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RESEARCH ARTICLE

Maximum Entropy Closure Relation for Higher Order Alignment and Orientation Tensors Compared to Quadratic and Hybrid Closure

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Abstract

A closure relation expresses the fourth order orientation tensor as a function of the second order one. Two well-known closure relations, the hybrid closure and the maximum entropy closure, are compared in the case of a rotation symmetric orientation distribution function. The maximum entropy closure predicts a positive fourth order parameter in the whole range of the second order parameter, whereas the hybrid closure results in negative fourth order parameters for small values of the second order one. For the maximum entropy closure quadratic fit polynomials are presented. For a general distribution without rotation symmetry, the expression for the entropy is exploited to derive an explicit form for the maximum entropy distribution. Lowest order approximation of this distribution function leads to simple closure forms for the fourth order alignment tensor and also for higher order alignment tensors.

Keywords: Orientation Tensor, Maximum Entropy Closure, Hybrid Closure

1 Introduction

Fiber reinforced materials become more and more important [1, 2] in many fields of application: In civil engineering short steel fibers are applied to improve the mechanical properties of concrete [3, 4]. Carbon fibers or glass fibers are introduced into polymers [5]. The mechanical (and eventually electrical) properties of a fiber-polymer composite depend on the orientational order of the fibers. In the case of concrete a uniform distribution of fiber orientations is often desirable. In other applications, a high elastic modulus in one direction or an electrically conducting polymer require more or less parallel orientation of the fibers. Therefore, the question of influencing the fiber orientation during the production process of a composite is of great technical interest. A reorientation of fibers is practically possible as long as the main component is in a fluid state. This is the case in fresh concrete or in a molten polymer.

Suspensions of (rigid) fibers in a low molar mass solvent play a role in the production process of most fiber composite materials. Usually, the suspension is flowing during the production process. Examples are the spinning process of carbon or glass fiber reinforced polymers or the filling of fresh fiber concrete into a formwork. During the flow process the fibers are reoriented, and the distribution of the (rigid) fiber orientations becomes anisotropic. After hardening, the fiber orientations are frozen. The material properties, like elastic modulus, heat conductivity and others are anisotropic, depending on the fiber orientation distribution. It is important to predict the influence of the flow field on the fiber orientation and consequently on the material properties.

Orientation and alignment tensors have been introduced to account for the orientational order in fiber suspensions [6, 7], liquid crystals, ferrofluids and other materials consisting of elongated particles. In all these examples the dynamics of the orientation tensor or the alignment tensor is relevant. For

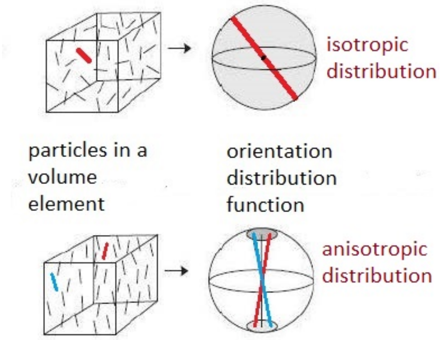


Figure 1: Orientation distribution function in case of isotropic particle orientations and in case of anisotropic particle orientations.

example, in moldflow of fiber composite materials the fibers are reorienting during the flow process of the suspension, thus determining the properties of the resulting composite. If the dynamics of the orientation or alignment tensor is derived from an orientation distribution function, in the differential equation for the lowest order tensor there is always involved a higher order tensor, which has to be eliminated by a closure relation. The predicted orientation tensor distribution in a flowing suspension - and consequently the properties of the resulting composite - depend essentially on the closure relation [8, 9, 10, 11].

1.1 Orientation distribution and orientation tensors

The orientations of the fiber particles may be described by an orientation distribution (ODF) on the unit sphere. Imagine all particles of the volume element attached with their centers of mass to the center of the unit sphere. The end points of the particles (scaled to length two) give rise to a distribution function on the unit sphere.

The orientation distribution is the probability density of finding a particle of the particular orientation $\boldsymbol{n}$, where $\boldsymbol{n}$ is a unit vector. It is homogeneous if all particle orientations are equally probable (see the first case in figure 1. This is the isotropic case. If there exists a preferred orientation, the ODF is concentrated more or less around that orientation (anisotropic case, second example in figure 1).

Since the whole distribution function is not easily measurable, the moments of the distribution function are introduced. They are macroscopic tensorial quantities. The symmetric tensors

$$\boldsymbol{A}^{(k)}(\boldsymbol{x},t) := \int_{S^2} f(\boldsymbol{x},t,\boldsymbol{n})\,\underbrace{\boldsymbol{n}\ldots\boldsymbol{n}}_{\text{k times}}\,d^2n \tag{1}$$

are denoted as orientation tensors. The set of infinitely many orientation tensors contains the same information as the ODF, but practically, only the lowest non-trivial moment, the second order tensor $\boldsymbol{A}^{(2)}(\boldsymbol{x},t) =: \boldsymbol{A} := \int_{S^2} f(\boldsymbol{x},t,\boldsymbol{n})\boldsymbol{n}\boldsymbol{n}d^2n$, is taken into account.

