Mesoscopically derived orientation dynamics of liquid crystals

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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 Fokker-Planck type.

Introduction

Liquid crystals are phases showing an orientational order of molecules which are of elongated or of plane shape, so that an orientational order of them can be defined. Besides the orientational order an additional spatial order of the centers of mass of the molecules is possible which causes a great variety of liquid crystal phases, such as nematics, smectics, cholesterics and a lot of other phases of various 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. So it is obvious to introduce a *microscopic director* which describes the alignment of a single molecule and which is different from the local macroscopic director. Because 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 is, but that no microscopic concept such as molecular interaction or potentials are used.

The degree of orientational order and the direction of the mean orientation can be influenced by a flow field or by electromagnetic fields [1]. Also shear induced phase transitions from the isotropic to liquid crystalline phases have been observed [2]. The theoretical work on the dynamics of the orientational order is based on a Fokker-Planck-equation for the orientation distribution function [3, 4, 5]. This equation is motivated by physical arguments, but has not been derived from basic principles. On the other hand the mesoscopic balance of mass yields an equation of motion for the orientation distribution function (ODF). However this is not a closed equation, as long as the field of orientation change velocity is not given. The aim of the present paper is the derivation of a closed equation for the ODF and for their first anisotropic moment, the alignment tensor, on the grounds of the mesoscopic theory.

1 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

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

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

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

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 *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. Because in general the alignment of the molecules is a function of position and time the ODF is defined on a six-dimensional space $S^2 \times \mathbb{R}^3 \times \mathbb{R}^1$

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

The 5-dimensional subspace $S^2 \times \mathbb{R}^3$ consisting of the orientation- and of the position-part is called *nematic space*. Because there are always two points of intersection opposite to each other on the S^2 the ODF shows the so-called head-tail symmetry

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

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

2 Orientational Balances

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

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

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

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

of the considered one-component liquid crystal. $\rho(\cdot)$ describes the mass density of those molecules having the orientation $\mathbf{n}$ at position $\mathbf{x}$ at time t. In this interpretation $\mathbf{v}(\cdot)$ is the material velocity of these molecules. Other such orientational fields (i.e. fields defined on 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* [6]. The general shape of these balances is

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

We obtain for the special balances the following identities [7]:
Mass

$$\phi(\cdot) \equiv 0, \quad \mathbf{\Phi}(\cdot) \ \equiv \ 1, \tag{8}$$

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

Momentum

$$\begin{aligned}
\phi(\cdot) \equiv 1,\ \boldsymbol{\Phi}(\cdot) &\equiv \mathbf{0}, && (10)\\
\mathbf{K}(\cdot) &\equiv \mathbf{k}(\cdot) = \text{external acceleration}, && (11)\\
\mathbf{S}(\cdot) &\equiv \mathbf{T}^\top(\cdot) = \text{transposed stress tensor}, && (12)
\end{aligned}$$

Angular Momentum

$$\begin{aligned}
\phi(\cdot) \equiv \mathbf{x}\times, \boldsymbol{\Phi}(\cdot) &\equiv I\mathbf{n} \times \mathbf{u}(\cdot), && (13)\\
\mathbf{K}(\cdot) &\equiv \mathbf{x} \times \mathbf{k}(\cdot) + \mathbf{n} \times \mathbf{g}(\cdot), && (14)\\
\mathbf{S}(\cdot) &\equiv [\mathbf{x} \times \mathbf{T}(\cdot)]^\top + \mathbf{n} \times \boldsymbol{\pi}(\cdot), && (15)
\end{aligned}$$

Total Energy

$$\begin{aligned}
\phi(\cdot) \equiv (1/2)\boldsymbol{\nu}(\cdot), \boldsymbol{\Phi}(\cdot) &\equiv (1/2)I[\mathbf{n} \times \mathbf{u}(\cdot)]^2 + \varepsilon(\cdot), && (16)\\
\mathbf{K}(\cdot) &\equiv \mathbf{k}(\cdot) \cdot \boldsymbol{\nu}(\cdot) + \mathbf{g}(\cdot) \cdot \mathbf{u}(\cdot) + r(\cdot), && (17)\\
\mathbf{S}(\cdot) &\equiv \boldsymbol{\nu}(\cdot) \cdot \mathbf{T}(\cdot) + \mathbf{u}(\cdot) \cdot \boldsymbol{\pi}(\cdot) - \mathbf{q}(\cdot). && (18)
\end{aligned}$$

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.

