

# Nonlinear dynamics of the alignment tensor in the presence of electric fields

*Dedicated to Prof. Henryk Zorski  
on the occasion of his 70-th birthday*

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IN THE MESOSCOPIC theory field quantities are introduced, which depend not only on position and time, but also on an additional orientational variable, the microscopic director. The orientation distribution function (ODF) gives the fraction of particles of a particular orientation. An equation of motion for the second order alignment tensor, including the influence of electromagnetic fields and of spatial inhomogeneities, is derived. The starting point are the mesoscopic balance equations of mass and of angular momentum. A constitutive equation on the mesoscopic level is discussed. The two balance equations together with the constitutive equation yield an equation of motion for the ODF. This finally leads to a nonlinear partial differential equation for the alignment tensor, which involves also the fourth and the sixth moment of the ODF.

## Notations

$\rho$	mass density,
$\underline{v}$	material velocity,
$\underline{t}$	stress tensor,
$\underline{\underline{f}}$	acceleration of external forces,
$\underline{s}$	specific density of internal angular momentum,
$\underline{\underline{\Pi}}$	couple stresses,
$\underline{g}$	couple forces,
$f$	orientation distribution function.

## 1. Introduction

THE DYNAMICS of the orientational order in liquid crystals under the influence of electromagnetic fields is interesting from the practical as well as from the theoretical point of view. The most important application is the switching of the preferred orientation in liquid crystalline devices. There are other experiments, where a rotating electric field is applied to a thin free-standing film [3]. The formation of dynamical orientation patterns is observed. In these orientation patterns the orientational order is spatially inhomogeneous. This importance of spatial gradients has not been taken into account in previous papers [9, 5] and will be considered now.

The orientational order in liquid crystals is described by the orientation distribution function or approximately, by the alignment tensor of second order. The aim of the present paper is a derivation of a differential equation for the orientation distribution function (ODF) and from there, the derivation of a differential equation for the alignment tensor. The starting point is the mesoscopic theory of liquid crystals.

Up to now, the theoretical work on the dynamics of the orientational order has been based on a Fokker-Planck-equation for the orientation distribution function [6, 7, 4]. This equation is motivated by physical arguments, but has not been derived from the 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 and the higher order alignment tensors are not given. In the present paper, an equation for the ODF is derived on the grounds of the mesoscopic theory.

## 2. Mesoscopic concept

Liquid crystals consist of non-spherical particles. Here it is assumed that the particles are rotationally symmetric, with the axis of rotation called microscopic director  $\underline{n}$ . In all liquid crystalline phases the microscopic directors exhibit a long-range orientational order, but no (three-dimensional) positional order. The orientational order can be described by introducing an orientation distribution function (ODF) (see Fig. 1).

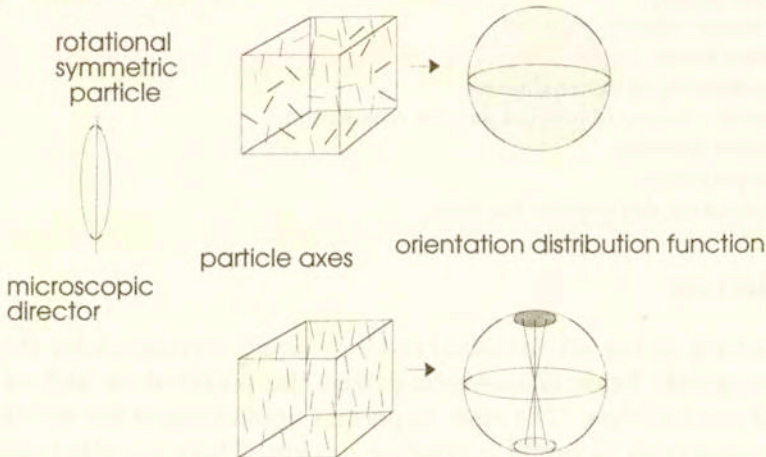


FIG. 1.