The alignment tensors are defined as the traceless part of the orientation tensors:

$$\boldsymbol{a}^{(k)}(\boldsymbol{x},t) := \int_{S^2} f(\boldsymbol{x},t,\boldsymbol{n})\,\underbrace{\overline{\boldsymbol{n}\ldots\boldsymbol{n}}}_{\text{k times}}\,d^2n, \tag{2}$$

where $\overline{}$ denotes the symmetric traceless part of a tensor. Contraction over any pair of indices of the alignment tensors is zero. They are a direct measure of orientational order, because all alignment tensors are zero in the isotropic phase, which is not the case for the orientation tensors. Different states of orientational order may be distinguished in terms of the second order orientation tensor or in terms of the alignment tensor. If the orientations are isotropic, the orientation tensor is proportional to the unit tensor, whereas for anisotropic distributions, it has a traceless part. If the orientation distribution is rotation symmetric, the alignment tensor is of the special form $\boldsymbol{a}^{(2)} = S^{(2)}\ \overline{\boldsymbol{dd}}$ with a scalar order parameter $S^{(2)}$ and unit vector $\boldsymbol{d}$.

In the context of liquid crystals the alignment tensors are considered usually, because they are a direct measure of the orientational order and of the anisotropic material properties. In the context of fiber suspensions, the orientation tensors are more familiar [6]. The quadratic closure relation has a very simple form in terms of the orientation tensors.

1.2 Relation between the alignment and orientation tensors

In the following, vector and tensor components refer to a Cartesian coordinate system with summation convention applied.

As the alignment tensors are defined as the traceless part of the orientation tensors, they fulfill the relations in components [12, 13]

$$a^{(2)}_{ij} = A^{(2)}_{ij} - \frac{1}{3}\delta_{ij} \tag{3}$$

$$\begin{aligned} a^{(4)}_{ijkl} &= A^{(4)}_{ijkl} - \frac{1}{7}\Big(\delta_{ij}A^{(4)}_{mmkl} + \delta_{ik}A^{(4)}_{mmjl} + \delta_{il}A^{(4)}_{mmjk} + \delta_{jk}A^{(4)}_{mmil} + \\ &\quad + \delta_{jl}A^{(4)}_{mmik} + \delta_{kl}\underbrace{A^{(4)}_{mmij}}_{=A^{(2)}_{ij}}\Big) + \frac{1}{35}\left(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right) \\ &= A^{(4)}_{ijkl} - \frac{1}{7}\Big(\delta_{ij}A^{(2)}_{kl} + \delta_{ik}A^{(2)}_{jl} + \delta_{il}A^{(2)}_{jk} + \delta_{jk}A^{(2)}_{il} + \\ &\quad + \delta_{jl}A^{(2)}_{ik} + \delta_{kl}A^{(2)}_{ij}\Big) + \frac{1}{35}\left(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right) \end{aligned} \tag{4}$$

For simplicity, we will apply the short notation for the second order tensors $\boldsymbol{a} := \boldsymbol{a}^{(2)}$ and $\boldsymbol{A} := \boldsymbol{A}^{(2)}$.

1.3 Expansion of the orientation distribution function

The traceless tensors $\overline{\boldsymbol{nn}}$ fulfill an orthogonality relation when integrated over the unit sphere [14, 13]:

$$\frac{1}{4\pi}\int_{S^2} \underbrace{\overline{n_{\mu 1}\dots n_{\mu k}}}_{\text{k times}}\ \underbrace{\overline{n_{\nu 1}\dots n_{\nu l}}}_{\text{l times}}\, d^2n = \delta_{kl}\frac{l!}{(2l+1)!!}\Delta_{\mu 1\dots\mu l,\nu 1\dots\nu l}, \tag{5}$$

where $\Delta_{\mu 1\dots\mu l,\nu 1\dots\nu l}$ is the projector on the traceless part of a tensor and $m!! = m(m-2)(m-4)\dots 1, m \in N$, odd. Especially, the integral is zero, if the order of the tensors is different $(k \neq l)$. Consequently, the alignment tensors appear as the coefficients in the expansion of the ODF with respect to the variable $\boldsymbol{n}$:

$$f(\boldsymbol{x}, t, \boldsymbol{n}) = \frac{1}{4\pi}\left(1 + \sum_{l\text{even}} \frac{(2l+1)!!}{l!} a_{\mu 1\dots\mu l}(\boldsymbol{x}, t)\ \overline{n_{\mu 1}\dots n_{\mu_l}}\right). \tag{6}$$

1.4 Dynamics of the orientation tensor

The equation of motion for the orientation tensors or for the alignment tensors results from the equation of motion for the orientation distribution function. There are many examples for a derivation in the literature [15, 16, 17, 18, 19, 7].

