

NONLINEAR HYDRODYNAMICS OF STRONGLY DEFORMED SMECTIC C AND C* LIQUID CRYSTALS

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The statics and dynamics of smectic C (and C*) liquid crystals has been a long-standing topic for investigations. All the previous descriptions, however, have been restricted in some sense. Either the theories were linear, or flat layers were assumed, or the layer thickness was taken to be constant, or only statics was considered, or the description (in the chiral case) included terms not compatible with layering. Here we will give a nonlinear hydrodynamic theory, which applies to strongly curved layers, non-flat 'ground' states, strongly compressed layers as well as strongly deformed director orientations. Some discrepancies between previous theories are resolved.

Keywords: hydrodynamics; ferroelectric liquid crystals; strong deformations

INTRODUCTION

A. Saupe was the first to realize that smectic C phases have a nematic degree of freedom. The direction of the tilt (the in-plane director) has to be specified in order to completely describe the phase. Deformations of that angle variable give rise to 4 Frank-like orientational elastic coefficients, since the phase is biaxial [1]. Thereafter, the elasticity of the layers has been added comprising one (layer) compressional modulus, 3 layer curvature coefficients (generalized splay coefficients – the generalized bend coefficients are usually neglected) and 2 contributions, where layer curvature and in-plane director bending are mixed [2]. On the basis of broken symmetries the linearized hydrodynamics of smectic C phases was given in [3]. Due to the monoclinic symmetry of that phase (requiring all equations to be invariant under the combined replacement of the layer normal $\hat{\mathbf{k}}$ and the in-plane director $\hat{\mathbf{c}}$ by $-\hat{\mathbf{k}}$ and $-\hat{\mathbf{c}}$, respectively) there are 13 (ordinary) flow viscosities, 4 thermal conductivities, one permeation coefficient, one director reorientation viscosity, 2 thermo-permeation coefficients, describing viscous cross-couplings between temperature gradients and layer deformations. As in any

system with nematic degrees of freedom there are reactive (non-dissipative) transport parameters relating flow and director orientation, i.e. for smectics C there are 2 (only one of which gives rise to shear flow alignment [4]). There are two additional hydrodynamic degrees of freedom (compared to simple fluids): the displacement of layers u along the layer normal (due to the broken translational symmetry) and rotations of the in-plane director $(\hat{\mathbf{k}} \times \hat{\mathbf{c}}) \cdot \delta \hat{\mathbf{c}}$ due to the broken rotational symmetry (in the layers). Of course, the existence of layers also breaks rotational symmetry, since the layer normal defines a preferred direction. However, deformations of that direction, $\delta \hat{\mathbf{k}}$, do not constitute independent variables, but are described by gradients of the layer displacement. This is true not only in linear approximation $\delta \hat{\mathbf{k}} = -\nabla_{\perp} u$, but generally

$$\hat{\mathbf{k}} = \nabla \Phi (|\nabla \Phi|)^{-1} \quad (1)$$

where Φ is the phase variable describing the periodic layer structure with $\Phi = z - u$ for flat layers along the z-direction.

In the chiral smectic C* phase the tilt direction changes from layer to layer in a helicoidal fashion and the director is conic helical, where the helix axis coincides with the layer normal. Simultaneously an in-plane polarization \mathbf{P} (perpendicular to both, the director and the layer normal) occurs, which is thus helical. In such a structure the translational symmetry along the layer normal (or helix axis) is broken twice and independently by the layer and the helix structure. Again there are two additional hydrodynamic variables, when compared to simple fluids [5]: layer displacement u_A and helix displacement u_C , where the latter is equivalent to helix rotation (about the helical axis). Although one could think that the helix displacement variable can only be used on length scales larger than the pitch $2\pi/q_0$ (global description), however, by using the helix rotation, $(\mathbf{P}_0 \times \delta \mathbf{P}) \cdot \hat{\mathbf{k}} = q_0 u_C$, it turns out that this description is useful also for much smaller length scales (local description). Again, curvature of the layers and of the helix structure is described by gradients of u_A and of u_C , respectively. Of course, if additionally the tilt angle or the magnitude of the polarization are used, non-hydrodynamic variables are introduced.