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*:

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

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

($\boldsymbol{\varepsilon}$ 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 $f(\cdot)$:

$$\frac{\partial}{\partial t}f(\cdot) + \nabla_x \cdot [\boldsymbol{\nu}(\cdot)f(\cdot)] + \nabla_n \cdot [\mathbf{u}(\cdot)f(\cdot)] = f(\cdot)\nabla_x \cdot \boldsymbol{\nu}(\cdot). \qquad (21)$$

Introducing the macroscopic fields by their mesoscopic definitions the orientational balance equations transform into the balances for micropolar media by integrating over the unit sphere [8].

3 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

$$1: \qquad \mathbf{v}(\cdot) \equiv \mathbf{v}(\mathbf{x}, t), \tag{22}$$

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

We are interested in stationary and uniform states

$$3: \qquad \frac{\partial}{\partial t}\mathbf{s}(\cdot) = 0, \tag{24}$$

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

There are no external forces

$$5: \qquad \mathbf{g} = 0. \tag{26}$$

Taking these approximations and the balance of mass (5) into account the orientational spin balance (20) results in

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

4 Constitutive Equations

Taking into account that ∇_n is the covariant derivative

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

we now can prove the

Proposition 1

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

Henceforth for convenience we will suppress the argument $(\cdot)$ denoting the nematic space. By use of proposition 1 we obtain from (27)

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

$$\mathbf{n} \cdot \boldsymbol{\varepsilon} : \mathbf{T} = 0. \tag{31}$$

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

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

By use of (31) we can prove the following

Proposition 2

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

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

$$\mathbf{u} = \mathbf{G} - \beta \mathbf{n} \tag{34}$$

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

$$\mathbf{N} := \mathbf{u} - \boldsymbol{\omega} \times \mathbf{n}, \qquad \mathbf{H} := \mathbf{G} - \boldsymbol{\omega} \times \mathbf{n}, \tag{35}$$

and (34) becomes

$$\mathbf{N} = \mathbf{H} - \beta \mathbf{n} = \mathbf{N}^* = \mathbf{H}^* - \beta \mathbf{n}. \tag{36}$$

Here the * denotes the quantities belonging to an other arbitrary observer, and β, $\mathbf{n}$, $\mathbf{N}$, 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 case of liquid crystals, the state space is chosen in such a way, that it consists of a mesoscopic and of a macroscopic part

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

$$\mathcal{Z}^{macro} = (\rho(\mathbf{x},t), T(\mathbf{x},t), \overline{\nabla_x \mathbf{v}}\ (\mathbf{x},t), \mathbf{a}(\mathbf{x},t)). \tag{38}$$

$$\mathcal{Z}^{meso} = (\mathbf{n}, \mathbf{N}(\cdot), \nabla_n \ln \rho(\cdot)), \tag{39}$$

According to the approximations (22) and (25) $\overline{\nabla_x \mathbf{v}}\,(\mathbf{x},t)$ (the symbol $\overline{\ldots}$ 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*

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

$\mathcal{Z}^{macro}$ is the simplest state space one can chose for macroscopic constitutive equations for liquid crystals under shear. It is supplemented by the mesoscopic variables $\mathcal{Z}^{meso}$, because we are dealing with mesoscopic constitutive equations.