For fiber suspensions, a widely used equation of motion for the second order orientation tensor is the Folgar-Tucker equation [19]

$$\begin{aligned} \frac{d\boldsymbol{A}^{(\mathbf{2})}}{dt} &+ \frac{1}{2}\left(\boldsymbol{\omega}\cdot\boldsymbol{A}^{(\mathbf{2})} - \boldsymbol{A}^{(\mathbf{2})}\cdot\boldsymbol{\omega}\right) = \\ &= \frac{1}{2}\lambda\left((\nabla\boldsymbol{v})^{(\mathrm{sym})}\cdot\boldsymbol{A}^{(\mathbf{2})} + \boldsymbol{A}^{(\mathbf{2})}\cdot(\nabla\boldsymbol{v})^{(\mathrm{sym})} - 2(\nabla\boldsymbol{v}):\boldsymbol{A}^{(\mathbf{4})}\right) + \\ &\quad +2D\left(\boldsymbol{\delta} - 3\boldsymbol{A}^{(\mathbf{2})}\right) \ . \end{aligned} \tag{7}$$

$\boldsymbol{\delta}$ is the unit tensor, and $\boldsymbol{\omega}$ is the antisymmetric part of the velocity gradient. The left hand side of this equation is the co-rotating time derivative of the orientation tensor - the time derivative of an observer moving and rotating with the flow field. The terms in the second line are due to the reorienting effect of the flow field (the velocity gradient) and due to the influence of the fluid component and neighboring fibers. D and λ are material parameters, λ depending on the ratio of diameter to length of the fibers with $\lambda = 1$ for infinitely long fibers.

In the Folgar-Tucker equation, as well as in other examples of differential equations for the second moment, the fourth order orientation tensor occurs, and it has to be eliminated by a closure relation. The closure relation expresses the fourth order tensor in terms of the second order one.

2 Closure relations

A simple and widely used closure relation is the quadratic closure [20]

$$\boldsymbol{A}^{(\mathbf{4})} = \boldsymbol{A}^{(\mathbf{2})}\boldsymbol{A}^{(\mathbf{2})} \ . \tag{8}$$

Another type of closure relation discussed in the literature is the linear closure, where the fourth order orientation tensor is expressed in terms linear in the second order tensor and the unit tensor, symmetric in any pair of indices and fulfilling the relation $A_{ijll} = A_{ij}$ see f.i. [21, 22]. In three dimensions it is of the form (in Cartesian components):

$$\begin{aligned} A^{(4)}_{ijkl} &= -\frac{1}{35}\left(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right) + \\ &\quad +\frac{1}{7}\left(\delta_{ij}A_{kl} + A_{ij}\delta_{kl} + A_{ik}\delta_{jl} + A_{jl}\delta_{ik} + A_{il}\delta_{jk} + A_{jk}\delta_{il}\right) \end{aligned} \tag{9}$$

Equation (4) shows, that the linear closure (9) is equivalent to the assumption $\boldsymbol{a}^{(\mathbf{4})} = 0$.

The quadratic closure is exact in the limiting case of perfect parallel alignment of the particles, whereas the linear closure becomes exact in the limit of an isotropic distribution of fiber orientations.

The hybrid closure is the convex combination of the linear and the quadratic closure [7], thus interpolating between the two closure forms

$$\boldsymbol{A}^{(4)}_{hybrid} = f\ \boldsymbol{A}^{(4)}_{quadratic} + (1-f)\ \boldsymbol{A}^{(4)}_{linear}. \tag{10}$$

with a scalar factor f depending on the shear rate. The quadratic and linear closure are special cases of the hybrid closure, setting $f = 1$ or $f = 0$, respectively. Therefore, we will treat the hybrid closure in section 3.2 and derive the other closure forms as special cases.

For a comparison of different closure approximations with respect to alignment predictions of a flowing fiber suspension, see f.i. [23] and for an overview over closure relations discussed in the literature see [24, 25, 26, 27]. The Reduce Strain Closure (RSC) model requires the determination of fit parameters from experimental data (see f.i. [27]). The choice of the closure relation is a crucial point in the simulation of the orientation tensor in flowing fiber suspensions (see f.i. for comparison [9, 11]). The viscosity of the suspension and the elasticity tensor show both significant flow-induced anisotropies as well as a strong dependence on the closure relation [28]. In a recent work [29], the orientation distribution function has been reconstructed in a certain class from the second-order orientation tensors obtained from injection molding simulations. Based on this ODF, a new closure approximation is developed.

The elastic modulus of the resulting fiber composite depends on the fourth order orientation tensor. Calculating the elasticity tensor or the stiffness tensor of the composite from a simulation of the second order orientation tensor requires again a closure relation. For planar orientation states, as they occur in sheet molding compounds [30], a minimal invariant set of structurally differing planar fourth-order fiber orientation tensors and orientation distribution functions derived from them have been constructed [30, 31].

2.1 A closure relation derived from entropy maximization

The idea is, that the second order orientation tensor or alignment tensor is the only moment of the orientation distribution function, which is measured. Apart from the constraint on the second order tensor, the ODF is determined as the most probable distribution function, using no additional information. This is the distribution function maximizing the statistical entropy [32, 33].