The theories described above are linear in the sense that deviations from the ground state (flat layers) are assumed to be small. However, they are nonlinear with respect to director and layer orientation, since all material tensors implicitly depend on those orientations giving rise to a host of intrinsic nonlinear effects. Nevertheless there is a need for dynamic theories that are applicable for situations of strong layer curvature or non-flat 'ground' states due to external fields or boundary conditions. An early, not quite satisfactory attempt in that direction has been made by the present authors [6]. For the chiral smectic C* phase an approach more in the spirit of a Ginzburg-Landau description has been proposed [7] allowing for chiral

terms that are not compatible with the layer structure, but would lead to a different ground state with a different symmetry and different broken symmetries. Quite recently the statics of strongly deformed (layered) smectic as well as discotic phases (including strong curvature and compression) has been discussed [8]. A dynamic theory based on continuum mechanics of smectic C and C* phases with strong layer curvature, but with constant layer spacing (no true elasticity, i.e. no compression or dilation and with flat layers as ground state) has been given also quite recently [9]. There is also a dynamic theory for smectic A liquid crystals using the phase Φ as variable [10].

THERMODYNAMICS AND STATICS

Using the phase Φ as variable it is obvious that homogeneous changes, $\delta\Phi$, do not cost energy, since they correspond to a rigid translation of the layer structure. Only changes of the layer thickness cost energy and are described by $d(|\nabla\Phi| - 1) = \hat{\mathbf{k}} \cdot d\nabla\Phi$, where $\hat{\mathbf{k}}$ is the layer normal defined in eq.(1). Its direction is not fixed energetically (in the absence of external fields), thus rigid rotations of $\hat{\mathbf{k}}$ (i.e. rigid rotations of the layers) do not cost energy. Thus, only gradients of the layer normal can enter the free energy, which are, however, expressed by higher order gradients of Φ (using eq.(1))

$$\nabla_l \hat{k}_i = |\nabla\Phi|^{-1} (\delta_{ij} - \hat{k}_i \hat{k}_j) \nabla_j \nabla_l \Phi. \quad (2)$$

This is the manifestation of the fact that the broken rotational symmetry due to the layer normal is slaved by the broken translational symmetry of the layers themselves and does not give rise to independent hydrodynamic degrees of freedom. Of course, eq.(2) ensures the identity $\hat{k}_i \nabla_l \hat{k}_i \equiv 0$.

The director $\hat{\mathbf{n}}$ constitutes a different broken rotational symmetry. Rigid rotations of the director do not cost energy, if they are done together with the whole structure, i.e. if the tilt angle ψ between $\hat{\mathbf{k}}$ and $\hat{\mathbf{n}}$ is kept fixed, $\hat{\mathbf{k}} \cdot \hat{\mathbf{n}} = \cos\psi = \text{const.}$. Thus, a rotation $\hat{\mathbf{k}} \cdot \delta\hat{\mathbf{n}}$ is equivalent to $-\hat{\mathbf{n}} \cdot \delta\hat{\mathbf{k}} = -|\nabla\Phi|^{-1} \sin\psi \hat{\mathbf{c}} \cdot \delta\nabla\Phi$, where we have made use of the in-plane director $\hat{\mathbf{c}}$ defined by $\hat{\mathbf{n}} = \hat{\mathbf{k}} \cos\psi + \hat{\mathbf{c}} \sin\psi$. For the same reason rotations $\hat{\mathbf{c}} \cdot \delta\hat{\mathbf{n}} = \cos\psi \hat{\mathbf{c}} \cdot \delta\hat{\mathbf{k}}$ are not independent, but described again by gradients of Φ . Only rotations of the director about the layer normal, $\hat{\mathbf{p}} \cdot \delta\hat{\mathbf{n}}$ are independent. Here we use the orthogonal triad $\hat{\mathbf{k}}$, $\hat{\mathbf{c}}$, and $\hat{\mathbf{p}} \equiv \hat{\mathbf{k}} \times \hat{\mathbf{c}}$ as basic coordinate system, although one could use also $\hat{\mathbf{n}}$, $\hat{\mathbf{p}}$, and $\hat{\mathbf{c}}' \equiv (\hat{\mathbf{k}} - \hat{\mathbf{n}} \cos\psi)(\sin\psi)^{-1}$. Of course, only inhomogeneous rotations, $\hat{p}_i \nabla_j n_i$ can enter the free energy.

