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# Balance Laws and Constitutive Equations for Liquid Crystals

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Liquid crystals consist of form-anisotropic particles which posses in most cases a uniaxial symmetry but in general the particles have a biaxial shape. We want to show how both cases can be treated within a uniform mathematical model. This model can be used to obtain a set of "mesoscopic" balance equations containing the orientation as additional variable. Introducing an orientation distribution function (ODF) for the rotational degrees of freedom the moments of the ODF – called alignment tensors – can be used as order parameters of the liquid crystal. Constitutive equations are formulated on the mesoscopic level and can be lifted to macroscopic fields by an averaging procedure. The resulting equations are valid for the whole physical range of the order parameters, covering the isotropic phase as well as the completely ordered nematic phase. To demonstrate the usefulness of the method we shortly discuss the viscous properties of nematic liquid crystals.

# Introduction

Liquid crystals are examples of continua with micro-structure. They exhibit at least one mesophase which is liquid but show additional properties usually associated with crystalline structures (e.g. spontaneous birefringence). These are caused by a long-range order of orientations of the non-spherical constituents (molecules in thermotropic liquid crystals or "micells" in lyotropic liquid crystals) whereas the fluid-like behaviour is due to the uncorrelated distribution of centers of mass over long distances.

Mathematical models of liquid crystals are obtained by regarding the particles forming the fluid as rigid bodies which are subjected to some kind of statistics characterizing the local degree of order of their molecular axes. In most of the cases the model can be further simplified by assuming additional symmetries as axial symmetric shapes of the particles, often in connection with a "head-tail" (inversion) symmetry. However, during the last ten years liquid crystals have been found which are clearly built up by biaxial particles[7]. Thus it is advantageous to describe both classes of liquid crystals in a generalized framework which can be specialized to obtain nematic liquid crystals of rod-like particles or liquid crystals with internal lower symmetries.

#### 1. MATHEMATICAL MODEL

1.1. Configuration Spaces. The orientation of a rod-like structure can be described by a single unit vector  $\underline{\mathbf{n}}$ . However, if the local distribution of particles does not distinguish between orientations  $\underline{\mathbf{n}}$  and  $-\underline{\mathbf{n}}$  (or if the particles themselves posses such a symmetry) we have a superimposed symmetry which is represented by choosing the real projective plane  $\mathbb{R}P^2$  to classify the orientation. Thus a natural configuration space for nematic liquid crystals is (including the degrees of freedom of displacement of the centers of mass)

$$CS_1 \equiv \mathbb{R}^3 \times \mathbb{R}P^2 \ . \tag{1}$$

The index refers to the fact that the constituents posses a uniaxial symmetry. For biaxial particles the orientation of the molecular axes can be described by elements of the proper orthogonal group SO(3). Thus our most general configuration space treated here is

$$CS_2 \equiv \mathbb{R}^3 \times SO(3) \ . \tag{2}$$

However, in both the cases the orientational part of  $CS_k$  ( $k \in \{1,2\}$ ) is neither orientable nor simply connected which complicates the formulation of balance laws. Using the fact that  $\mathbb{R}P^2$  and SO(3) can be covered by  $S^2$  and  $S^3$ , respectively,

$$S^2/\mathbb{Z}_2 = \mathbb{R}P^2$$
,  $S^3/\mathbb{Z}_2 = SO(3)$ 

the substitution of CS<sub>k</sub> by its (universal) covering yields

$$CS'_k \equiv \mathbb{R}^3 \times S^k \ (k \in \{1, 2\}) \ . \tag{3}$$

as new configuration spaces endowed with an inherent symmetry due to the lift of physical quantities (defined in  $\mathrm{CS}_k$ ) to  $\mathrm{CS}_k'$ . It should be noted that the

projection  $\pi_k$  of the bundle  $(CS'_k, CS_k, \pi_k)$  can be chosen in such a manner that all balance laws and constitutive equations have to be invariant under the substitution of  $-\underline{n} \in S^k$  for  $\underline{n} \in S^k$  which is often called "nematic symmetry". For details we refer to [9].

