

# ALIGNMENT TENSOR DYNAMICS INDUCED BY THE MESOSCOPIC BALANCE OF THE ORIENTATION DISTRIBUTION FUNCTION

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**Abstract.** Starting out with the mesoscopic balance equation of the orientation distribution function (ODF) of an orientable Cosserat continuum, we eliminate the orientation change velocity by the mesoscopic spin balance. We obtain balance equations of the second moment of the ODF – the alignment tensor of second order – and an expression for the orientation change velocity. The remaining balance equation of the orientation distribution is of the Fokker–Planck type.

**Key words:** liquid crystals, orientation distribution function, balance equations.

## 1. INTRODUCTION

Liquid crystals are phases showing an orientational order of molecules which are of elongated or plane shape, so that an orientational order of them can be defined. Besides the orientational order, an additional spatial order of the centres of mass of the molecules is possible, which causes a large variety of liquid crystal phases, such as nematic, smectic, cholesteric, and a lot of other phases of different structures. Because of thermal fluctuations the molecules are not totally aligned, but they have a certain distribution around a “mean orientation” which can be described by a normalized *macroscopic director field*. The name “macroscopic” originates from the fact that the macroscopic director field belongs to all molecules of a volume element, whereas a special molecule may be aligned differently. Thus it is necessary to introduce a *microscopic director* which describes the alignment of a single molecule and which is different from the local macroscopic director. Since the microscopic director is defined on a molecular level, it is not a macroscopic

field, but a so-called *mesoscopic variable*. Here “mesoscopic” means that the level of description is finer than the macroscopic level, but that no microscopic concept, such as molecular interaction or potentials, is used.

## 2. MESOSCOPIC CONCEPT

As discussed above, a liquid crystal is composed of differently orientated particles, for which we presuppose that their orientation can be described individually by the direction of their microscopic directors. Thus, the microscopic director  $\mathbf{n}$  is defined as a unit vector pointing in the temporary direction of a needle-shaped rigid particle, or, if the particle is of plane shape, the microscopic director is perpendicular to the particle. Because the microscopic director is normalized,

$$\mathbf{n}^2 = 1, \quad \mathbf{n} \in S^2, \quad (1)$$

the microscopic alignment has two degrees of freedom. Since the particles may rotate, the microscopic director changes in time and we define the microscopic orientation change velocity  $\mathbf{u}$  by

$$\mathbf{u} := \frac{d}{dt}\mathbf{n}, \quad \text{with } \mathbf{u} \cdot \mathbf{n} = 0. \quad (2)$$

If we consider molecules of a volume element, the alignment of their axes is specified by a distribution function  $f$  on the unit sphere, called the *orientation distribution function* (ODF). The ODF describes the density generated by intersection points on the unit sphere  $S^2$  between the molecule axes and the sphere. As, in general, the alignment of the molecules is a function of position and time, the ODF is defined on a six-dimensional space  $S^2 \times \mathbb{R}^3 \times \mathbb{R}^1$

$$f(\mathbf{n}, \mathbf{x}, t) \equiv f(\cdot), \quad (\cdot) \equiv (\mathbf{n}, \mathbf{x}, t) \in S^2 \times \mathbb{R}^3 \times \mathbb{R}^1. \quad (3)$$

The five-dimensional subspace  $S^2 \times \mathbb{R}^3$ , consisting of the orientation- and the position-part, is called the *nematic space*. As there are always two points of intersection opposite to each other on the  $S^2$ , the ODF shows the so-called head-tail symmetry

$$f(-\mathbf{n}, \mathbf{x}, t) = f(\mathbf{n}, \mathbf{x}, t). \quad (4)$$

By this ODF a mesoscopic classification of different types of orientation, so-called *phases*, becomes possible.

### 3. ORIENTATIONAL BALANCES

In the nematic space  $\Omega$  each particle is represented by its five coordinates  $(\mathbf{x}, \mathbf{n})$  at the time  $t$ . We define the field in the nematic space as the field of orientational mass density which is defined by

$$\rho(\cdot) := \rho(\mathbf{x}, t)f(\cdot), \quad (5)$$

where  $\rho(\mathbf{x}, t)$  is the macroscopic mass density

$$\rho(\mathbf{x}, t) := \int_{S^2} \rho(\cdot) d^2n \quad (6)$$

of the considered one-component liquid crystal,  $\rho(\cdot)$  describes the mass density of the molecules having the orientation  $\mathbf{n}$  at the position  $\mathbf{x}$  at the time  $t$ . In this interpretation  $\mathbf{v}(\cdot)$  is the material velocity of these molecules. Other such orientational fields (i.e., fields defined in the nematic space) are, e.g., the external acceleration  $\mathbf{k}(\cdot)$ , the stress tensor  $\mathbf{T}(\cdot)$ , and the heat flux density  $\mathbf{q}(\cdot)$ .