We will sketch here only very shortly the entropy maximization. A detailed derivation with the alignment tensor as variable is given in [34, 35].

In order to introduce an entropy, it is necessary to go to the microscopic level of single particles with positions, velocities, orientations and angular velocities. On this level, a phase space distribution is defined, and the continuum mechanical fields, like mass density, energy density and orientation tensors are defined as averages with this phase space distribution function f^Γ. The entropy density is defined as

$$\eta(\boldsymbol{x}, t) = -K \int_\Gamma \sum_{\alpha=1}^{N} f^\Gamma(\bar{\Gamma}, t) \ln f^\Gamma(\bar{\Gamma}, t) \delta(\boldsymbol{x} - \boldsymbol{x}^\alpha) d\bar{\Gamma} \tag{11}$$

with Boltzmann constant K, number of particles N, the δ-distribution and integral over the phase space.

This entropy is maximized with respect to the phase space distribution function under the constraints, that the field quantities of mass density, energy density, momentum density, angular momentum density and second order alignment tensor have correct values, when calculated with this distribution function. A phase space distribution of exponential form results. The orientation distribution function (ODF), introduced previously, is derived by summing up over all particles inside the volume element. The ODF derived by entropy maximization is of the form

$$f(\boldsymbol{x}, t, \boldsymbol{n}) = \frac{\exp\left(-\boldsymbol{\Lambda}(\boldsymbol{x}, t) : \boldsymbol{n}\boldsymbol{n}\right)}{\int_{S^2} \exp\left(-\boldsymbol{\Lambda}(\boldsymbol{x}, t) : \boldsymbol{n}\boldsymbol{n}\right) d^2 n} = \frac{\exp\left(-\boldsymbol{\Lambda}(\boldsymbol{x}, t) : \overline{\boldsymbol{n}\boldsymbol{n}}\right)}{\int_{S^2} \exp\left(-\boldsymbol{\Lambda}(\boldsymbol{x}, t) : \overline{\boldsymbol{n}\boldsymbol{n}}\right) d^2 n} \; . \tag{12}$$

The parameter $\boldsymbol{\Lambda}$ is determined by the second order alignment tensor, and all higher order alignment tensors may be calculated with this distribution function.

3 The uniaxial case

An important special case is a rotation symmetric distribution function with rotation symmetry axis $\boldsymbol{d}$. In the case of liquid crystals a uniaxial distribution function is often observed in the absence of electromagnetic fields. In this case, the alignment tensors may be expressed in terms of the unit vector $\boldsymbol{d}$ and scalar parameters:

$$\boldsymbol{a} = S^{(2)} \ \overline{\boldsymbol{dd}} \tag{13}$$

$S^{(2)}$ is denoted as scalar order parameter, and

$$\boldsymbol{a}^{(4)} = S^{(4)} \ \overline{\boldsymbol{dddd}} \tag{14}$$

In the uniaxial case, the closure problem reduces to calculating the fourth order parameter $S^{(4)}$ in terms of the second order parameter $S^{(2)}$.

3.1 Maximum entropy closure in the uniaxial case

In the uniaxial case the orientation distribution function has a rotational symmetry around an axis $\boldsymbol{d}$. For symmetry reasons it is of the form

$$\begin{aligned} f(\boldsymbol{x},t,\boldsymbol{n}) &= \frac{\exp\{-\boldsymbol{\Lambda}(\boldsymbol{x},t) : \boldsymbol{nn}\}}{\int_{S^2} \exp\{-\boldsymbol{\Lambda}(\boldsymbol{x},t) : \boldsymbol{nn} d^2 n\}} = \\ &= \frac{\exp\{-l(\boldsymbol{x},t)\boldsymbol{d}(\boldsymbol{x},t)\boldsymbol{d}(\boldsymbol{x},t) : \boldsymbol{nn}\}}{\int_{S^2} \exp\{-l(\boldsymbol{x},t)\boldsymbol{d}(\boldsymbol{x},t)\boldsymbol{d}(\boldsymbol{x},t) : \boldsymbol{nn} d^2 n\}} \ . \end{aligned} \tag{15}$$

l is a scalar parameter. The scalar order parameters $S^{(2)}$ and $S^{(4)}$ depend on the parameter l. The dependence has been expressed in terms of the error-function [35]. It is not possible to eliminate l analytically from these relations, but a parametric plot shows $S^{(4)}$ as a function of $S^{(2)}$, see right hand side of figure 2 from [35].

3.2 Quadratic, linear and hybrid closure in the uniaxial case

In this subsection we derive the closure relation for the scalar order parameters in the case of the well known closure relations for the alignment tensor. These closure relations will be compared to the maximum entropy closure in the uniaxial case.