The gradient free energy is of the form

$$\begin{aligned}
f_g &= \frac{B}{2}(|\nabla\Phi| - 1)^2 + \frac{1}{2}K_{jl}^{(n)}p_i p_k(\nabla_j n_i)(\nabla_l n_k) \\
&+ \frac{1}{2}K_{ijkl}^{(k)}(\delta_{im} - k_i k_m)(\delta_{kn} - k_k k_n)(\nabla_j k_m)(\nabla_l k_n) \\
&+ \frac{1}{2}K_{ijl}^{(m)}p_k(\delta_{im} - k_i k_m)(\nabla_j k_m)(\nabla_l n_k)
\end{aligned} \tag{3}$$

where the gradients of $\hat{\mathbf{k}}$ have to be expressed by gradients of Φ by eq.(2). One can add higher order compression or dilation, i.e. terms $\sim (k_i k_j \nabla_i \nabla_j \Phi)^2$ and crosscouplings between compression/dilation with temperature and density variations. Note that gradients of $\hat{\mathbf{c}}$ are already contained in eq.(3). Of course, the $\hat{\mathbf{k}}$, $\hat{\mathbf{c}}$, and $\hat{\mathbf{p}}$ occurring explicitly and implicitly in eq.(3) are not constant but time and space dependent. Their variations are again expressed by $\delta\Phi$ and $\hat{\mathbf{p}} \cdot \delta\mathbf{n}$. The actual structure of the material tensors, their dependence on $\hat{\mathbf{k}}$, $\hat{\mathbf{c}}$, and $\hat{\mathbf{p}}$, follows from the monoclinic symmetry and has already been discussed in the literature.

It is somewhat tempting to use – as in biaxial nematics – the three angles $\delta\theta_1 = \hat{\mathbf{c}} \cdot \delta\hat{\mathbf{k}}$, $\delta\theta_2 = \hat{\mathbf{p}} \cdot \delta\hat{\mathbf{k}}$, and $\delta\theta_3 = \hat{\mathbf{p}} \cdot \delta\hat{\mathbf{c}}$, in order to describe the orientational degrees of freedom. In that case, however, one must consider that these angles are not defined globally, since three-dimensional rotations about different axes are generally non-commutable. This leads to the so-called Mermin-Ho relations for biaxial nematics [11]

$$(\delta_1 \delta_2 - \delta_2 \delta_1)\boldsymbol{\theta} = (\delta_1 \boldsymbol{\theta}) \times (\delta_2 \boldsymbol{\theta}) \tag{4}$$

where δ_1 and δ_2 are any first order differential operator and where $\boldsymbol{\theta} = (\theta_1, \theta_2, \theta_3)$. Because of these rather complicated relations and because θ_1, θ_2 are not independent of each other due to eq.(2), it is not very appropriate to use this angle-variable description for the dynamics below.

In the presence of external fields some or all of the rotational symmetries are broken externally by these fields. Thus, already homogeneous rotations enter the free energy as can be seen from the dielectric energy

$$4\pi f_{diel} = \frac{1}{2}\epsilon_1(\hat{\mathbf{n}} \cdot \mathbf{E})^2 + \frac{1}{2}\epsilon_2(\hat{\mathbf{k}} \cdot \mathbf{E})^2 + \frac{1}{2}\epsilon_3(\hat{\mathbf{p}} \cdot \mathbf{E})^2 + \epsilon_4(\hat{\mathbf{n}} \cdot \mathbf{E})(\hat{\mathbf{k}} \cdot \mathbf{E}) \tag{5}$$

which could also be expressed in terms of $\hat{\mathbf{c}}$ instead of $\hat{\mathbf{n}}$. Eq.(5) as well as the flexoelectric energy

$$f_{flex} = e_{ij}^{(n)}\hat{p}_k E_i \nabla_j n_k + e_{kij}^{(k)}(\delta_{kl} - \hat{k}_k \hat{k}_l)E_i \nabla_j k_l \tag{6}$$

can be expressed by the variables Φ and $\hat{\mathbf{p}} \cdot \delta\hat{\mathbf{n}}$.