1.2. ORIENTATION DISTRIBUTION FUNCTION. To describe the orientational order of the anisotropic fluid at given time and position a one-particle distribution function called orientation distribution function (ODF) is introduced

$$f_k: CS'_k \longrightarrow \mathbb{R}_0^+$$
 (4)

which is normalized and symmetric on  $\mathrm{CS}_k'$  according to

$$\oint_{S^k} f(\underline{x}, \underline{n}, t) d^k n = 1$$

$$f(\underline{x}, -\underline{n}, t) = f(\underline{x}, \underline{n}, t) .$$
(5)

$$f(\underline{x}, -\underline{n}, t) = f(\underline{x}, \underline{n}, t) . \tag{6}$$

Thus  $f_k(\underline{x},\underline{n})$  can be interpreted as probability density describing the orientation of all particles in a spatial volume element located at position  $\underline{x}$  at time t.

1.3. KINEMATICS IN  $CS'_k$ . Using the "nematic spaces"  $CS'_k$  the evolution of the liquid crystal is traced with respect to spatial translations as well as to orientational changes. Thus, a transport theorem in  $\mathrm{CS}_k'$  is needed.

PROPOSITION 1. Let  $\mathfrak{G}$  be a (time dependent) region in  $CS'_k$  and  $X \in$  $C^1(CS'_k)$  a function. Let the evolution of  $\mathfrak{G}$  be given by velocity fields  $(\underline{\nu},\underline{\mathfrak{u}})$ of the tangential bundle  $TCS'_k$ . We denote the covariant derivatives of the Levi-Civita connection on  $\mathbb{R}^3$  and  $S^k$  with  $\nabla_x$  and  $\nabla_n$ , respectively. Then

$$\begin{split} \frac{d}{dt} \iint\limits_{\mathfrak{G}} X(\underline{x},\underline{n},t) \, d^k n \, d^3 x &= \iint\limits_{\mathfrak{G}} \left\{ \frac{\partial}{\partial t} X(\underline{x},\underline{n},t) + \nabla_x \cdot (\underline{\nu}(\underline{x},\underline{n},t) X(\underline{x},\underline{n},t)) + \right. \\ &\left. + \nabla_n \cdot (\underline{u}(\underline{x},\underline{n},t) X(\underline{x},\underline{n},t)) \right\} \, d^k n \, d^3 x \end{split} \tag{7}$$

is valid.

Applying this theorem to material volumes in CS<sub>k</sub> (or "modified bodies", see [3, 4, 5, 6]) balance equations on nematic space can be obtained.

1.4. ALIGNMENT TENSORS. Although the knowledge of the ODF  $f_k$  is sufficient to calculate the properties of a liquid crystal concerning orientation,  $f_k$  itself is unknown in most cases. Thus, a set of (tensorial) order parameters called alignment tensors [12, 13, 15] is introduced.

PROPOSITION 2. Let  $f \in L^2(S^k)$  be a square-integrable function on the k-dimensional unit sphere with  $k \geq 2$ . Then the symmetric irreducible tensors  $\{\underbrace{\overline{n\cdots n}}_{\ell \text{ conies}}\}_{\ell \in \mathbb{N}}$  form a basis of  $L^2(S^k)$  and f can be expanded in a series (denoting

Cartesian components with Greek indices and using summation convention for them)

$$f(\underline{n}) = \frac{1}{A(k)} \left[ f_0 + \sum_{\ell=1}^\infty \frac{(2\ell+k-1)!!}{\ell!(k-1)!!} \alpha_{\mu_1 \dots \mu_\ell} \ \overline{n_{\mu_1} \cdots n_{\mu_\ell}} \right] \ . \label{eq:force}$$

Here, A(k) denotes the surface area of the sphere  $S^k$  (in fact  $A(2) = 4\pi$  and  $A(3) = 2\pi^2$ ), and  $f_0$ ,  $a_{\mu_1...\mu_\ell}$  are given by

$$\begin{array}{rcl} f_0 & = & \oint\limits_{S^k} f(\underline{n}) \, d^k n \ , \\ \\ a_{\mu_1 \dots \mu_\ell} & = & \oint\limits_{S^k} f(\underline{n}) \ \overline{n_{\mu_1} \cdots n_{\mu_\ell}} \ d^k n \end{array}$$

as the moments of f.