These orientational fields satisfy local balance equations which are defined on the nematic space and which are therefore denoted as *local orientational balances* [1]. The general shape of these balances is

$$\begin{aligned} \frac{\partial}{\partial t} \{ \rho(\cdot) [\phi(\cdot)\mathbf{v}(\cdot) + \Phi(\cdot)] \} + \nabla_x \cdot \{ \rho(\cdot)\mathbf{v}(\cdot) [\phi(\cdot)\mathbf{v}(\cdot) + \Phi(\cdot)] \} \\ + \nabla_n \cdot \{ \rho(\cdot)\mathbf{u}(\cdot) [\phi(\cdot)\mathbf{v}(\cdot) + \Phi(\cdot)] \} \\ = \rho(\cdot)\mathbf{K}(\cdot) + \nabla_x \cdot \mathbf{S}(\cdot). \end{aligned} \quad (7)$$

For the special balances we obtain the following identities [2]:

*mass*

$$\phi(\cdot) \equiv 0, \quad \Phi(\cdot) \equiv 1, \quad (8)$$

$$\mathbf{K}(\cdot) \equiv \mathbf{0}, \quad \mathbf{S}(\cdot) \equiv \mathbf{0}; \quad (9)$$

*momentum*

$$\phi(\cdot) \equiv 1, \quad \Phi(\cdot) \equiv \mathbf{0}, \quad (10)$$

$$\mathbf{K}(\cdot) \equiv \mathbf{k}(\cdot) = \text{external acceleration}, \quad (11)$$

$$\mathbf{S}(\cdot) \equiv \mathbf{T}^T(\cdot) = \text{transposed stress tensor}; \quad (12)$$

*angular momentum*

$$\phi(\cdot) \equiv \mathbf{x} \times, \quad \Phi(\cdot) \equiv I\mathbf{n} \times \mathbf{u}(\cdot), \quad (13)$$

$$\mathbf{K}(\cdot) \equiv \mathbf{x} \times \mathbf{k}(\cdot) + \mathbf{n} \times \mathbf{g}(\cdot), \quad (14)$$

$$\mathbf{S}(\cdot) \equiv [\mathbf{x} \times \mathbf{T}(\cdot)]^T + \mathbf{n} \times \boldsymbol{\pi}(\cdot); \quad (15)$$

total energy

$$\phi(\cdot) \equiv (1/2)\mathbf{v}(\cdot), \Phi(\cdot) \equiv (1/2)I[\mathbf{n} \times \mathbf{u}(\cdot)]^2 + \varepsilon(\cdot), \quad (16)$$

$$\mathbf{K}(\cdot) \equiv \mathbf{k}(\cdot) \cdot \mathbf{v}(\cdot) + \mathbf{g}(\cdot) \cdot \mathbf{u}(\cdot) + r(\cdot), \quad (17)$$

$$\mathbf{S}(\cdot) \equiv \mathbf{v}(\cdot) \cdot \mathbf{T}(\cdot) + \mathbf{u}(\cdot) \cdot \boldsymbol{\pi}(\cdot) - \mathbf{q}(\cdot). \quad (18)$$

Here  $\mathbf{u}(\cdot)$  is the field of the orientational change velocity,  $\mathbf{g}(\cdot)$  the orientational couple force density,  $\boldsymbol{\pi}(\cdot)$  the orientational couple stress tensor,  $\varepsilon(\cdot)$  the density of orientational internal energy, and  $r(\cdot)$  the orientational radiation supply,  $I$  is the constant moment of inertia of a needle-shaped molecule.