We now presuppose that the constitutive equation for $\mathbf{H}$ is linear in all variables of $\mathcal{Z}$, except of ρ, T, $\mathbf{a}$ and $\mathbf{n}$. Thus we obtain with 7 functions α_j j = 1,...,5, and γ_k k = 1, 2,

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

Taking into account this linear ansatz for $\mathbf{H}$ we can calculate the orientation change velocity $\mathbf{u}$ by (36) and $(35)_1$

Proposition 3 *The orientation change velocity is*

$$\begin{aligned} \mathbf{u} &= \boldsymbol{\omega} \times \mathbf{n} + \beta_3 \nabla_n \ln \rho + \mathcal{P} \cdot \{\beta_4 \overline{\nabla_x \boldsymbol{v}} + \beta_5 \mathbf{a}\} \cdot \mathbf{n}, && (42) \\ \mathcal{P} &:= \mathbf{1} - \mathbf{nn}, && (43) \\ \beta_j &:= \alpha_j / [1 - \alpha_2], \qquad j = 3,4,5. && (44) \end{aligned}$$

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

Proposition 4

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

5 Alignment Tensor Dynamics

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

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

Introducing the total time derivative

$$\frac{D}{Dt} f := \frac{\partial}{\partial t} f + \boldsymbol{v} \cdot \nabla_x f \tag{47}$$

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

Proposition 5 *The ODF satisfies a Fokker-Planck type equation*

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

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

$$\mathbf{A} := \{\beta_4 \overline{\nabla_x \boldsymbol{v}} + \beta_5 \mathbf{a}\} \tag{49}$$

and the fourth rank alignment tensor

$$\underline{\mathbf{a}}_4(\mathbf{x}, t) := \int_{S^2} f(\cdot) \, \overline{\mathbf{nnnn}} \, d^2 n \tag{50}$$

yields the

Proposition 6

$$\frac{\mathrm{D}}{\mathrm{Dt}}\mathbf{a} - 2\,\overline{\boldsymbol{\omega} \times \mathbf{a}} = 6\beta_3\mathbf{a} + \frac{2}{5}\mathbf{A} + \frac{6}{7}\,\overline{\mathbf{A} \cdot \mathbf{a}} - 2\,\overline{\mathbf{A} \cdot \underline{\mathbf{a}}_4}\ . \tag{51}$$

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 constrains 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. (51). The left hand side of (51) 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 non-linear terms in $\mathbf{a}$ and $\underline{\mathbf{a}}_4$ (responsible for the phase transition *isotropic-nematic*) and some terms which are due to the flow field. Thus it is possible – taking into account the restrictions of section 3 – 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.
The application of this equation to shear experiments on liquid crystals in planar Couette flow is in preparation. There also the coefficients β_3, β_4, and β_5 are identified as experimentally measurable coefficients. In order to deal with experiments, where a rotating electric field is applied to a thin free standing film [9], the electric field and spatial gradient of the variables have to be included into the state space, which is left for a future work. Finally the approach presented here can also be extended to liquid crystalline mixtures of different chemical components.

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Appendix

The proofs to all propositions run as follows:

Proposition 1

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

Proof. Since $\mathbf{s} = \mathrm{I}\mathbf{n} \times \mathbf{u}$ we obtain

$$\begin{aligned} \mathbf{u} \cdot \nabla_n \mathbf{s} =& \mathrm{I}\mathbf{u} \cdot \nabla_n \Big(\mathbf{n} \times \mathbf{u}\Big) = \mathrm{I}\mathbf{u} \cdot \mathbf{1} \times \mathbf{u} + \mathrm{I}\mathbf{n} \times \Big(\mathbf{u} \cdot \nabla_n \mathbf{u}\Big) \\ =& \mathrm{I}\mathbf{u} \times \mathbf{u} + \mathrm{I}\mathbf{n} \times \Big(\mathbf{u} \cdot \nabla_n \mathbf{u}\Big) = \mathrm{I}\mathbf{n} \times \Big(\mathbf{u} \cdot \nabla_n \mathbf{u}\Big) \end{aligned}$$

□

Proposition 2 *For every solution* $\mathbf{s}$ *of the stationary spin balance the implication*

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

holds true.

