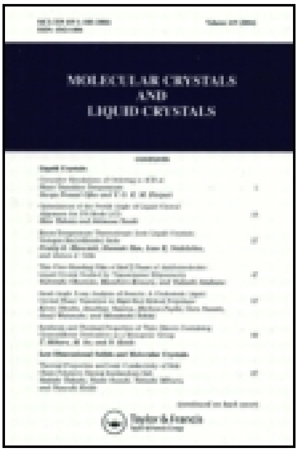

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BALANCE LAWS AND CONSTITUTIVE EQUATIONS OF MICROSCOPIC RIGID BODIES: A MODEL FOR BIAXIAL LIQUID CRYSTALS

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Abstract A reasonably general model of a liquid crystal is achieved by a continuum consisting of microscopic rigid bodies. Within this model it is possible to deal with chiral molecules as well as simple rod-like particles forming a conventional nematic liquid crystal. The configuration space of a rigid body is the rotation group SO(3); the configuration space of a "nematic" is - with regard to the head-tail symmetry - the projective plane P(2). By replacing both manifolds by their universal coverings (S^3 and S^2, resp.) the internal symmetry of the fluid can be represented by a generalized (non-normalized) director. Within this mathematical framework mesoscopic balance equations are formulated which are applicable in the biaxial case of chiral molecules and in the uniaxial case of rod-like particles. Finally it is shown how constitutive equations can be derived by calculating averages of mesoscopic quantities with respect to an orientation distribution function which characterizes the orientational order of the liquid crystal.

INTRODUCTION

When Reinitzer and Lehmann examined the first liquid crystal in 1888 they observed an internal symmetry under rotations with rotation angle π. The optical effect of this symmetry – visible under a polarization microscope – gave a whole class of materials its name: The "nematic" liquid crystals had been discovered. The mathematical picture used to model the symmetry is to introduce a pair of unit vectors $\{\underline{n}, -\underline{n}\}$ ("director") to describe the orientational alignment of molecules in a volume element.

Nowadays the number of nematic substances has dramatically increased, but in addition new materials have been synthetized[1] which form liquid crystalline phases with slightly different symmetries. Especially chiral substances (e.g. ferro-electric liquid crystals) or discotic liquid crystals might no longer be sufficiently described by a single director but call for a "biaxial" description.

There are different approaches in literature to establish a theory of such biaxial molecules. The first (and most frequently used) one is the introduction of a second director $\underline{m}^2$ to express the break of symmetry but there are also more complicated theories using Lie groups (e.g. $SU(3)$)^3 to describe biaxiality. The way used in the following is to classify the orientation of single molecules by rotation matrices and to reduce the mathematical model to a formal (mesoscopic) director theory. However, the resulting director will be four-dimensional or, alternatively, we can choose a non-normalized 3-vector. A general advantage of the presented approach will be the close analogy to the well developed theory of uniaxial liquid crystals. It should be noted that the phrase "biaxial liquid crystal" is used in the following to refer to phases of biaxial particle and does not necessarily imply a lack of symmetry of the orientational distribution function of the particles.

CONFIGURATION SPACES

At first, the mathematical model of nematic liquid crystals is recalled.

The experimentally observed symmetry of nematics – often called head-tail symmetry – leads to the choice of the two-dimensional projective plane P^2 as configuration space with respect to orientation. Because of technical difficulties (e.g. P^2 is not orientable) the unit sphere S^2 is usually substituted for P^2 and the orientation within a nematic liquid crystal is described by unit vectors $\underline{n} \in \mathrm{S}^2 \subset \mathbb{R}^3$ instead of pairs of unit vectors $[n] := \{\underline{n}, -\underline{n}\} \in \mathrm{P}^2 \equiv \mathrm{S}^2/\mathbb{Z}^2$. Head-tail symmetry requires invariance of all physical quantities under the transformation $\underline{n} \rightarrow -\underline{n}$. Thus the following topological procedure was used:

1. In accordance with experimental evidence a configuration $\mathcal{G}$ is chosen.

2. The space $\mathcal{G}$ is replaced by its universal covering $< \tilde{\mathcal{G}}, \mathcal{G}, \pi >$, $\tilde{\mathcal{G}}$ denoting the total space and $\pi : \tilde{\mathcal{G}} \rightarrow \mathcal{G}$ denoting the (fibre bundle) projection .