Besides the local balances of mass, momentum, angular momentum, and total energy, we obtain from the balance of momentum by subtracting the balance of angular momentum the *orientational spin balance*:

$$\mathbf{s}(\cdot) : = I\mathbf{n} \times \mathbf{u}(\cdot) = \text{orientational spin density}, \quad (19)$$

$$\begin{aligned} \frac{\partial}{\partial t} [\rho(\cdot)\mathbf{s}(\cdot)] + \nabla_x \cdot [\rho(\cdot)\mathbf{v}(\cdot)\mathbf{s}(\cdot) - (\mathbf{n} \times \boldsymbol{\pi}(\cdot))^T] \\ + \nabla_n \cdot [\rho(\cdot)\mathbf{u}(\cdot)\mathbf{s}(\cdot)] = \boldsymbol{\varepsilon} : \mathbf{T}(\cdot) + \rho(\cdot)\mathbf{n} \times \mathbf{g}(\cdot) \end{aligned} \quad (20)$$

( $\boldsymbol{\varepsilon}$  is the Levi-Civita tensor). The right-hand side of this equation indicates that one part of the spin supply is caused by the antisymmetric part of the stress tensor.

According to the definition of the orientational mass density (5), we obtain from the orientational mass balance an additional balance of the ODF by inserting the definition of  $f(\cdot)$ :

$$\begin{aligned} \frac{\partial}{\partial t} f(\cdot) + \nabla_x \cdot [\mathbf{v}(\cdot)f(\cdot)] + \nabla_n \cdot [\mathbf{u}(\cdot)f(\cdot)] \\ = f(\cdot)\nabla_x \cdot \mathbf{v}(\cdot). \end{aligned} \quad (21)$$

If we introduce the macroscopic fields by their mesoscopic definitions, the orientational balance equations are transformed into the balances for micropolar media by integrating over the unit sphere [3].

#### 4. APPROXIMATIONS

We now introduce some approximations: The orientational velocity does not depend on the orientation, that means, it is equal to the barycentric velocity which is assumed to be incompressible

$$1. \quad \mathbf{v}(\cdot) \equiv \mathbf{v}(\mathbf{x}, t), \quad (22)$$

$$2. \quad \nabla_x \cdot \mathbf{v}(\mathbf{x}, t) = 0. \quad (23)$$

We are interested in stationary and uniform states

$$3. \quad \frac{\partial}{\partial t} \mathbf{s}(\cdot) = \mathbf{0}, \quad (24)$$

$$4. \quad \nabla_x \equiv \mathbf{0}, \quad \text{except } \nabla_x \mathbf{v}(\cdot) = \text{const.} \quad (25)$$

There are no external forces

$$5. \quad \mathbf{g} = \mathbf{0}. \quad (26)$$

If we consider these approximations and the balance of mass (5), the orientational spin balance (20) results in

$$\rho(\cdot) \mathbf{u}(\cdot) \cdot \nabla_n \mathbf{s}(\cdot) = \boldsymbol{\varepsilon} : \mathbf{T}(\cdot). \quad (27)$$

## 5. CONSTITUTIVE EQUATIONS

Taking into account that  $\nabla_n$  is the covariant derivative

$$\nabla_n := \partial_n - \mathbf{n}(\mathbf{n} \cdot \partial_n), \quad (28)$$

we can now prove

### Proposition 1.

$$\mathbf{u}(\cdot) \cdot \nabla_n \mathbf{s}(\cdot) = I \mathbf{n} \times (\mathbf{u}(\cdot) \cdot \nabla_n) \mathbf{u}(\cdot). \quad (29)$$

Henceforth, for convenience we will suppress the argument  $(\cdot)$  denoting the nematic space. Using Proposition 1, we obtain from (27)

$$\nabla_n \mathbf{s} = \mathbf{0} \longrightarrow \boldsymbol{\varepsilon} : \mathbf{T} = \mathbf{0}, \quad (30)$$

$$\mathbf{n} \cdot \boldsymbol{\varepsilon} : \mathbf{T} = 0. \quad (31)$$

From (30) we see that  $\boldsymbol{\varepsilon} : \mathbf{T}$  is homogeneous in  $\nabla_n \mathbf{s}$ . Thus we obtain the general constitutive equation

$$\boldsymbol{\varepsilon} : \mathbf{T} = \rho \mathbf{G} \cdot \nabla_n \mathbf{s}. \quad (32)$$

Applying (31), we can prove the following

### Proposition 2.

$$\mathbf{G} \cdot \nabla_n \mathbf{s} \cdot \mathbf{n} = 0 \longrightarrow \mathbf{G} = \mathbf{u} + \beta \mathbf{n}. \quad (33)$$