In the case of the ODF  $f_k$  the odd moments vanish (because of the symmetry of  $f_k$ ). The remaining moments are called the *alignment tensors* of order  $\ell$ , where  $\ell$  runs through all even integers.

In many physical situations only a few of them (e.g. only  $\underline{\underline{a}}$ ) are relevant and experimentally available. Thus the problem of obtaining  $f_k$  can often be reduced to the task of determining the alignment tensor of second and sometimes of fourth or higher order.

# 2. Balance Equations.

In the spirit of a mesoscopic theory of liquid crystals [3, 4, 5, 6, 9] balance equations on  $CS'_k$  are needed. For more details see the references listed above, here we restrict ourselves to the presentation of the resulting equations.

The general case of an orientational balance equation is formulated as (the argument  $(\underline{x}, \underline{n}, t)$  is suppressed)

$$\frac{\partial}{\partial t}X + \nabla_x \cdot \left(\underline{\nu}X + \underline{J}_X^x\right) + \nabla_n \cdot \left(\underline{u}X + \underline{J}_X^n\right) = \operatorname{Prod}(X) \tag{8}$$

with  $\underline{J}_X^x$  and  $\underline{J}_X^n$  denoting spatial and orientational non-convective fluxes of X, respectively, and  $\operatorname{Prod}(X)$  referring to production and supply of X. Special cases are listed below.

- 2.1. Mass.  $X=\rho(\underline{x},\underline{n},t)$  mass density per orientation  $\underline{J}_X^x=\underline{0}$   $\underline{J}_X^n=\underline{0}$   $\mathrm{Prod}(X)=0$
- 2.2. Momentum.  $X = \rho(\underline{x}, \underline{n}, t)\underline{\nu}(\underline{x}, \underline{n}, t)$  momentum density per orientation  $\underline{J}_X^x = -\underline{\underline{t}}^\top(\underline{x}, \underline{n}, t)$  (transposed) pressure tensor per orientation  $\underline{J}_X^n = -\underline{\underline{T}}^\top(\underline{x}, \underline{n}, t)$  (transposed) pressure tensor per orientation on  $S^k$   $\operatorname{Prod}(X) = \rho(x, n, t)k(x, n, t)$  external force density per orientation
- 2.3. Angular momentum.  $X = \rho(\underline{x},\underline{n},t) \big(\underline{x} \times \underline{\nu}(\underline{x},\underline{n},t) + \underline{\underline{\Theta}} \cdot \underline{u}(\underline{x},\underline{n},t) \big)$  angular momentum density per orientation,  $\underline{\underline{\Theta}}$  tensor of inertia  $\underline{J}_X^x = \underline{\underline{t}}^\top(\underline{x},\underline{n},t) \times \underline{x} + \underline{\underline{m}}^\top(\underline{x},\underline{n},t)$  torque density due to stresses and couple stresses  $\underline{J}_X^n = \underline{\underline{T}}^\top(\underline{x},\underline{n},t) \times \underline{x} + \underline{\underline{m}}^\top(\underline{x},\underline{n},t)$  torque density due to stresses and couple stresses on  $S^k$   $\operatorname{Prod}(X) = \rho(\underline{x},\underline{n},t)\underline{x} \times \underline{k}(\underline{x},\underline{n},t) + \underline{m}(\underline{x},\underline{n},t)$  torque density due to external forces and couple forces
- 2.4. Energy.  $X = \rho(\underline{x}, \underline{n}, t) \left(\frac{1}{2}\underline{\nu}^2(\underline{x}, \underline{n}, t) + \frac{1}{2}\underline{\Theta} : \underline{u}\underline{u} + e(\underline{x}, \underline{n}, t)\right)$  angular energy density per orientation,  $e(\underline{x}, \underline{n}, t)$  internal energy per orientation  $\underline{J}_X^x = -\underline{\nu}(\underline{x}, \underline{n}, t) \cdot \underline{\underline{t}}(\underline{x}, \underline{n}, t) \underline{u}(\underline{x}, \underline{n}, t) \cdot \underline{\underline{\pi}}(\underline{x}, \underline{n}, t) \underline{q}(\underline{x}, \underline{n}, t)$  energy flux density due to stresses and couple stresses and heat flux density  $\underline{J}_X^n = -\underline{\nu}(\underline{x}, \underline{n}, t) \cdot \underline{\underline{T}}(\underline{x}, \underline{n}, t) \underline{u}(\underline{x}, \underline{n}, t) \cdot \underline{\underline{\Pi}}(\underline{x}, \underline{n}, t) \underline{Q}(\underline{x}, \underline{n}, t)$  their counterparts on  $S^k$   $\operatorname{Prod}(X) = \rho(\underline{x}, \underline{n}, t) \underline{k}(\underline{x}, \underline{n}, t) \cdot \underline{\nu}(\underline{x}, \underline{n}, t) + \underline{m}(\underline{x}, \underline{n}, t) \cdot \underline{u}(\underline{x}, \underline{n}, t) + r(\underline{x}, \underline{n}, t)$  power due to external forces and couple forces and radiation supply