The scalar order parameters $S^{(2)}$ and $S^{(4)}$ are defined in terms of the alignment tensors. However, linear, quadratic and hybrid closure are formulated for the orientation tensors. Because $\boldsymbol{a} = \overline{\boldsymbol{A}}$ and $\boldsymbol{a}^{(4)} = \overline{\boldsymbol{A}^{(4)}}$, we have for the orientation tensors

$$\boldsymbol{A} = S^{(2)} \ \overline{\boldsymbol{dd}} \ + \frac{1}{3}\boldsymbol{\delta} \tag{16}$$

and consequently

$$\boldsymbol{A} : \boldsymbol{dd} = \frac{2}{3} S^{(2)} + \frac{1}{3}. \tag{17}$$

The fourth order alignment tensor

$$\boldsymbol{a}^{(4)} = S^{(4)} \ \overline{\boldsymbol{dddd}} \tag{18}$$

reads in components

$$a^{(4)}_{ijkl} = S^{(4)} \Big(d_i d_j d_k d_l - \frac{1}{7} (\delta_{ij} d_k d_l + \delta_{ik} d_j d_l + \delta_{il} d_j d_k + $$
$$+ \delta_{jk} d_i d_l + \delta_{jl} d_i d_k + \delta_{kl} d_i d_j) + \frac{1}{35} (\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) \Big) \tag{19}$$

The scalar order parameters are obtained by contractions with the unit vector $\boldsymbol{d}$.
From equation (19) it follows

$$\boldsymbol{a}^{(4)} :: \boldsymbol{dddd} = \frac{8}{35} S^{(4)}. \tag{20}$$

On the other hand with the closure relations (8), (9), (10) and equations (4) and (17) we have

$$\boldsymbol{a}^{(4)} :: \boldsymbol{dddd} = \boldsymbol{A}^{(4)} :: \boldsymbol{dddd} - \frac{6}{7}\boldsymbol{A} : \boldsymbol{dd} + \frac{3}{35}$$
$$= (1-f)\left(-\frac{3}{35} + \frac{6}{7}\boldsymbol{A} : \boldsymbol{dd}\right) + f\,(\boldsymbol{A} : \boldsymbol{dd})^2 - \frac{6}{7}\boldsymbol{A} : \boldsymbol{dd} + \frac{3}{35} =$$
$$= f\left(\frac{4}{9} S^{(2)^2} - \frac{8}{63} S^{(2)} - \frac{4}{45}\right), \tag{21}$$

Equations (20) and (21) together result in the closure relation for the scalar order parameter:

$$S^{(4)} = f\left(\frac{35}{18} S^{(2)^2} - \frac{5}{9} S^{(2)} - \frac{7}{18}\right) \tag{22}$$

In the special case of the linear closure $f = 0$, the fourth order parameter $S^{(4)} = 0$. The quadratic closure $f = 1$ results in

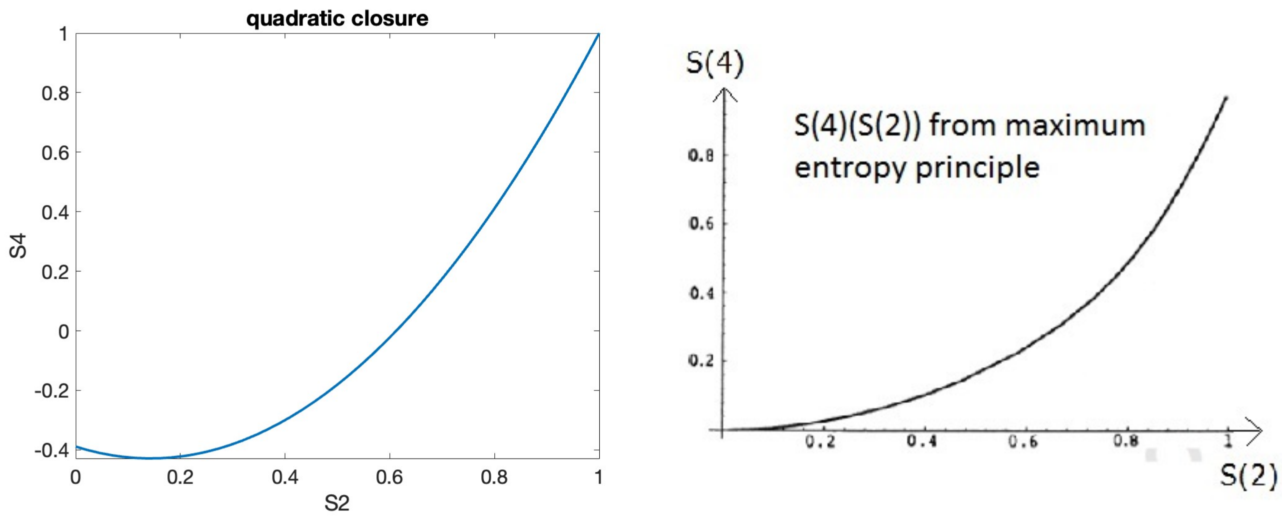
$$S^{(4)} = \frac{35}{18} S^{(2)^2} - \frac{5}{9} S^{(2)} - \frac{7}{18} \tag{23}$$

The result is shown for $f = 1$ on the left hand side of figure 2 compared to the maximum entropy closure relation on the right hand side. $f = 1$ corresponds to the quadratic closure, but the factor f results only in a scaling of the graph, not altering the graph qualitatively. We observe, that the maximum entropy closure results in $S^{(4)} > 0$ for any $S^{(2)} > 0$ in contrast to the quadratic closure. $S^{(4)} > 0$ means, that the fourth order alignment tensor term is a correction term, making the distribution function stronger concentrated around the preferred axis.