Thus, we have proved that the orientation change velocity  $\mathbf{u}$  is in accordance with the spin balance (27) if the constitutive equation (32) holds. Equation (33) is a constraint for  $\mathbf{u}$

$$\mathbf{u} = \mathbf{G} - \beta \mathbf{n}, \quad (34)$$

which we have to transform into an objective formulation. Introducing the local angular velocity  $\boldsymbol{\omega}$  of the observer with respect to the material, we can define

$$\mathbf{N} := \mathbf{u} - \boldsymbol{\omega} \times \mathbf{n}, \quad \mathbf{H} := \mathbf{G} - \boldsymbol{\omega} \times \mathbf{n}, \quad (35)$$

and (34) becomes

$$\mathbf{N} = \mathbf{H} - \beta \mathbf{n} = \mathbf{N}^* = \mathbf{H}^* - \beta \mathbf{n}. \quad (36)$$

Here  $*$  denotes the quantities belonging to another arbitrary observer, and  $\beta$ ,  $\mathbf{n}$ ,  $\mathbf{N}$ , and  $\mathbf{H}$  are objective quantities.

We now have to define what the domain of the constitutive equations is. This domain is called the *state space*. Here, in the case of liquid crystals, the state space is chosen so that it consists of mesoscopic and macroscopic parts

$$\mathcal{Z} = (\mathcal{Z}^{\text{meso}}, \mathcal{Z}^{\text{macro}}), \quad (37)$$

$$\mathcal{Z}^{\text{meso}} = (\mathbf{n}, \mathbf{N}(\cdot), \nabla_n \ln \rho(\cdot)), \quad (38)$$

$$\mathcal{Z}^{\text{macro}} = (\rho(\mathbf{x}, t), T(\mathbf{x}, t), \overline{\nabla_x \mathbf{v}}(\mathbf{x}, t), \mathbf{a}(\mathbf{x}, t)). \quad (39)$$

According to the approximations (22) and (25),  $\overline{\nabla_x \mathbf{v}}(\mathbf{x}, t)$  (the symbol  $\overline{\dots}$  refers to the symmetric traceless part of a tensor) is a constant, and  $\mathbf{a}(\mathbf{x}, t)$  is the macroscopic field of the *alignment tensor*

$$\mathbf{a}(\mathbf{x}, t) := \int_{S^2} f(\cdot) \overline{\mathbf{nn}} d^2n. \quad (40)$$

We now presuppose that the constitutive equation for  $\mathbf{H}$  is linear in all variables of  $\mathcal{Z}$ , except  $\rho(\cdot)$  and  $\mathbf{n}$ . Thus, we obtain with 7 functions  $\alpha_j$ ,  $j = 1, \dots, 5$ , and  $\gamma_k$ ,  $k = 1, 2$ ,

$$\begin{aligned} \mathbf{H}(\cdot) &= \alpha_1 \mathbf{n} + \alpha_2 \mathbf{N} + \alpha_3 \nabla_n \ln \rho + \alpha_4 \overline{\nabla_x \mathbf{v}} \cdot \mathbf{n} \\ &+ \gamma_1 \mathbf{n} \cdot \overline{\nabla_x \mathbf{v}} \cdot \mathbf{nn} + \gamma_2 \mathbf{n} \cdot \mathbf{a} \cdot \mathbf{nn} + \alpha_5 \mathbf{a} \cdot \mathbf{n}. \end{aligned} \quad (41)$$

Taking this linear ansatz for  $\mathbf{H}$  into account, we can calculate the orientation change velocity  $\mathbf{u}$  by (36) and (35)<sub>1</sub>.

**Proposition 3.** *The orientation change velocity is*

$$\mathbf{u} = \boldsymbol{\omega} \times \mathbf{n} + \beta_3 \nabla_n \ln \rho + \mathcal{P} \cdot \{\beta_4 \overline{\nabla_x \mathbf{v}} + \beta_5 \mathbf{a}\} \cdot \mathbf{n}, \quad (42)$$

$$\mathcal{P} := \mathbf{1} - \mathbf{nn}, \quad (43)$$

$$\beta_j := \alpha_j / [1 - \alpha_2], \quad j = 3, 4, 5. \quad (44)$$

By use of (28) we obtain for the divergence of the orientation change velocity

**Proposition 4.**

$$\nabla_n \cdot \mathbf{u} = \beta_3 \nabla_n \cdot \nabla_n \ln \rho - 3\mathbf{n} \cdot \{\beta_4 \overline{\nabla_x \mathbf{v}} + \beta_5 \mathbf{a}\} \cdot \mathbf{n}. \quad (45)$$