3. All physical quantities defined on $\mathcal{G}$ are "lifted" to $\tilde{\mathcal{G}}$. The lift introduces a natural symmetry of the constitutive functions.

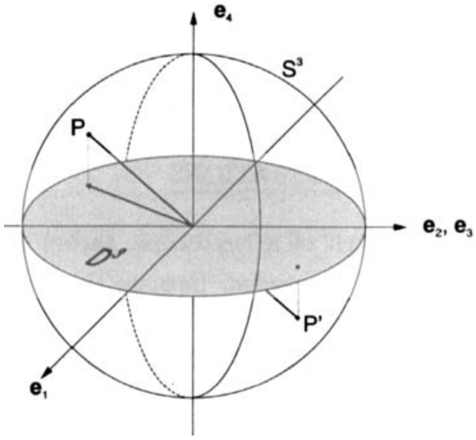

Figure 1: Substitution of S^3 for SO(3). Antipodes P and P' are projected onto the same rotation matrix, represented by points of the ball D^3

For a continuum of microscopic rigid bodies the situation is quite similar: The configuration space is the group SO(3) [4] and again it is worth looking if the universal covering of SO(3) provides a simpler model space than SO(3) itself. In fact, the universal covering of SO(3) is given by S^3, the three-dimensional unit sphere and it is simpler to differentiate and integrate on S^3. Keeping in mind that a rotation in $\mathbb{R}^3$ is determined by an angle and an axis of the rotation, it is sometimes convenient to think of SO(3) as the three-dimensional ball D^3 with radius π (maximum angle) with antipodes on its boundary identified: $\mathrm{SO}(3) \equiv D^3 \cup S^2/\mathbb{Z}^2$ – the rotation angle is obtained from $\underline{n}' \in D^3$ by $|\underline{n}'|$, the axis is represented by $\underline{n}'/|\underline{n}'|$ for $\underline{n} \neq \underline{0}$, the null vector represents the identity.
Thus we use the model described below:

The fibre bundle $< S^3, \mathrm{SO}(3), \pi >$ with projection

$$\begin{aligned} \pi : S^3 &\rightarrow \mathrm{SO}(3) \qquad (1)\\ \underline{n} &\mapsto \exp\left(\pi \underline{\underline{\epsilon}} \cdot \underline{P}(\underline{n})\right) \\ P : S^3 &\rightarrow D^3 \qquad (2)\\ \underline{n} &\mapsto \mathrm{sgn}(\underline{n} \cdot \underline{e}_4)\,(\underline{n} - \underline{n} \cdot \underline{e}_4 \underline{e}_4)) \end{aligned}$$

is substituted for SO(3). $\{\underline{e}_i\}_{i=1,2,3,4}$ is the Euclidean basis of $\mathbb{R}^4$, and $\underline{\underline{\epsilon}}$ is the Levi-Civitá tensor. Here, we used the identification of the Lie algebra so(3) with the skew-symmetric 3×3 matrices, written down by help of the $\underline{\underline{\epsilon}}$-tensor, to construct the bundle projection π. The natural symmetry of this model is obtained by the physical equivalence of all elements in $\pi^{-1}(\underline{\underline{Q}}) = \{\underline{n}, -\underline{n}\}$ for given $\underline{\underline{Q}} \in \mathrm{SO}(3)$.