Proof. The stationary spin balance (27) reads

$$\rho \mathbf{u} \cdot \nabla_n \mathbf{s} = \boldsymbol{\varepsilon} : \mathbf{T} = \mathbf{G} \cdot \nabla_n \mathbf{s} \ .$$

Representing $\mathbf{G}$ by

$$\mathbf{G} = \alpha \mathbf{u} + \beta \mathbf{n} + \gamma \mathbf{n} \times \mathbf{u}$$

we obtain

$$\begin{aligned} \mathbf{G} \cdot \nabla_n =& \alpha \mathbf{u} \cdot \nabla_n \mathbf{s} + \beta \cdot \nabla_n \mathbf{s} + \gamma \mathbf{n} \times \mathbf{u} \cdot \nabla_n \mathbf{s} \\ =& \alpha \mathbf{G} \cdot \nabla_n \mathbf{s} + 0 + \gamma \mathbf{n} \times \mathbf{u} \cdot \nabla_n \Big(\mathrm{I}\mathbf{n} \times \mathbf{u}\Big) \end{aligned}$$

or

$$(1 - \alpha)\mathbf{G} \cdot \nabla_n \mathbf{s} = \mathrm{I}\gamma \mathbf{n} \times \mathbf{u} \cdot \nabla_n (\mathbf{n} \times \mathbf{u}) \ .$$

Thus we infer from $\mathbf{G} \cdot \nabla_n \mathbf{s} \cdot \mathbf{n} = 0$ the relation

$$0 = (1 - \alpha)\mathbf{G} \cdot (\nabla_n \mathbf{s}) \cdot \mathbf{n} = \mathrm{I}\gamma \mathbf{n} \times \mathbf{u} \cdot \nabla_n (\mathbf{n} \times \mathbf{u}) \cdot \mathbf{n} \ .$$

Taking into account that

$$\nabla_n (\mathbf{n} \times \mathbf{u}) \cdot \mathbf{n} = \nabla_n (\mathbf{n} \times \mathbf{u} \cdot \mathbf{n}) - (\nabla_n \mathbf{n}) \cdot \mathbf{n} \times \mathbf{u} = -\mathbf{n} \times \mathbf{u}$$

holds true we arrive at

$$I\gamma\mathbf{n}\times\mathbf{u}\cdot(-\mathbf{n}\times\mathbf{u}) = -I\gamma|\mathbf{n}\times\mathbf{u}|^2 = -I\gamma|\mathbf{u}|^2 = 0\ .$$

Since $\mathbf{u}$ is in general not the null vector we conclude $\gamma = 0$ and consequently $\alpha = 1$. Thus the final relation is

$$\mathbf{G} = \mathbf{u} + \beta\mathbf{n}$$

with some $\beta \in \mathbb{R}$. □

Proposition 3 *The orientation change velocity is*

$$\begin{aligned}
\mathbf{u} &= \boldsymbol{\omega}\times\mathbf{n} + \beta_3\nabla_n \ln\rho + \mathcal{P}\cdot\{\beta_4 \overline{\nabla_x\mathbf{v}} + \beta_5\mathbf{a}\}\cdot\mathbf{n},\\
\mathcal{P} &:= \mathbf{1} - \mathbf{nn},\\
\beta_j &:= \alpha_j/[1-\alpha_2], \qquad j = 3,4,5.
\end{aligned}$$

Proof. The representation theorem (41) together with the Eqs. (33) and (35) yields

$$\begin{aligned}
\mathbf{H} =&\alpha_1\mathbf{n} + \alpha_2\mathbf{N} + \alpha_3\nabla_n\ln\rho + \alpha_4 \overline{\nabla_x\mathbf{v}}\cdot\mathbf{n} +\\
&+\gamma_1\mathbf{n}\cdot \overline{\nabla_x\mathbf{v}}\cdot\mathbf{nn} + \gamma_2\mathbf{n}\cdot\mathbf{a}\cdot\mathbf{nn} + \alpha_5\mathbf{a}\cdot\mathbf{n}\\
=&\mathbf{G} - \omega\times\mathbf{n} = \mathbf{u} + \beta\mathbf{n} - \omega\times\mathbf{n}\ .
\end{aligned}$$