The axes of the rotationally symmetric particles are the microscopic directors. Their orientations are distributed at random in the isotropic phase, and they are more or less parallel in the liquid crystalline phases. This corresponds to

an isotropic orientation distribution function on the unit sphere, or an ODF concentrated around one direction, respectively. If the orientation distribution function is rotationally symmetric, the axis of rotational symmetry is called the macroscopic director  $\underline{d}$ .

This distribution function is defined on the so-called nematic space. The nematic space is spanned by positions  $\underline{x}$ , orientations  $\underline{n}$ , and time  $t$ .  $\underline{x}$  and  $\underline{n}$  determine the configuration of the rotationally symmetric particle. In all liquid crystalline phases the ODF is anisotropic, whereas in the isotropic phase it is a homogeneous distribution on the unit sphere. In the liquid crystalline phase the ODF can be uniaxial or biaxial. In the nematic phase it is observed experimentally in most cases to be uniaxial. For uniaxial orientation distribution functions there exists an axis of rotational symmetry called macroscopic director  $\underline{d}$ . The anisotropic moments of the ODF are called alignment tensors. They are defined as

$$(2.1) \quad a^{(k)} := \int_{S^2} f(\underline{x}, \underline{n}, t) \underbrace{\underline{n} \dots \underline{n}}_k d^2 n,$$

where  $\underbrace{\dots}_k$  denotes the symmetric irreducible part of a tensor. Because of the symmetry of the ODF  $f(-\underline{n}) = f(\underline{n})$ , only even order alignment tensors are nonzero. If the ODF is uniaxial, the alignment tensors can be expressed in terms of the order parameters  $S^{(k)}$  and the macroscopic director:

$$(2.2) \quad a^{(k)} = S^{(k)} \underbrace{\underline{d} \dots \underline{d}}_k$$

for symmetry reasons, and analogously for the even-order moments which are not traceless:

$$(2.3) \quad \int_{S^2} f(\underline{x}, \underline{n}, t) \underbrace{\underline{n} \dots \underline{n}}_k d^2 n = \sigma^{(k)} \underbrace{\underline{d} \dots \underline{d}}_k.$$

The second order parameter  $S$  is the Maier–Saupe order parameter.

The idea of the mesoscopic theory is to define fields on a higher-dimensional domain, in contrast to the Ericksen–Leslie theory, which introduces an additional macroscopic field (the macroscopic director  $\underline{d}$ ). So-called mesoscopic fields are introduced, which are fields of physical quantities defined on the nematic space, i.e. depending not only on position and time, but also on the microscopic director (the orientation). For instance, the mesoscopic mass density  $\hat{\rho}(\underline{x}, \underline{n}, t)$  is the mass density of particles of the particular orientation  $\underline{n}$ . Mesoscopic fields are denoted by a mark  $\hat{\phantom{x}}$ . Macroscopic fields are obtained from the mesoscopic ones by averaging with the ODF. The ODF gives the fraction of particles of orientation  $\underline{n}$ :

$$(2.4) \quad f(\underline{x}, \underline{n}, t) = \frac{\hat{\rho}(\underline{x}, \underline{n}, t)}{\rho(\underline{x}, t)}.$$

For the mesoscopic quantities, the balance equations have been derived [1, 2]. From the mesoscopic balance of mass one obtains for the ODF in the case of an incompressible fluid:

$$(2.5) \quad \frac{\partial f}{\partial t} + \hat{v} \cdot \nabla f + \nabla_n \cdot (\underline{u}f) = 0.$$

$\hat{v}$  is the material velocity of particles of orientation  $\underline{n}$ .  $\underline{u}$  is the orientation change velocity. It is tangential to the unit sphere.  $\nabla_n$  denotes the covariant derivative with respect to the orientation variable. The last term on the left-hand side of Eq. (2.5) is the flux in orientation space. It expresses the fact that the fraction of particles of orientation  $\underline{n}$  changes if the particles change their orientation (see Fig. 2). The orientation change velocity  $\underline{u}$  is tangential to the unit sphere.