2.5. "Orientation".  $X = f(\underline{x}, \underline{n}, t)$  orientation distribution function  $J_X^x = \underline{0}$   $J_X^n = \underline{0}$   $\operatorname{Prod}(X) = -f(\underline{x}, \underline{n}, t) \left( \frac{\partial}{\partial t} + \underline{\nu}(\underline{x}, \underline{n}, t) \cdot \nabla_x \right) \log \rho(\underline{x}, t)$ Here

$$\rho(\underline{x}, \underline{n}, t) =: f(\underline{x}, \underline{n}, t) \rho(\underline{x}, t)$$
(9)

connects mass density per orientation and (macroscopic) mass density of the liquid. The last balance equation is obtained from the orientation mass balance using this definition and the equation of conservation of mass.

Finally, we remark that balance laws on  $\mathbb{R}^3$  can be obtained by integrating the balances on  $\mathrm{CS}'_k$  over  $S^k$  [4]. They have the usual form of balances of a micro-polar medium.

# 3. Constitutive Equations.

Physical fields are tensor fields defined on (subsets of)  $\mathbb{R}^3$ . Thus, connecting them with orientational variables is simple in the case of nematics (represented by  $CS'_1$ ) where  $\{-\underline{n},\underline{n}\}$  are 3-vectors. However, for biaxial particles the orientational variables are 4-vectors and we have to lift all 3-tensor fields to 4-tensor fields.

3.1. PROJECTIONS AND LIFTS FOR  $CS_2'$ . We restrict the discussion on the orientation part of  $CS_2'$ . Let

be the restriction to unit vectors,

inv: 
$$S^3 \rightarrow S^3$$
  
 $\underline{n} \mapsto \operatorname{sgn}(\underline{n} \cdot \underline{e}_4)\underline{n}$  (11)

be an inversion and

$$P_4: \mathbb{R}^4 \to \mathbb{R}^3$$

$$\underline{\mathbf{n}} \mapsto \underline{\mathbf{n}} - \underline{\mathbf{n}} \cdot \underline{\mathbf{e}}_4 \underline{\mathbf{e}}_4 , \qquad (12)$$

the projector along the 4-direction with  $\{\underline{e}_i\}_{i=1,2,3,4}$  denoting the Euclidean basis of  $\mathbb{R}^4$ . Using the Levi-Civita tensor  $\underline{\varepsilon}$  we define a projection

$$\begin{array}{ccc} \tilde{\pi} : \ \overline{D^3} & \to & \mathrm{SO}(3) \\ & \underline{n} & \mapsto & \exp(\pi\underline{\varepsilon} \cdot P_4 \circ \mathrm{inv}(\underline{n})) \ , \end{array} \tag{13}$$

from the closed unit ball in 3 dimensions to SO(3). Then the following diagram commutes and  $(S^3, SO(3), \pi)$  is a universal covering.