Thus we are able to use an orientation distribution function

$$f : \mathbb{R}^3 \times S^3 \times \mathbb{R} \rightarrow \mathbb{R}^+ \qquad (3)$$

$$f(\underline{x}, \underline{n}, t) = f(\underline{x}, -\underline{n}, t) \qquad (4)$$

$$\int_{S^3} f(\underline{x}, \underline{n}, t)\, d^3n = 1 \qquad (5)$$

very similar to that one used in the context of nematics to describe orientational order of biaxial liquid crystal phases. The only difference between both models is the increase of dimension in case of biaxial molecules in comparison to the nematic model.

BALANCE EQUATIONS

In the spirit of a mesoscopic theory of liquid crystals[5, 6, 7, 8] orientational balance equations are needed. Because the transport theorems (6) on S^2 and S^3

$$\frac{d}{dt}\int_{\mathcal{W}(t)} X(\underline{x},\underline{n},t)\, d^k n\, d^3x = \tag{6}$$
$$\int_{\mathcal{W}(t)} \left\{ \frac{\partial}{\partial t} X(\underline{x},\underline{n},t) + \nabla_x \cdot (\underline{v}(\underline{x},\underline{n},t) X(\underline{x},\underline{n},t)) + \nabla_n \cdot (\underline{u}(\underline{x},\underline{n},t) X(\underline{x},\underline{n},t)) \right\} d^k n\, d^3 x$$

are very similar ($\underline{v}$ and $\underline{u}$ are the velocities of the time dependent set $\mathcal{W}(t) \subset \mathbb{R}^3 \times \mathrm{S}^k$ with respect to position and orientation changes, respectively, $k \in \{2,3\}$) we obtain the orientational balance equations of microscopic rigid bodies by replacing two-dimensional covariant derivatives and tangential vector fields in the nematic balances with their three-dimensional counterparts on S^3. Hence the general case of an orientational balance equation is formulated as (the argument $(\underline{x},\underline{n},t)$ is suppressed)

$$\frac{\partial}{\partial t} X + \nabla_x \cdot (\underline{v} X + \underline{J}^x_X) + \nabla_n \cdot (\underline{u} X + \underline{J}^n_X) = \mathrm{Prod}(X) \tag{7}$$

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

Mass
$X = \rho(\underline{x},\underline{n},t)$ mass density per orientation
$\underline{J}^x_X = \underline{0}$
$\underline{J}^n_X = \underline{0}$
$\mathrm{Prod}(X) = 0$

Momentum
$X = \rho(\underline{x},\underline{n},t)\underline{v}(\underline{x},\underline{n},t)$ momentum density per orientation
$\underline{J}^x_X = -\underline{\underline{t}}^\top(\underline{x},\underline{n},t)$ (transposed) pressure tensor per orientation
$\underline{J}^n_X = -\underline{\underline{T}}^\top(\underline{x},\underline{n},t)$ (transposed) pressure tensor per orientation on S^3
$\mathrm{Prod}(X) = \rho(\underline{x},\underline{n},t)\underline{k}(\underline{x},\underline{n},t)$ external force density per orientation

Angular Momentum

$X = \rho(\underline{x},\underline{n},t)\left(\underline{x}\times\underline{v}(\underline{x},\underline{n},t) + \underline{\underline{\Theta}}\cdot\underline{u}(\underline{x},\underline{n},t)\right)$

angular momentum density per orientation, $\underline{\underline{\Theta}}$ tensor of inertia

$\underline{J}^{x}_{X} = \underline{\underline{t}}^{\top}(\underline{x},\underline{n},t)\times\underline{x} + \underline{\underline{\pi}}^{\top}(\underline{x},\underline{n},t)$

torque density due to stresses and couple stresses

$\underline{J}^{n}_{X} = \underline{\underline{T}}^{\top}(\underline{x},\underline{n},t)\times\underline{x} + \underline{\underline{\Pi}}^{\top}(\underline{x},\underline{n},t)$

torque density due to stresses and couple stresses on S^3

$\mathrm{Prod}(X) = \rho(\underline{x},\underline{n},t)\underline{x}\times\underline{k}(\underline{x},\underline{n},t) + \underline{m}(\underline{x},\underline{n},t)$

torque density due to external forces and couple forces

Energy

$X = \rho(\underline{x},\underline{n},t)\left(\frac{1}{2}\underline{v}^2(\underline{x},\underline{n},t) + \frac{1}{2}\underline{\underline{\Theta}} : \underline{u}\underline{u} + e(\underline{x},\underline{n},t)\right)$

angular energy density per orientation, $e(\underline{x},\underline{n},t)$ internal energy per orientation