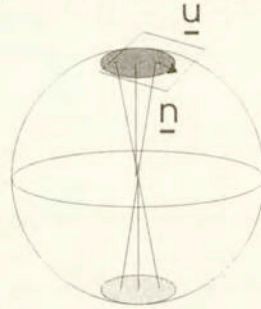


FIG. 2.

### 3. Balance of spin and simplifications

The mesoscopic balance of spin reads:

$$(3.1) \quad \frac{\partial \hat{\underline{s}}}{\partial t} + \nabla \cdot \left( \hat{\underline{v}}\hat{\underline{s}} - (\underline{n} \times \hat{\underline{\Pi}})^T \right) + \nabla_n \cdot (\hat{\underline{v}}\underline{u}\hat{\underline{s}}) = \underline{\underline{\epsilon}} : \hat{\underline{t}} + \hat{\underline{v}}\underline{n} \times \hat{\underline{g}}.$$

The symbols are:  $\hat{\underline{s}}$ : mesoscopic specific spin density,  $\hat{\underline{\Pi}}$ : couple stresses ( $\nabla \cdot \hat{\underline{\Pi}}$  are short-range forces acting on the orientation),  $\hat{\underline{t}}$ : mesoscopic stress tensor,  $\hat{\underline{g}}$ : couple forces (long-range forces acting on the orientation). Equation (3.1) shows that the antisymmetric part of the stress tensor produces the spin.

#### 3.1. Approximations

In contrast to the previous work [9], the system is not assumed to be uniform. The following simplifications are made:

1. The spin is stationary:  $\frac{\partial \hat{\underline{s}}}{\partial t} = 0.$

2. There is no material flux:  $\hat{v} \equiv 0.$  In the case of freestanding liquid crystalline films in a rotating electric field, there are experimental hints that there is no flow of material [3].

Then the spin balance (3.1) simplifies to

$$(3.2) \quad \widehat{\rho} \underline{u} \cdot \nabla_n \widehat{s} = \underline{\underline{\epsilon}} : \widehat{\underline{t}} + \widehat{\rho} \underline{n} \times \widehat{\underline{g}} + \nabla \cdot \left( \underline{n} \times \widehat{\underline{\Pi}} \right)^T ;$$

$\widehat{\underline{t}}$ ,  $\widehat{\underline{\Pi}}$  and  $\widehat{\underline{g}}$  are constitutive quantities. They depend on the variables in the state space (see below) in a material-dependent manner. From Eq. (3.2) we see that  $\underline{\underline{\epsilon}} : \widehat{\underline{t}} + \widehat{\rho} \underline{n} \times \widehat{\underline{g}} + \nabla \cdot \left( \underline{n} \times \widehat{\underline{\Pi}} \right)^T$  vanishes if  $\nabla_n \widehat{s} = 0$ . Therefore we can write down a constitutive equation of the form

$$(3.3) \quad \underline{\underline{\epsilon}} : \widehat{\underline{t}} + \widehat{\rho} \underline{n} \times \widehat{\underline{g}} + \nabla \cdot \left( \underline{n} \times \widehat{\underline{\Pi}} \right)^T = \widehat{\rho} \widehat{\underline{G}} \cdot \nabla_n \widehat{s}.$$

The orientation change velocity  $\underline{u}$  is a solution of the stationary spin balance (3.2) if

$$(3.4) \quad \underline{u} = \underline{\underline{P}} \cdot \widehat{\underline{G}}, \quad \underline{\underline{P}} = \underline{\underline{\delta}} - \underline{n} \underline{n}.$$

It has been taken into account that  $\underline{u}$  is orthogonal to  $\underline{n}$ , with the projector  $\underline{\underline{P}}$  onto the space orthogonal to  $\underline{n}$  (see Eq. (3.4)), because  $\underline{u} = \dot{\underline{n}}$ , and  $\underline{n}$  is a unit vector.