Thus we obtain for $\mathbf{u}$

$$\begin{aligned}
\Big(1-\alpha_2\Big)\mathbf{u} =& \Big(1-\alpha_2\Big)\omega\times\mathbf{n} + \Big(\alpha_1 - \beta\Big)\mathbf{n} + \alpha_3\nabla_n\ln\rho +\\
&+\alpha_4 \overline{\nabla_x\mathbf{v}}\cdot\mathbf{n} + \gamma_1\mathbf{n}\cdot \overline{\nabla_x\mathbf{v}}\cdot\mathbf{nn} + \gamma_2\mathbf{n}\cdot\mathbf{a}\cdot\mathbf{nn} + \alpha_5\mathbf{a}\cdot\mathbf{n}\ .
\end{aligned}$$

Since $\mathbf{u}$ is perpendicular to $\mathbf{n}$ we know in addition

$$\begin{aligned}
0 =& \mathbf{n}\cdot\mathbf{u} =\\
&= \alpha_1 - \beta + \alpha_4\mathbf{nn} : \overline{\nabla_x\mathbf{v}} + \gamma_1\mathbf{nn} : \overline{\nabla_x\mathbf{v}} + \gamma_2\mathbf{nn} : \mathbf{a} + \alpha_5\mathbf{nn} : \mathbf{a}
\end{aligned}$$

and conclude

$$\alpha_1 = \beta, \qquad \alpha_4 = -\gamma_1, \qquad \alpha_5 = -\gamma_2\ .$$

Finally, we arrive at

$$\begin{aligned}
\mathbf{u} =& \frac{1}{(1-\alpha_2)}\Big((1-\alpha_2)\omega\times\mathbf{n} + \alpha_3\nabla_n\ln\rho +\\
&+\alpha_4(\mathbf{1}-\mathbf{nn})\cdot \overline{\nabla_x\mathbf{v}}\cdot\mathbf{n} + \alpha_5(\mathbf{1}-\mathbf{nn})\cdot\mathbf{a}\cdot\mathbf{n}\Big)\\
=&\omega\times\mathbf{n} + \beta_3\nabla_n\ln\rho + \mathcal{P}\cdot\Big\{\beta_4 \overline{\nabla_x\mathbf{v}} + \beta_5\mathbf{a}\Big\}\cdot\mathbf{n}\ .
\end{aligned}$$

□

Proposition 4

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

Proof. From the previous proposition we know already the velocity field $\mathbf{u}$. Its divergence is calculated straight forward:

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

which yields (note that $\beta_4 \overline{\nabla_x \mathbf{v}} + \beta_5 \mathbf{a}$ shall not depend on $\mathbf{n}$)

$$\begin{aligned}\nabla_n \cdot \mathbf{u} = & \underbrace{\nabla_n \cdot (\omega \times \mathbf{n})}_{=0} + \beta_3 \nabla_n \cdot \nabla_n \ln \rho + \\ & + \nabla_n \cdot \Big[\big(\mathbf{1} - \mathbf{nn}\big) \cdot \{\beta_4 \overline{\nabla_x \mathbf{v}} + \beta_5 \mathbf{a}\} \cdot \mathbf{n}\Big] \\ = & \beta_3 \nabla_n \cdot \nabla_n \ln \rho + \underbrace{\{\beta_4 \overline{\nabla_x \mathbf{v}} + \beta_5 \mathbf{a}\} : \mathbf{1}}_{=0} - \\ & - 3\mathbf{n} \cdot \{\beta_4 \overline{\nabla_x \mathbf{v}} + \beta_5 \mathbf{a}\} \cdot \mathbf{n} - \underbrace{\mathbf{n} \cdot \nabla_n \Big[\mathbf{n} \cdot \{\ldots\} \cdot \mathbf{n}\Big]}_{=0}\end{aligned}$$

since $\beta_4 \overline{\nabla_x \mathbf{v}} + \beta_5 \mathbf{a}$ is a traceless tensor and $\mathbf{n} \cdot \nabla_n \ldots$ vanishes due to the definition of the covariant derivative – yielding the wanted relation. □

