 q_0^2)(f_z \sin \alpha - f_x \cos \alpha + \lambda_3 q_0 T_{xz}). \tag{41}$$

Dubois-Violette and de Gennes (1975) discussed the situation with $\lambda_3 = 0$. The flow patterns are very different from the roll instability we find in normal fluids. With λ_3 alone we can get similar effects. For example for an imposed temperature profile (Ranganath 1983)

$$T = B \sinh \overline{q}z \cos qx$$
,

the motions in the central region are given by

$$v_1 = -B(\lambda_3/\lambda_1)(\overline{q}^2/qq_0)\sinh \overline{q}z \sin qx,$$

$$v_2 = 0,$$

$$v_3 = B(\lambda_3/\lambda_1)(\overline{q}/q_0)\cosh \overline{q}z \cos qx.$$

However near the boundary, velocities will be different since we have to satisfy the boundary condition

$$\psi = 0, \quad \psi_z = 0.$$

But it can be shown that these altered velocities are confined to a narrow region of thickness $\delta \sim [1/(Kq)^{1/2}]$ near the walls.

5. Chiral smectic C hydrodynamics

Finally, it may be remarked that many of the ideas discussed earlier are applicable to the chiral smectic C (or smectic C*) phase which consists of a helical stack of smectic C layers. The hydrodynamics of smectic C has been developed by Martin et al (1972) who have shown that as regards first order elasticity, smectic C is not any different from smectic A. It allows a second sound at velocities given essentially by (19). This transverse sound mode has layer displacements (or fluctuation in the phase of the layers) accompanying it. Now in many respects (if one ignores the in-plane distortion of the C* director) smectic C* is like a cholesteric. Hence at long wavelengths, low amplitidues we should have another second sound mode, similar to that for the cholesteric, with a

velocity much smaller than that for the normal smectic C and which has fluctuations in the phase of the C* director. Thus in smectic C* we have two types of second sound, a fast mode with layer displacements and a slow mode with the phase of the C* director fluctuating in it and the layers moving more or less as a whole. In the smectic C phase such director fluctuations are non-propagating.

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