For the mesoscopic quantity  $\widehat{\underline{G}}$  we make a constitutive assumption.

#### 4. Constitutive equations

In order to make a constitutive assumption it is necessary to fix the domain of the constitutive functions, i.e. to choose the state space. The state space has to be a mesoscopic one, because we are dealing with mesoscopic constitutive functions. It also has to include spatial gradients, because orientation patterns are inhomogeneous in space. The electric field  $\underline{E}$  is also included. It is chosen here as

$$(4.1) \quad Z = \left( \widehat{\rho}, T, \underline{E}, \underline{a}, \nabla \underline{a}, \nabla \nabla \underline{a}, \frac{\nabla \widehat{\rho}}{\widehat{\rho}}, \frac{\nabla \nabla \widehat{\rho}}{\widehat{\rho}}, \frac{\nabla_n \widehat{\rho}}{\widehat{\rho}}, \underline{n} \right),$$

here  $T$  is the temperature. It includes macroscopic  $(T, \underline{E}, \underline{a}, \nabla \underline{a}, \nabla \nabla \underline{a})$  and mesoscopic variables  $\left( \widehat{\rho}, \frac{\nabla \widehat{\rho}}{\widehat{\rho}}, \frac{\nabla \nabla \widehat{\rho}}{\widehat{\rho}}, \frac{\nabla_n \widehat{\rho}}{\widehat{\rho}}, \underline{n} \right)$ . Especially the macroscopic quantity, the second order alignment tensor  $\underline{a}$ , has been included in the state space. This inclusion of an integral quantity is necessary, because the action on particles of one orientation depends not only on particles of similar orientations, but also on particles of very different orientations.

Then a representation theorem for  $\widehat{\underline{G}}$ , linear in all derivatives and quadratic in  $\underline{E}$ , gives:

$$(4.2) \quad \underline{u} = \underline{\underline{P}} \cdot \widehat{\underline{G}} = \beta_3 \nabla_n \ln \widehat{\rho} + \underline{\underline{P}} \cdot \left( \beta_5 \underline{a} + \beta_6 \underline{a} \cdot \underline{a} + \beta_9 \underline{a} \cdot \underline{n} \underline{n} \cdot \underline{a} + \beta_7 \underline{E} \underline{E} \right. \\ \left. + \beta_8 \underline{n} \underline{n} : \nabla \nabla \underline{a} + \beta_{10} \frac{\nabla \nabla \widehat{\rho}}{\widehat{\rho}} + \beta_{11} \underline{n} \cdot \nabla \nabla \underline{a} \cdot \underline{n} + \beta_{12} \underline{n} \underline{n} : \underline{a} \underline{a} \right) \cdot \underline{n}.$$

Here it has been taken into account that the orientation change velocity must be an even function of  $\underline{n}$

$$(4.3) \quad \underline{u}(-\underline{n}) = -\underline{u}(\underline{n}).$$

The coefficients  $\beta_j$  are functions of the macroscopic mass density  $\rho(\underline{x}, t)$  and the temperature  $T(\underline{x}, t)$ .

## 5. The equations of motion for the orientation distribution function and for the second order alignment tensor

The second tensorial irreducible moment of Eq. (2.5) reads

$$(5.1) \quad \frac{\partial}{\partial t} \int_{S^2} f \underline{nn} d^2n + \underline{v} \cdot \nabla \int_{S^2} f \underline{nn} d^2n + \int_{S^2} \underline{nn} \nabla_n \cdot (f \underline{u}) d^2n = 0.$$