Now we can write down the Gibbs relation connecting the differentials of variables with entropy density ($d\sigma$) or energy density changes ($d\epsilon$)

$$d\epsilon = \mu d\rho + T d\sigma + \mathbf{v} \cdot d\mathbf{g} + \frac{1}{4\pi} \mathbf{E} \cdot d\mathbf{D} + \tilde{\Omega} \hat{\mathbf{k}} \cdot d(\nabla\Phi) \quad (7)$$

$$\begin{aligned} & + \Psi'' \hat{k}_i \hat{k}_j d(\nabla_i \nabla_j \Phi) + \tilde{\Psi}_{il} d\left(|\nabla\Phi|^{-1} (\delta_{ij} - \hat{k}_i \hat{k}_j) \nabla_j \nabla_l \Phi\right) \\ & + \Omega' \hat{\mathbf{c}} \cdot d(\nabla\Phi) + \Omega'' \hat{\mathbf{p}} \cdot d(\nabla\Phi) + H_j d(\hat{p}_i \nabla_j n_i) + h' \hat{\mathbf{p}} \cdot d\mathbf{n} \\ = & \mu d\rho + T d\sigma + \mathbf{v} \cdot d\mathbf{g} + \frac{1}{4\pi} \mathbf{E} \cdot d\mathbf{D} + \boldsymbol{\Omega} \cdot d(\nabla\Phi) \quad (8) \\ & + \Psi_{ij} d(\nabla_i \nabla_j \Phi) + H_j \hat{p}_i d(\nabla_j n_i) + h \hat{\mathbf{p}} \cdot d\mathbf{n} \end{aligned}$$

with

$$\begin{aligned} \boldsymbol{\Omega} = & \hat{\mathbf{k}} \left(\tilde{\Omega} - \tilde{\Psi}_{il} |\nabla\Phi|^{-2} (\delta_{ij} - \hat{k}_i \hat{k}_j) (\nabla_l \nabla_j \Phi) \right) \quad (9) \\ & + \hat{\mathbf{c}} \left(\tilde{\Omega}' - \tilde{\Psi}_{il} |\nabla\Phi|^{-2} (\hat{k}_i \hat{c}_j + \hat{k}_j \hat{c}_i) (\nabla_j \nabla_l \Phi) \right) \\ & + \hat{\mathbf{p}} \left(\Omega'' - \tilde{\Psi}_{il} |\nabla\Phi|^{-2} (\hat{k}_i \hat{p}_j + \hat{k}_j \hat{p}_i) (\nabla_j \nabla_l \Phi) \right. \\ & \left. + (\sin\psi)^{(-1)} H_l |\nabla\Phi|^{-2} \hat{c}_j (\nabla_l \nabla_j \Phi) \right) \end{aligned}$$

and

$$\Psi_{ij} = \Psi'' \hat{k}_i \hat{k}_j + \tilde{\Psi}_{qi} |\nabla\Phi|^{-1} (\delta_{qj} - \hat{k}_q \hat{k}_j) \quad (10)$$

and

$$h = h' - \cot\psi H_l |\nabla\Phi|^{-1} \hat{c}_j (\nabla_l \nabla_j \Phi) \quad (11)$$

where we have introduced the mass density ρ , the momentum density \mathbf{g} and the dielectric displacement field \mathbf{D} as well as the conjugate quantities chemical potential μ , temperature T , velocity \mathbf{v} and electrical field \mathbf{E} . The elastic stress conjugate to layer compression or dilation is $\tilde{\Omega}$, to inhomogeneous compression or dilation is Ψ'' , while $\tilde{\Psi}_{il}$ is conjugate to inhomogeneous layer rotations (i.e. curvature of the layers). The conjugate H_j is related to director orientational elasticity, while h' , Ω' and Ω'' are conjugate to director and layer (normal) rotations, respectively. The latter quantities have to vanish either with external field strength (squared) or they are nonlinear and vanishing with the wave vector squared at least. Explicit expressions for the conjugate quantities can be obtained by partial derivatives of the energy expression with respect to the appropriate variable, but will not be given here for lack of space.

Instead of the phase Φ one could use the displacement u along the layer (z -) axis by the transformation $\Phi = z - u$. As long as gradients of u are kept to any order, this is still an exact description, although it is no longer

coordinate-free and the manifest rotational invariance is lost. For almost flat layers, however, where one can expand the expression in the (small) gradients of u , this leads back to the well-known linear or weakly nonlinear theories.