$\underline{J}^{x}_{X} = -\underline{v}(\underline{x},\underline{n},t)\cdot\underline{\underline{t}}(\underline{x},\underline{n},t) - \underline{u}(\underline{x},\underline{n},t)\cdot\underline{\underline{\pi}}(\underline{x},\underline{n},t) - \underline{q}(\underline{x},\underline{n},t)$

energy flux density due to stresses and couple stresses and heat flux density $\underline{q}(\underline{x},\underline{n},t)$

$\underline{J}^{n}_{X} = -\underline{v}(\underline{x},\underline{n},t)\cdot\underline{\underline{T}}(\underline{x},\underline{n},t) - \underline{u}(\underline{x},\underline{n},t)\cdot\underline{\underline{\Pi}}(\underline{x},\underline{n},t) - \underline{Q}(\underline{x},\underline{n},t)$

their counterparts on S^3

$\mathrm{Prod}(X) = \rho(\underline{x},\underline{n},t)\underline{k}(\underline{x},\underline{n},t)\cdot\underline{v}(\underline{x},\underline{n},t) + \underline{m}(\underline{x},\underline{n},t)\cdot\underline{u}(\underline{x},\underline{n},t) + r(\underline{x},\underline{n},t)$

power due to external forces and couple forces and radiation supply $r(\underline{x},\underline{n},t)$

"Orientation"

$X = f(\underline{x},\underline{n},t)$ orientation distribution function

$\underline{J}^{x}_{X} = \underline{0}$

$\underline{J}^{n}_{X} = \underline{0}$

$\mathrm{Prod}(X) = -f(\underline{x},\underline{n},t)\left(\frac{\partial}{\partial t} + \underline{v}(\underline{x},\underline{n},t)\cdot\nabla_{x}\right)\log\rho(\underline{x},t)$

Here,

$$\rho(\underline{x},\underline{n},t) =: f(\underline{x},\underline{n},t)\rho(\underline{x},t) \tag{8}$$

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.

The balance equations mentioned so far include vector fields on $\mathbb{R}^3$ – such as $\underline{v}(\underline{x},\underline{n},t)$ – as well as tangential vector fields on S^3, e.g. $\underline{u}(\underline{x},\underline{n},t)$, together with covariant derivatives. By embedding S^3 in $\mathbb{R}^4$ we can regard $\underline{n}$ and $\underline{u}(\underline{x},\underline{n},t)$ as 4-vectors which simplifies the calculation of the derivatives but leads to equations mixing 3- and 4-vector fields. However, an uniform description is often advantageous when constitutive equations are needed.

REMARKS ON CONSTITUTIVE EQUATIONS

A way to construct constitutive functions is the use of representation theorems for a given state space $\mathcal{Z}$. In general, $\mathcal{Z}$ will contain tensors of different orders, but all tensor fields are formulated on $\mathbb{R}^n$ with fixed n. From this point of view we have to lift all tensor fields of the previous section to $\mathbb{R}^4$.