The first two terms are derivatives of the alignment tensor. The third term is integrated by parts using the Gauss theorem on the unit sphere. The resulting equation reads:

$$(5.2) \quad \frac{\partial \underline{a}}{\partial t} + \underline{v} \cdot \nabla \underline{a} = \frac{d \underline{a}}{dt} = \int_{S^2} f \underline{u} \cdot \nabla_n (\underline{nn}) d^2n.$$

In our case we have  $\underline{v} \equiv 0$ . Then inserting Eq. (4.2) and integrating the first term by parts, we obtain:

$$(5.3) \quad \begin{aligned} \frac{\partial \underline{a}}{\partial t} = & 6\beta_3 \underline{a} + 2\beta_{10} (\nabla \nabla \cdot \underline{a})^{\text{symm}} + \left( \left( \underline{a} + \frac{1}{3} \underline{\delta} \right) \cdot \underline{A} \right)^{\text{symm}} \\ & - 2 \left( \underline{A} + \beta_{10} \nabla \nabla \right) : \int_{S^2} \underline{nnnn} f d^2n \\ & + 2 \left( \underline{\underline{B}} : \int_{S^2} \underline{nnnn} f d^2n \right)^{\text{symm}} \\ & - 2 \left( (\beta_8 + \beta_{11}) \nabla \nabla \underline{a} + \beta_{12} \underline{aa} \right) :: \int_{S^2} \underline{nnnnnn} f d^2n, \end{aligned}$$

where  $(\dots)^{\text{symm}}$  denotes the symmetric part of a tensor, and we have introduced  $(\nabla \nabla \underline{a})_{ijkl}^T := \nabla_k \nabla_l a_{ij}$ . It has been taken into account that

$$(5.4) \quad \nabla_n \underline{n} = \underline{P} = \underline{\delta} - \underline{nn},$$

and we have introduced the abbreviations

$$(5.5) \quad \underline{A} := \beta_5 \underline{a} + \beta_6 \underline{a} \cdot \underline{a} + \beta_7 \underline{EE},$$

$$(5.6) \quad \underline{\underline{B}} = \left( \beta_8 (\nabla \nabla \underline{a})^T + \beta_{11} \nabla \nabla \underline{a} + \beta_{12} \underline{aa} \right).$$

The higher moments  $\int_{S^2} \underline{nnnn} f d^2n$  and  $\int_{S^2} \underline{nnnnnn} f d^2n$  can be expressed by the second order one using the closure relation, which can be derived using the principle of maximum entropy [8]. This derivation of a closure relation will be published elsewhere.

## 6. Conclusions

Starting with the orientational balances, we have derived a Fokker–Planck type differential equation for the orientation distribution function (ODF). The special form of this equation was obtained under several assumptions mentioned in Subsec. 3.1. Because the second order alignment tensor, which is the second moment of the orientation distribution function, enters this equation, the equation for the distribution function is an integro-differential equation. Taking the second moment of this equation, a nonlinear partial differential equation for the second order alignment tensor is obtained. The orientation distribution function is hardly experimentally accessible, and the second order alignment tensor is the most important quantity from the experimental point of view. The fourth and the sixth moment of the ODF enter the equation for the second order alignment tensor. Closure relations expressing these higher moments in terms of the second one can be derived by applying the principle of maximum entropy on the phase space. The entropy is maximized under the constraints that the phase space averages of the fields of mass density, internal energy density, spin density and second order alignment tensor are prescribed at every  $\underline{x}$  and  $t$ .

In most cases the ODF is uniaxial and the alignment tensor can be expressed by the scalar order parameter  $S$  and the macroscopic director  $\underline{d}$ . For this uniaxial case, coupled differential equations for  $S$  and  $\underline{d}$  can be derived.

## Acknowledgments

Many helpful discussions with W. MUSCHIK and H. EHRENTAUT are gratefully acknowledged. The work has been supported financially by Draloric Company (D-95100 Selb), the European Union (grant ERBCHRXCT920007) and the Sfb 335 "Anisotrope Fluide".

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Received November 19, 1997.