3.3 Approximation of the maximum entropy closure by second order polynomials

The maximum entropy closure relation in the uniaxial case may be approximated by a second order polynomial. In figure 3 it is shown a piecewise approximation with second order polynomials in subintervals of length 0.2. The fit polynomials are

$$S^{(4)} = 0,729\ S^{(2)^2} - 0,021\ S^{(2)}\ , \quad S^{(2)} \in [0; 0,2] \tag{24}$$
$$S^{(4)} = 0,833\ S^{(2)^2} - 0,075\ S^{(2)} + 0,007\ , \quad S^{(2)} \in [0,2; 0,4] \tag{25}$$
$$S^{(4)} = 1,042\ S^{(2)^2} - 0,417\ S^{(2)} + 0,11\ , \quad S^{(2)} \in [0,4; 0,6] \tag{26}$$
$$S^{(4)} = 1,312\ S^{(2)^2} - 0,607\ S^{(2)} + 0,127\ , \quad S^{(2)} \in [0,6; 0,8] \tag{27}$$
$$S^{(4)} = 3,062\ S^{(2)^2} - 3,057\ S^{(2)} + 0,967\ , \quad S^{(2)} \in [0,8; 1]\ . \tag{28}$$



(a) (b)

Figure 2: Quadratic closure on the left hand side and maximum entropy closure relation on the right hand side, (maximum entropy closure taken from [35]).

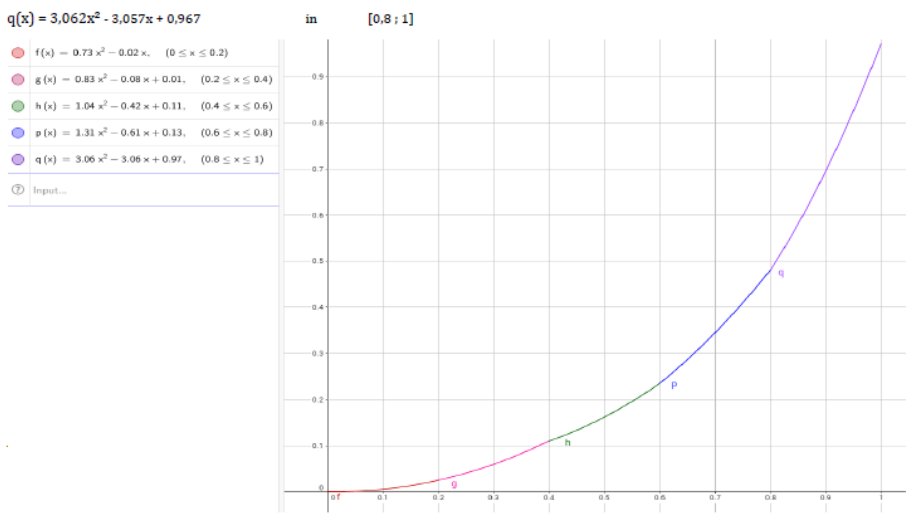


Figure 3: Second order fit polynomials to the maximum entropy closure.

A quadratic fit polynomial for the whole range of second order parameter is

$$S^{(4)} = 1,365\ S^{(2)^2} - 0,393\ S^{(2)}\ , \quad S^{(2)} \in [0;1]\ . \tag{29}$$

The piecewise approximation as well as the approximation in the whole range are not of the form of the quadratic closure, equation (23) and also not of the form of the hybrid closure. In the case of a piecewise approximation, the coefficients depend on the range of the scalar order parameter.

4 The maximum entropy closure in the general case

The distribution function maximizing the statistical entropy under constraints is [35]:

$$f(\boldsymbol{x},t,\boldsymbol{n}) = \frac{e^{-\boldsymbol{\Lambda}(\boldsymbol{x},t):\overline{\boldsymbol{nn}}}}{\int_{S^2} e^{-\boldsymbol{\Lambda}(\boldsymbol{x},t):\overline{\boldsymbol{nn}}}\, d^2n} =: \frac{e^{-\boldsymbol{\Lambda}(\boldsymbol{x},t):\overline{\boldsymbol{nn}}}}{Z}\ . \tag{30}$$

We obtain for the alignment tensor:

$$\begin{aligned}\boldsymbol{a}(\boldsymbol{x},t) &= \int_{S^2} \overline{\boldsymbol{nn}}\ \frac{e^{-\boldsymbol{\Lambda}(\boldsymbol{x},t):\overline{\boldsymbol{nn}}}}{Z} = \frac{1}{Z}\int_{S^2} -\frac{\partial}{\partial \boldsymbol{\Lambda}} e^{-\boldsymbol{\Lambda}:\overline{\boldsymbol{nn}}}\, d^2n \\ &= -\frac{1}{Z}\frac{\partial}{\partial \boldsymbol{\Lambda}}\int_{S^2} e^{-\boldsymbol{\Lambda}:\overline{\boldsymbol{nn}}}\, d^2n = -\frac{1}{Z}\frac{\partial Z}{\partial \boldsymbol{\Lambda}}\end{aligned} \tag{31}$$

This is an implicit relation between the alignment tensor and the parameter $\boldsymbol{\Lambda}$. We will use the entropy density for the identification of $\boldsymbol{\Lambda}$.