## 6. ALIGNMENT TENSOR DYNAMICS

Taking the approximations (22) and (23) into account, we get the differential equation of the ODF (21)

$$\frac{\partial}{\partial t} f(\cdot) + \mathbf{v} \cdot \nabla_x f(\cdot) + f(\cdot) \nabla_n \cdot \mathbf{u}(\cdot) + \mathbf{u}(\cdot) \cdot \nabla_n f(\cdot) = 0. \quad (46)$$

Introducing the total time derivative

$$\frac{D}{Dt} f := \frac{\partial}{\partial t} f + \mathbf{v} \cdot \nabla_x f, \quad (47)$$

and inserting the relations of (42) and (45), we can prove the following statement using a straightforward calculation (note that the terms quadratic in  $\nabla_n f$  cancel without any additional assumptions).

**Proposition 5.** *The ODF satisfies a Fokker–Planck type equation*

$$\begin{aligned} \frac{D}{Dt} f + \beta_3 \nabla_n \cdot \nabla_n f - 3f \mathbf{n} \cdot \{ \beta_4 \overline{\nabla_x \mathbf{v}} + \beta_5 \mathbf{a} \} \cdot \mathbf{n} \\ + [ \boldsymbol{\omega} \times \mathbf{n} + \mathbf{n} \cdot \{ \beta_4 \overline{\nabla_x \mathbf{v}} + \beta_5 \mathbf{a} \} ] \cdot \nabla_n f = 0. \end{aligned} \quad (48)$$

Multiplication of (48) by  $\overline{\mathbf{n}\mathbf{n}}$ , integration over the microscopic directors, and introduction of the abbreviation

$$\mathbf{A} := \{ \beta_4 \overline{\nabla_x \mathbf{v}} + \beta_5 \mathbf{a} \} \quad (49)$$

yields

**Proposition 6.**

$$\frac{D}{Dt} \mathbf{a} - 2 \overline{\boldsymbol{\omega} \times \mathbf{a}} = 6\beta_3 \mathbf{a} + \frac{2}{5} \mathbf{A} + \frac{6}{7} \overline{\mathbf{A} \cdot \mathbf{a}} - 2\mathbf{A} : \mathbf{a}_4 \quad (50)$$

with

$$\mathbf{a}_4 := \int_{S^2} \overline{\mathbf{n}\mathbf{n}\mathbf{n}\mathbf{n}} f(\cdot) d^2 n.$$

## 7. CONCLUSIONS

Starting with the orientational balances, we derived a Fokker–Planck type relaxation equation for the orientation distribution function. The special form of this equation was obtained under several constraints mentioned in Section 3

with the most restrictive conditions being concerned with the absence of external (electric or magnetic) fields and the uniformity of the system. Nevertheless, such a situation frequently occurs when rheological properties of freely flowing nematics are examined. For this case we developed a relaxation equation for the alignment tensor  $\mathbf{a}$  in the form of Eq. (50). The left-hand side of (50) is Jaumann's time derivative (convected and corotational time derivative) of the alignment tensor  $\mathbf{a}$ , the right-hand side is the alignment production containing nonlinear terms in  $\mathbf{a}$  (responsible for the phase transition *isotropic-nematic*) and some terms which are due to the flow field. Thus, taking into account the restrictions of Section 3, it is possible to extract an evolution equation for the order parameters of nematic liquid crystals from the mesoscopic balances of spin and mass and to obtain an explicit expression for the alignment production.

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### ORIENTATSIOONI JAOTUSFUNKTSIOONI MESOSKOOPILISEST TASAKAALUST MÕJUTATUD REASTUSTENSORI DÜNAAMIKA

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Cosserat' tüüpi pideva keskkonna iseloomustamiseks on kasutatud orientatsiooni jaotusfunktsiooni mesoskoopilise tasakaalu võrrandit, millest on elimineeritud orientatsiooni muutuse kiirus mesoskoopilise spinni tasakaalu arvestades. Tulemus kujutab endast teist järku reastustensori teise momendi tasakaaluvõrrandeid, millele lisandub orientatsiooni muutuse kiiruse võrrand. Orientatsiooni jaotusfunktsiooni tasakaal on seejuures Fokkeri-Plancki tüüpi.