Regarding  $\mathbb{R}^3$  as linear subspace of  $\mathbb{R}^4$  we denote the canonical embedding by  $\mathcal{E}: \mathbb{R}^3 \to \mathbb{R}^4$ , i.e.

$$P_4 \circ \mathcal{E} = \mathrm{id}_{\mathbb{R}^3} . \tag{14}$$

Definition 3.1. The lift of a 3-tensor field  $\underline{A}_{\ell}$  of order  $\ell$  to  $\mathbb{R}^4$  shall be denoted by  $\underline{A}_{\ell}^{\uparrow}$ . Using Cartesian coordinates we define

$$A^\uparrow_{\mu_1\ldots\mu_\ell} = P_{4\nu_1\mu_1}\cdots P_{4\nu_\ell\mu_\ell}A_{\nu_1\ldots\nu_\ell} \ .$$

Now a term like  $\underline{\mathbf{n}} \cdot (\nabla_{\mathbf{x}}\underline{\mathbf{v}})^{\uparrow} \cdot \underline{\mathbf{n}} = P_4(\underline{\mathbf{n}}) \cdot (\nabla_{\mathbf{x}}\underline{\mathbf{v}}) \cdot P_4(\underline{\mathbf{n}})$  has a well defined meaning without adding or neglecting any information and we can use the *same* methods in the constitutive theory of biaxial liquid crystals as before in the case of nematics.

3.2. APPLICATION: VISCOSITY COEFFICIENTS. For simplicity we specialize in this paragraph to nematic liquid crystals described by  $CS_1'$  (this is only a brief sketch of the necessary calculations, for a detailed treatment see [8]). The crucial quantity describing viscous properties of liquid crystals is the friction-pressure tensor  $\underline{p}$ . Choosing a non-equilibrium state space on  $CS_1'$   $\mathcal{Z} = \{\underline{n}, \underline{N}, \underline{D}, \nabla_x \cdot \underline{\nu}\}$  with

$$\underline{\nu}(\underline{x},\underline{n},t) = \underline{\nu}(\underline{x},t) \qquad \qquad \omega = \frac{1}{2}\nabla_{x} \times \underline{\nu}$$
 (15)

$$\underline{\underline{D}} = \frac{1}{2} \left( \nabla_{\mathbf{x}} \underline{\nu} + (\nabla_{\mathbf{x}} \underline{\nu})^{\top} \right) - \nabla_{\mathbf{x}} \cdot \underline{\mathbf{x}} \underline{\mathbf{1}} \qquad \underline{\mathbf{N}} = \underline{\dot{\mathbf{n}}} - \underline{\omega} \times \underline{\mathbf{n}}$$
 (16)

and decomposing  $p_{\mu\nu}$  in its isotropic, skew-symmetric and symmetric traceless parts the general form linear in derivatives of  $\underline{\nu}$  is [14]

$$\overline{p_{\nu\mu}} = -2\eta^{\text{ord}} D_{\nu\mu} - 2\tilde{\eta}_{1}^{\text{ord}} \overline{n_{\nu}n_{\lambda}} D_{\lambda\mu} \overline{n_{\nu}n_{\lambda}} D_{\lambda\mu} - 2\tilde{\eta}_{2}^{\text{ord}} \overline{n_{\nu}N_{\mu}} 
- 2\tilde{\eta}_{3}^{\text{ord}} \overline{n_{\nu}n_{\mu}} \overline{n_{\lambda}n_{\kappa}} D_{\lambda\kappa} - \zeta_{2}^{\text{ord}} \overline{n_{\nu}n_{\mu}} \nabla_{\lambda}\nu_{\lambda} ,$$
(17)

$$(p_{\nu\mu})^{\alpha} = \gamma_1^{\text{ord}} (n_{\nu} N_{\mu})^{\alpha} + \gamma_2^{\text{ord}} (\overline{n_{\nu} n_{\lambda}} D_{\lambda\mu})^{\alpha} ,$$
 (18)

$$\frac{1}{3}p_{\lambda\lambda} = -\eta_V^{\text{ord}} \nabla_{\lambda} \nu_{\lambda} - \kappa^{\text{ord}} n_{\lambda} n_{\kappa} D_{\lambda\kappa} . \tag{19}$$

Measured viscosities are derived in flow experiments with constrained geometry. One example are the so-called Miesowicz viscosities measured in a plane Couette flow (simple shear flow) under the influence of a strong magnetic field. The magnetic field is used to overcome the influence of the flow on the mean orientation of the molecules, described by a "director"  $\underline{\mathbf{d}}$ . Thus it is possible to distinguish three main viscosities according to the orientation of the director  $\mathbf{d}$  with respect to the flow field

 $\eta_1$ : <u>d</u> parallel to the flow direction

 $\eta_2$ : <u>d</u> parallel to the "gradient" direction

 $\eta_3$ : d orthogonal to flow and gradient direction.