Proposition 5 *The ODF satisfies a Fokker-Planck type equation*

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

Proof. Inserting the expressions for $\nabla_n \cdot \mathbf{u}$ of proposition 4 and 3 (for $\mathbf{u}$ itself) into the Fokker-Planck equation (46) and using the definition of the material time derivative Eq. (47) yields the proposed relation. □

Proposition 6

$$\frac{D}{Dt}\mathbf{a} - 2\,\overline{\boldsymbol{\omega} \times \mathbf{a}} = 6\beta_3 \mathbf{a} + \frac{2}{5}\mathbf{A} + \frac{6}{7}\,\overline{\mathbf{A} \cdot \mathbf{a}} - 2\,\overline{\mathbf{A} \cdot \underline{\mathbf{a}}_4}\ .$$

Proof. We multiply Eq. (48) with $\overline{\mathbf{nn}}$ and integrate over all orientations. Additionally, we use the abbreviation

$$\mathbf{A} := \beta_4 \overline{\nabla_x \mathbf{v}} + \beta_5 \mathbf{a}\ .$$

This leads to ($\Delta_n := \nabla_n \cdot \nabla_n$)

$$\frac{D}{Dt}\oint f\ \overline{\mathbf{nn}}\ d^2n + \beta_3\oint(\Delta_n f)\ \overline{\mathbf{nn}}\ d^2n - 3\mathbf{A} : \oint f\ \overline{\mathbf{nn}}\ \overline{\mathbf{nn}}\ d^2n +$$
$$+\oint \omega \times \mathbf{n} \cdot (\nabla_n f)\ \overline{\mathbf{nn}}\ d^2n + \mathbf{A} : \oint (\nabla_n f)\mathbf{n}\ \overline{\mathbf{nn}}\ d^2n = 0$$

We shall calculate each term of the equation for its own.

Obviously (from the definition of the alignment tensor) we have

$$\frac{D}{Dt}\oint f\ \overline{\mathbf{nn}}\ d^2n = \frac{D}{Dt}\mathbf{a}\ .$$

Secondly, the components of $\overline{\mathbf{nn}}$ are eigenfunctions of Δ_n[10], i.e.

$$\Delta_n\ \overline{\mathbf{nn}} = -6\ \overline{\mathbf{nn}}\ ,$$

which yields (shifting the symmetric operator Δ_n from the left to the right)

$$\beta_3\oint(\Delta_n f)\ \overline{\mathbf{nn}}\ d^2n = -6\beta_3\mathbf{a}\ .$$

For the third term we remark that for any tensor $\mathbf{B}$ the relation [10]

$$\mathbf{B} : \overline{\mathbf{nn}}\ \overline{\mathbf{nn}} = \mathbf{B} : \overline{\mathbf{nnnn}} + \frac{4}{7}\ \overline{\mathbf{B}\cdot\ \overline{\mathbf{nn}}} + \frac{2}{15}\ \overline{\mathbf{B}}$$

holds true. The fourth rank alignment tensor is defined analogously to $\mathbf{a}$ as

$$\underline{\mathbf{a}}_4 := \oint f\ \overline{\mathbf{nnnn}}\ d^2n$$

and we obtain

$$-3\mathbf{A} : \oint f\ \overline{\mathbf{nn}}\ \overline{\mathbf{nn}}\ d^2n = -3\mathbf{A} : \underline{\mathbf{a}}_4 - \frac{12}{7}\ \overline{\mathbf{A}\cdot\mathbf{a}} - \frac{2}{5}\mathbf{A}\ .$$