The definition of the projection π in Eq. (1) contains a projection $P:\ S^3 \subset \mathbb{R}^4 \to D^3 \subset \mathbb{R}^3$. Its definition (2) can be easily extended to $\mathbb{R}^4$ and we obtain

$$\begin{aligned} \underline{\underline{P}}:\ \mathbb{R}^4 &\to \mathbb{R}^3 \\ \underline{n} &\mapsto \operatorname{sgn}(\underline{n}\cdot\underline{e}_4)\,(\underline{n}-\underline{n}\cdot\underline{e}_4\underline{e}_4))\ . \end{aligned} \tag{9}$$

We can now define the "lift" by $\underline{\underline{P}}^{\top}:\ \mathbb{R}^3 \to \mathbb{R}^4$ and

$$\underline{\underline{P}}\cdot\underline{\underline{P}}^{\top} = \mathrm{id} \tag{10}$$

is valid. For any 3-vector field $\underline{A}'$ we define the lifted vector field $\underline{A}$ by

$$\underline{A} := \underline{\underline{P}}^{\top}\cdot\underline{A}'\ . \tag{11}$$

A tensor field $\underline{\underline{B}}'$ is lifted analogously

$$\underline{\underline{B}} := \underline{\underline{P}}^{\top}\cdot\underline{\underline{B}}'\cdot\underline{\underline{P}}\ . \tag{12}$$

Thus it is possible to lead back the constitutive theory of biaxial liquid crystals to a (formal) Ericksen-Leslie theory in four dimensions.

NEMATICS AS UNIAXIAL LIMIT OF MICROSCOPIC RIGID BODIES

If biaxial molecules in a volume element are aligned in such a way that only the distribution of one major axis is of importance and the other axes are uniformly distributed the orientation distribution function f possesses a symmetry which results in a phase of (effectively) uniaxial particles.

Let $\underline{\underline{Q}}' \in \mathrm{SO}(3)$ be a rotation matrix which belongs to a configuration of a molecule with major axis aligned parallel to $\underline{n}' \in S^2 \subset \mathbb{R}^3$. Then the symmetry mentioned above is expressed by

$$f(\underline{x},\underline{n},t) = \text{const} \quad \text{for all } \underline{n} \text{ with } \pi(\underline{n})\cdot\underline{n}' = \underline{n}'\ . \tag{13}$$

Now we can define an equivalence relation by

$$\underline{n}_1 \sim \underline{n}_2 \quad \Leftrightarrow \quad \pi(\underline{n}_1) \cdot \underline{n}' = \pi(\underline{n}_2) \cdot \underline{n}' = \underline{n}' \tag{14}$$

and it is easy to verify that the equivalence classes $[\underline{n}']$ ($\underline{n}' \in \mathrm{S}^2$) are given by lifts of the 1-parameter subgroups of SO(3)

$$[\underline{n}'] = \pi^{-1}\left(\left\{\exp(\xi \underline{\underline{\underline{\epsilon}}} \cdot \underline{n}') | \xi \in (-\pi, \pi]\right\}\right) \equiv \pi^{-1}(\,\mathrm{S}^1)\ . \tag{15}$$

Thus we have obtained

$$\mathrm{S}^2 \equiv \mathrm{S}^3/\pi^{-1}(\mathrm{S}^1) \equiv \mathrm{SO}(3)/\mathrm{S}^1 \tag{16}$$

and indeed the configuration space of a nematic liquid crystal is recovered.

CONCLUSIONS

Liquid crystals of biaxial molecules can be described by a theory using 4-vectors $\underline{n} \in \mathrm{S}^3$ as microscopic directors. Hence many calculations concerning biaxial particles can be led back to nematic liquid crystals. The form of orientational balance equations is identical for biaxial and nematic liquid crystals. Both of them result in macroscopic balance equations of a Cosserat continuum. By lifting three-dimensional vector or tensor fields of physical importance (e.g. flow field $\underline{v}(\underline{x}, t)$ or $\nabla\underline{v}(\underline{x}, t)$) to S^3 constitutive equations can be formulated analogously to the well known examples of Ericksen-Leslie theory. The lifting procedure provides a natural symmetry of the constitutive equations and of the orientational distribution function describing the alignment of the molecules. Finally the case of nematic liquid crystals can be recovered as a limiting case of the general biaxial model.

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