These are the Miesowicz viscosities, they can be expressed as linear combinations of the viscosity coefficients introduced in Eqs. (17 - 19).

Integrating Eqs. (17 – 19) over  $S^2$  and taking into account the special flow geometry we obtain for an ODF  $f_1$  with symmetry axis  $\underline{\mathbf{d}}$  (i.e.  $f_1(\underline{\mathbf{x}},\underline{\mathbf{n}},\mathbf{t}) = \tilde{f}_1(\underline{\mathbf{x}},|\underline{\mathbf{n}}\cdot\underline{\mathbf{d}}|,\mathbf{t})$ )

$$\begin{split} \eta_1 &= \eta^{\mathrm{ord}} + \frac{1}{6} S_2 \tilde{\eta}_1^{\mathrm{ord}} + \frac{1}{2} S_2 \tilde{\eta}_2^{\mathrm{ord}} + \frac{2}{15} \left( 1 + \frac{5}{7} S_2 - \frac{12}{7} S_4 \right) \tilde{\eta}_3^{\mathrm{ord}} \\ &+ \frac{1}{12} \left( 2 + S_2 \right) \gamma_1^{\mathrm{ord}} + \frac{1}{4} S_2 \gamma_2^{\mathrm{ord}} \;, \end{split} \tag{20}$$

$$\begin{split} \eta_2 &= \eta^{\mathrm{ord}} + \frac{1}{6} S_2 \tilde{\eta}_1^{\mathrm{ord}} - \frac{1}{2} S_2 \tilde{\eta}_2^{\mathrm{ord}} + \frac{2}{15} \left( 1 + \frac{5}{7} S_2 - \frac{12}{7} S_4 \right) \tilde{\eta}_3^{\mathrm{ord}} \\ &+ \frac{1}{12} \left( 2 + S_2 \right) \gamma_1^{\mathrm{ord}} - \frac{1}{4} S_2 \gamma_2^{\mathrm{ord}} \;, \end{split} \tag{21}$$

$$\begin{split} \eta_3 &= \eta^{\mathrm{ord}} - \frac{1}{3} S_2 \tilde{\eta}_1^{\mathrm{ord}} + \frac{2}{15} \left( 1 - \frac{10}{7} S_2 + \frac{3}{7} S_4 \right) \tilde{\eta}_3^{\mathrm{ord}} \\ &+ \frac{1}{6} \left( 1 - S_2 \right) \gamma_1^{\mathrm{ord}} \ . \end{split} \tag{22}$$

Here,  $S_2$  and  $S_4$  denote the averages of the second and fourth Legendre polynomials with respect to  $f_1$ .  $S_2$  is usually referred to as Maier-Saupe order parameter. In an ordered state with complete alignment we have  $S_2 = S_4 = 1$  and Eqs. (20 - 22) reduce to Eqs. (17 - 19).

After having chosen a suitable model for the "ordered" viscosities (as the affine transformation model [10, 11, 1, 2]) these viscosities can be compared with experimentally obtained values. Such a comparison can be found in [8].

### Conclusions

The description of liquid crystals within a mesoscopic model containing the orientation as additional variable allows a systematic treatment both on the levels of balance equations and constitutive relations. Physical fields are obtained by averaging the mesoscopic quantities with an orientation distribution function. There the introduction of universal coverings is advantageous and allows a uniform formalism suitable for uni- and biaxial particles. Symmetries of the ODF (as in the case of nematic liquid crystals of uniaxial molecules) simplify the resulting constitutive laws on  $\mathbb{R}^3$  and include order parameters in a natural way.

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