To deal with the fourth and fifth term we consider an expression with a traceless (symmetric or anti-symmetric) second order tensor $\mathbf{C}$

$$\mathbf{C} : \oint(\nabla_n f)\mathbf{n}\ \overline{\mathbf{nn}}\ d^2n =$$
$$= \underbrace{\oint \nabla_n \cdot \left(f\mathcal{P}\cdot\mathbf{C}\cdot\mathbf{n}\ \overline{\mathbf{nn}}\right) d^2n}_{=0} - \oint f\nabla_n \cdot \left(\mathcal{P}\cdot\mathbf{C}\cdot\mathbf{n}\ \overline{\mathbf{nn}}\right) d^2n$$
$$= -\oint f\nabla_n \cdot \left(\mathcal{P}\cdot\mathbf{C}\cdot\mathbf{n}\ \overline{\mathbf{nn}}\right) d^2n\ .$$

The term $\oint \nabla_n \cdot (\mathcal{P} \cdot \dots)\, \mathrm{d}^2 n$ vanishes due to Gauss' theorem (the boundary of S^2 is the empty set). Note, that we inserted the projector $\mathcal{P}$, since only the tangential part of $\mathbf{C}$ contributes in $\mathbf{C}^\top \cdot \nabla_n f$. A short calculation shows

$$
\begin{aligned}
\nabla_n \cdot \left(\mathcal{P} \cdot \mathbf{C} \cdot \mathbf{n}\, \overline{\mathbf{nn}} \right) &= \mathbf{C} \cdot \mathbf{nn} + \mathbf{n} \mathbf{C} \cdot \mathbf{n} - 3 \mathbf{C} : \overline{\mathbf{nn}}\, \overline{\mathbf{nn}} - 2 \mathbf{C} : \overline{\mathbf{nn}}\, \mathbf{nn} = \\
&= 2\, \overline{\mathbf{C} \cdot \overline{\mathbf{nn}}} + \frac{2}{3} \overline{\mathbf{C}} - 5 \mathbf{C} : \overline{\mathbf{nn}}\, \overline{\mathbf{nn}} = \\
&= 2\, \overline{\mathbf{C} \cdot \overline{\mathbf{nn}}} + \frac{2}{3} \overline{\mathbf{C}} - 5 \mathbf{C} : \overline{\mathbf{nnnn}} - \frac{20}{7} \overline{\mathbf{C} \cdot \overline{\mathbf{nn}}} - \frac{2}{3} \overline{\mathbf{C}} = \\
&= -\frac{6}{7} \overline{\mathbf{C} \cdot \overline{\mathbf{nn}}} - 5 \mathbf{C} : \overline{\mathbf{nnnn}} \; .
\end{aligned}
$$

Since we can write

$$
\omega \times \mathbf{n} = \Omega \cdot \mathbf{n}, \qquad \Omega := -\epsilon \cdot \omega \; ,
$$

we obtain (with $\mathcal{P} \cdot \Omega = \Omega$)

$$
\nabla_n \cdot \left(\Omega \cdot \mathbf{n}\, \overline{\mathbf{nn}} \right) = 2\, \overline{\Omega \cdot \overline{\mathbf{nn}}} = 2\, \overline{\omega \times \overline{\mathbf{nn}}}
$$

and by integration

$$
\oint \omega \times \mathbf{n} \cdot (\nabla_n f)\, \overline{\mathbf{nn}}\, \mathrm{d}^2 n = -2\, \overline{\omega \times \mathbf{a}} \; .
$$

The fifth term, finally, turns out as (replace $\mathbf{C}$ by $\mathbf{A}$)

$$
\mathbf{A} : \oint (\nabla_n f) \mathbf{n}\, \overline{\mathbf{nn}}\, \mathrm{d}^2 n = \frac{6}{7} \overline{\mathbf{A} \cdot \mathbf{a}} + 5 \mathbf{A} : \underline{\mathbf{a}}_4 \; .
$$

Thus the final equation contains $\underline{\mathbf{a}}_4$ in addition to $\mathbf{a}$ and we conclude

$$
\frac{\mathrm{D}}{\mathrm{D}t} \mathbf{a} - 2\, \overline{\omega \times \mathbf{a}} = 6 \beta_3 + \frac{2}{5} \mathbf{A} + \frac{6}{7} \overline{\mathbf{A} \cdot \mathbf{a}} - 2\, \overline{\mathbf{A} : \underline{\mathbf{a}}_4} \; .
$$

□

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