The entropy density η is introduced on the microscopic level in terms of a statistical distribution function. It has been shown [35], that the orientation dependent part of the statistical entropy can be expressed in terms of the mesoscopic distribution function $f(\boldsymbol{x},t,\boldsymbol{n})$ in the form:

$$\eta(\boldsymbol{x},t) = \int_{S^2} f(\boldsymbol{x},t,\boldsymbol{n}) \ln f(\boldsymbol{x},t,\boldsymbol{n}) d^2n\ . \tag{32}$$

With the distribution function eq. (30) this leads to:

$$\eta(\boldsymbol{x},t) = \int_{S^2} -\boldsymbol{\Lambda}(\boldsymbol{x},t) : \overline{\boldsymbol{nn}}\ f(\boldsymbol{x},t,\boldsymbol{n}) d^2n - \ln Z = -\boldsymbol{\Lambda} : \boldsymbol{a} - \ln Z\ . \tag{33}$$

The time derivative of the entropy density is calculated as:

$$\dot{\eta} = -\dot{\boldsymbol{\Lambda}} : \boldsymbol{a} - \boldsymbol{\Lambda} : \dot{\boldsymbol{a}} - \frac{\dot{Z}}{Z} = -\boldsymbol{\Lambda} : \dot{\boldsymbol{a}} \tag{34}$$

because

$$\begin{aligned}\dot{Z} &= \frac{d}{dt}\left(\int_{S^2} e^{-\boldsymbol{\Lambda}(\boldsymbol{x},t):\overline{\boldsymbol{nn}}}\, d^2n\right) = -\dot{\boldsymbol{\Lambda}} : \int_{S^2} e^{-\boldsymbol{\Lambda}(\boldsymbol{x},t):\overline{\boldsymbol{nn}}}\ \overline{\boldsymbol{nn}}\ d^2n = \\ &= -\dot{\boldsymbol{\Lambda}} : \boldsymbol{a}Z\ .\end{aligned} \tag{35}$$

On the other hand, the entropy density is a constitutive function, depending on the alignment tensor $\boldsymbol{a}$. Therefore, we have for the time derivative of the orientation dependent entropy density:

$$\dot{\eta}(\boldsymbol{a}(\boldsymbol{x},t)) = \frac{\partial \eta}{\partial \boldsymbol{a}} : \dot{\boldsymbol{a}}\ . \tag{36}$$

Comparing the time derivatives of the entropy density, equations (34) and (36), leads to the identification of the parameter $\mathbf{\Lambda}$ in the distribution function:

$$\mathbf{\Lambda} = -\frac{\partial \eta}{\partial \boldsymbol{a}} \; . \tag{37}$$

We have identified the maximum entropy distribution function to be:

$$f(\boldsymbol{x}, t, \boldsymbol{n}) = \frac{e^{\frac{\partial \eta}{\partial \boldsymbol{a}}(\boldsymbol{x},t) : \overline{\boldsymbol{nn}}}}{\int_{S^2} e^{\frac{\partial \eta}{\partial \boldsymbol{a}}(\boldsymbol{x},t) : \overline{\boldsymbol{nn}}} \, d^2 n} \; . \tag{38}$$

With this distribution function alignment tensors and orientation tensors of any order can be calculated.

4.1 Lowest order approximation

The entropy density is supposed to be a continuously differentiable function of the alignment tensor (with $trace(\boldsymbol{a}) = 0$). If the power series

$$\eta(\boldsymbol{a}) = \eta_0 + \frac{1}{2}\eta_2 trace(\boldsymbol{a} \cdot \boldsymbol{a}) + \frac{1}{6}\eta_3 trace(\boldsymbol{a} \cdot \boldsymbol{a} \cdot \boldsymbol{a}) + \mathcal{O}(\boldsymbol{a}^4) \tag{39}$$

is truncated after the second order term, it results

$$\frac{\partial \eta}{\partial \boldsymbol{a}} = \eta_2 \boldsymbol{a}. \tag{40}$$