HYDRODYNAMICS

Having discussed the static properties in the preceding section we are prepared to set up the hydrodynamic equations, which are either conservation laws or balance equations for variables associated with spontaneously broken continuous symmetries. They are of the general form [12]

$$\dot{\rho} + \nabla_i(\rho v_i) = 0 \quad (12)$$

$$\dot{g}_i + \nabla_j(g_i v_j) + \nabla_i(p - \frac{\mathbf{E}^2}{8\pi}) + \nabla_j(\sigma_{ij}^{(s)} + \sigma_{ij}) = \rho_e E_i + P_j \nabla_j E_i \quad (13)$$

$$\dot{\sigma} + \nabla_i(\sigma v_i) + \text{div} \mathbf{j}^\sigma = \frac{R}{T} \quad (14)$$

$$\dot{\epsilon} + \nabla_i([\epsilon + p]v_i) + \text{div} \mathbf{j}^\epsilon = 0 \quad (15)$$

$$\dot{D}_i + v_i \nabla_j D_j + 4\pi j_i^e = 0 \quad (16)$$

$$\dot{\Phi} + v_i \nabla_i \Phi + X = 0 \quad (17)$$

$$\hat{\mathbf{p}} \cdot \hat{\mathbf{n}} + \hat{p}_i v_j \nabla_j n_i + (\hat{\mathbf{p}} \times \hat{\mathbf{n}}) \cdot \boldsymbol{\omega} + \hat{p}_i Y_i = 0 \quad (18)$$

where the dot denotes a partial time derivative $\partial/\partial t$. In eq.(13) $\rho_e = (1/4\pi)\text{div} \mathbf{D}$ is the electric charge density and $\mathbf{P} = (1/4\pi)(\mathbf{D} - \mathbf{E})$ is the polarization. The nonlinear convective terms shown in eqs.(12-18) are required by Galilean invariance. The nonlinear term in the director equation (18) involving the vorticity $\boldsymbol{\omega}$ ($\boldsymbol{\omega} \equiv (1/2)\text{curl} \mathbf{v}$) is required by rotational symmetry. These contributions are balanced, in order to give zero entropy production ($R = 0$), by the pressure and the part of the stress tensor determined by symmetry $\sigma_{ij}^{(s)}$. For the pressure the Gibbs-Duhem relation gives

$$p = -\epsilon + \mu\rho + T\sigma + v_i g_i + \frac{1}{4\pi} E_i D_i \quad (19)$$

$$\begin{aligned} dp &= -\rho d\mu - \sigma dT - g_i dv_i - \frac{1}{4\pi} D_i dE_i - \boldsymbol{\Omega} \cdot d(\nabla\Phi) \\ &\quad - \Psi_{ij} d(\nabla_i \nabla_j \Phi) - H_j \hat{p}_i d(\nabla_j n_i) - h \hat{\mathbf{p}} \cdot d\mathbf{n} \end{aligned} \quad (20)$$

while for $\sigma_{ij}^{(s)}$ we find

$$\begin{aligned} \sigma_{ij}^{(s)} &= \frac{1}{2} h^{(tot)} (\hat{n}_j \hat{p}_i - \hat{n}_i \hat{p}_j) + \frac{1}{2} \cot\psi H_q (\hat{k}_j \hat{c}_i - \hat{k}_i \hat{c}_j) \hat{p}_k \nabla_q n_k \\ &\quad - \frac{1}{2} (\sin\psi)^{(-1)} H_q |\nabla\Phi|^{-1} (\hat{k}_j \hat{c}_i - \hat{k}_i \hat{c}_j) \hat{p}_k \nabla_k \nabla_q \Phi \\ &\quad + \Psi_{jl} (\nabla_i \nabla_l \Phi) + \Omega_j^{(tot)} \nabla_i \Phi + H_j \hat{p}_l \nabla_i n_l \end{aligned} \quad (21)$$

where we have introduced the convenient abbreviations

$$\Omega_i^{(tot)} = \Omega_j - \nabla_j \Psi_{ij} \quad (22)$$

$$h^{(tot)} = h - \nabla_i H_i \quad (23)$$

In eq.(21) the first three terms are antisymmetric contributions due to the nematic degree of freedom, while the last three are generalizations (due to the layers) of the Leslie-Ericksen stress.