Inserting the series expansion of the distribution function for the calculation of the fourth order alignment tensor

$$\begin{aligned} \boldsymbol{a}^{(4)} = \int_{S^2} f(\boldsymbol{x}, t, \boldsymbol{n}) \; \overline{\boldsymbol{nnnn}} \; d^2 n = \frac{\int_{S^2} e^{\eta_2 \boldsymbol{a}(\boldsymbol{x},t) : \overline{\boldsymbol{nn}}} \; \overline{\boldsymbol{nnnn}} \; d^2 n}{\int_{S^2} e^{\eta_2 \boldsymbol{a}(\boldsymbol{x},t) : \overline{\boldsymbol{nn}}} \, d^2 n} = \\ = \frac{\int_{S^2} \sum_{i=0}^{\infty} \left(\frac{1}{i!} (\eta_2 \boldsymbol{a}(\boldsymbol{x}, t) : \overline{\boldsymbol{nn}} \,)^i \right) \; \overline{\boldsymbol{nnnn}} \; d^2 n}{\int_{S^2} \sum_{i=0}^{\infty} \left(\frac{1}{i!} (\eta_2 \boldsymbol{a}(\boldsymbol{x}, t) : \overline{\boldsymbol{nn}} \,)^i \right) d^2 n} \end{aligned} \tag{41}$$

results in integrals of the form [14, 13] (with the projector $\mathbf{\Delta}^{(4)}$ on the symmetric traceless part of a fourth order tensor):

$$\int_{S^2} 1 d^2 n = 4\pi \tag{42}$$

$$\int_{S^2} \overline{\boldsymbol{nn}} \; d^2 n = 0 \tag{43}$$

$$\int_{S^2} \overline{\boldsymbol{nnnn}} \; d^2 n = 0 \tag{44}$$

$$\int_{S^2} \overline{\boldsymbol{nn}} \; \overline{\boldsymbol{nnnn}} \; d^2 n = 0 \tag{45}$$

$$\int_{S^2} \overline{\boldsymbol{nn}} \; \overline{\boldsymbol{nn}} \; \overline{\boldsymbol{nnnn}} \; d^2 n = 4\pi \frac{4!}{9!!} \mathbf{\Delta}^{(4)} = 4\pi \frac{8}{315} \mathbf{\Delta}^{(4)} \tag{46}$$

and higher orders. Keeping only the lowest order non-zero terms in equation (41) results in

$$\boldsymbol{a}^{(4)} = \frac{8}{315}\frac{\eta_2^2}{2}\boldsymbol{\Delta}^{(4)} : :\boldsymbol{aa} = \frac{4}{315}\eta_2^2\ \overline{\boldsymbol{aa}}\,, \tag{47}$$

i.e. in a closure relation of the form of a quadratic closure. A term linear in $\boldsymbol{a}$ does not appear due to equation (45). However, higher order terms have been neglected in the nominator and in the denominator. The closure relation (47) fulfills the symmetry condition ($\boldsymbol{a}^{(4)}$ is symmetric with respect to any pair of indices) and the constraint $a^{(4)}_{ijll} = 0$.

Remark on the sixth order alignment tensor in the lowest order approximation
Because

$$\int_{S^2} \overline{\boldsymbol{nnnnnn}}\ d^2n = 0 \tag{48}$$

$$\int_{S^2} \overline{\boldsymbol{nn}}\ \overline{\boldsymbol{nnnnnn}}\ d^2n = 0 \tag{49}$$

$$\int_{S^2} \overline{\boldsymbol{nn}}\ \overline{\boldsymbol{nn}}\ \overline{\boldsymbol{nnnnnn}}\ d^2n = 0 \tag{50}$$

the lowest order approximation leads to a closure form

$$\boldsymbol{a}^{(6)} \propto \overline{\boldsymbol{aaa}} \tag{51}$$

without lower order terms in the second order alignment tensor.

5 Conclusions

In the uniaxial case the closure problem reduces to a relation between the scalar order parameters of fourth and second order. An implicit relation between these order parameters has been derived previously from the orientation distribution function, which maximizes the statistical entropy. This closure relation has been compared here to the quadratic, linear and hybrid closure. One important difference is the sign of the fourth order parameter, which is always positive for the maximum entropy closure. In case of the quadratic and hybrid closure the fourth order parameter has negative sign for small values and positive sign for larger values of the second order parameter. Negative sign means, that the fourth order correction term in the expansion of the ODF decreases the degree of orientational order. Second order fit polynomials for the maximum entropy closure have been presented and could be used in numerical solutions of the orientation tensor dynamics, where a closure relation is always necessary. In the general case without rotation symmetry of the orientation distribution function, the parameter $\boldsymbol{\Lambda}$ in the maximum entropy distribution has been identified as the derivative of the entropy density with respect to the second order alignment tensor. In the lowest order approximation this derivative is proportional to the alignment tensor. Expansion of the exponential function leads to an approximation of the fourth order alignment tensor proportional to the square of the second order alignment tensor without linear terms, i.e. a quadratic closure form. The same way approximations for the higher order alignment tensors may be calculated. The result for the sixth order alignment tensor is proportional to $\overline{\boldsymbol{aaa}}$ without lower order terms. These closure relations may also be expressed in terms of the non-traceless orientation tensors, but do not have such a simple form then.

6 Declarations

6.1 Competing Interests

I have no competing interests to declare.

6.2 Publisher's Note

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