The phenomenological currents σ_{ij} , \mathbf{j}^σ , \mathbf{j}^e and the quasi-currents X and Y_i are each a sum of a reversible and an irreversible part, due to zero ($R = 0$) and positive definite ($R > 0$) entropy production, respectively. They are characterized by reactive and dissipative transport parameters. The energy current density then follows from the Gibbs relation. For the reversible parts we find

$$Y_i^{(R)} = \hat{p}_i \lambda_{jk} \frac{1}{2} (\nabla_j v_k + \nabla_k v_j) \quad (24)$$

$$\sigma_{ij}^{(R)} = -\frac{1}{2} (\lambda_{ij} + \lambda_{ji}) h^{(tot)} \quad (25)$$

$$\mathbf{j}^{\sigma(R)} = 0 = \mathbf{j}^{e(R)} = 0 = X^{(R)} \quad (26)$$

with $\lambda_{ij} = \lambda_1 \hat{p}_i \hat{k}_j + \lambda_3 \hat{p}_i \hat{c}_j$ containing two reactive transport parameters [4], one of which corresponds to the flow alignment parameter of uniaxial nematics. Note that there are no flow-alignment-like terms with respect to the layer normal $\hat{\mathbf{k}}$. Such terms are possible in a biaxial nematic, but they are not possible in a layered structure.

The irreversible parts of the currents and quasi-currents are obtained from an entropy production functional that is bilinear in the thermodynamic forces, i.e. we restrict ourselves here to linear irreversible thermodynamics (cubic and quartic terms in the entropy production have been discussed in [13]). Nevertheless, the expressions obtained will be highly nonlinear when written in the variables.

$$j_i^{\sigma(D)} = -\kappa_{ij} \nabla_j T - \kappa_{ij}^{(E)} E_j - \xi_i^{(T)} \text{div} \mathbf{\Omega}^{(tot)} \quad (27)$$

$$j_i^{e(D)} = \sigma_{ij}^{(E)} E_j + \kappa_{ij}^{(E)} \nabla_j T + \xi_i^{(E)} \text{div} \mathbf{\Omega}^{(tot)} + \nabla_j (\zeta_{ji}^{(E)} h^{(tot)}) \quad (28)$$

$$\sigma_{ij}^{(D)} = -\frac{1}{2} \nu_{ijkl} (\nabla_k v_l + \nabla_l v_k) \quad (29)$$

$$X^{(D)} = -\xi \text{div} \mathbf{\Omega}^{(tot)} - \xi_i^{(T)} \nabla_i T - \xi_i^{(E)} E_i \quad (30)$$

$$Y_i^{(D)} = \frac{1}{\gamma_1} \hat{p}_i h^{(tot)} - \hat{p}_i \zeta_{jk}^{(E)} \nabla_j E_k \quad (31)$$

The energy density current \mathbf{j}^e is an extremely complicated expression, but fortunately it will not be needed, because the energy conservation law is redundant due to the Gibbs relation eq.(8).

Eqs. (27-31) contain in the field-free case the same number of dissipative coefficients as Ref. [3] (cf. the discussion in the Introduction). In [9] much more viscosity-like coefficients were found. This is not because ref. [3] used linear irreversible thermodynamics for the dissipative dynamics (so did we in the present paper as well as ref. [9]), but because in [9] *three* dynamic variables (the three rotation angles of the triad $\hat{\mathbf{k}}$, $\hat{\mathbf{c}}$, and $\hat{\mathbf{p}}$) were used as is suitable for monoclinic biaxial nematics, while for smectic C liquid crystals only *two* dynamic variables (in-plane director rotation and the phase of the layering) are appropriate. Thus, the existence of layers (i.e. the requirement for $\hat{\mathbf{k}}$ being a layer normal as is expressed in eq.(1)) reduces the number of coefficients as has been seen above for the flow alignment parameters. With external electric fields eqs. (27-31) contain additionally 4 electric conductivities, 4 thermo-electric diffusivities, 2 electro-permeative and 2 dynamic flexoelectric coefficients.

A more extended discussion including chiral smectic C* liquid crystals will be given elsewhere.

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