

Application of the Mesoscopic Theory to Dipolar Media

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Mesoscopic continuum theory is a way to deal with complex materials, i.e. materials with an internal structure, which can change under the action of external fields, within continuum theory. In the mesoscopic theory field quantities are introduced, which depend not only on position and time, but also on an additional, so called mesoscopic variable. In our case this additional variable is the orientation of a dipole. The orientation distribution function (ODF) gives the fraction of dipoles of a particular orientation. The magnetization is proportional to the first moment of the ODF. Balance equations for the mesoscopic fields, and an equation of motion for the distribution function have been derived in the general case. With some additional assumptions these equations are used here to derive a relaxation equation for the magnetization. The linear limit case of this relaxation equation is the well known DEBYE equation.

1 Introduction

Let us call substances consisting of spherical particles with interactions depending only on the interparticle distance *simple materials*. However most of the interesting and practically important materials have a more complicated internal structure. We will call these materials with internal degrees of freedom *complex materials*. Examples of such complex materials are materials with electric or magnetic dipole moments, which can be oriented under the action of external fields. For simplicity we will restrict ourselves here to magnetic dipoles, i.e. paramagnetic materials. The case of electric dipoles works analogously.

Electric and magnetic dipolar media have been studied extensively with the methods of irreversible non-equilibrium thermodynamics. Decomposing the vector of electric polarization into an elastic part and an irreversible part (an internal variable) a relaxation equation for the electric polarization \mathbf{P}

$$\xi_{(EP)}^{(0)} \mathbf{E} + \frac{d\mathbf{E}}{dt} = \xi_{(PE)}^{(0)} \mathbf{P} + \xi_{(PE)}^{(1)} \frac{d\mathbf{P}}{dt} \quad (1)$$

has been obtained (Kluitenberg, 1973, 1977), which was known before as the DEBYE equation (Debye, 1945). The same form of equation can be received replacing the polarization by the magnetization \mathbf{M} , and the electric field \mathbf{E} by the magnetic induction \mathbf{B} . This DEBYE equation has been generalized to a second order differential equation for the polarization, or magnetization (Kluitenberg, 1981; Ciancio et al., 1996), and also the cross coupling effects between relaxation of the polarization, heat conduction, and viscous flow have been taken into account (Ciancio, 1989).

The aim of the present paper is an introduction to the mesoscopic concept, the application to paramagnetic materials, and a derivation of a differential equation for the magnetization in dipolar media from the mesoscopic theory.

2 Mesoscopic Concept and Balance Equations

There are two principally different possibilities to deal with complex materials within continuum thermodynamics: The first way is to introduce additional fields depending on position and time. These fields can be internal variables (Muschik, 1990; Maugin and Muschik, 1994), order or damage parameters (Maugin, 1992), Cosserat triads (Cosserat and Cosserat, 1907, 1909; Truesdell and Noll, ...), directors (Eriksen, 1960; Leslie, 1965), alignment and conformation tensors (Hess, 1975; Maugin and Drouot, 1988). The other way is a so called mesoscopic theory. The idea is to enlarge the domain of the field quantities. The new mesoscopic fields are defined on the space $\mathbb{R}_x^3 \times \mathbb{R}_t \times M$. The manifold M is given by the set of values the internal degree of freedom can take. Therefore the choice of M depends on the complex material under consideration, but if the internal structure is known, the nature of M is clear.

We call this way of dealing with the internal structure of complex materials a mesoscopic concept, because it includes more information than a macroscopic theory on $\mathbb{R}_x^3 \times \mathbb{R}_t$, but the molecular level is not considered like in a microscopic approach. The mesoscopic level is between the microscopic and the macroscopic level. The domain of the mesoscopic field quantities $\mathbb{R}_x^3 \times \mathbb{R}_t \times \mathcal{M}$ is called mesoscopic space.

Beyond the use of additional variables \mathbf{m} the mesoscopic concept introduces a statistical element, the so-called *mesoscopic distribution function* (MDF) $f(\mathbf{m}, \mathbf{x}, t)$ generated by the different values of the mesoscopic variable of the particles in a volume element

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

The MDF describes the distribution of \mathbf{m} in a volume element around \mathbf{x} at time t , and therefore it is normalized

$$\int f(\mathbf{x}, \mathbf{m}, t) d\mathcal{M} = 1. \quad (3)$$

It is the probability density of having the particular value \mathbf{m} of the mesoscopic variable in the continuum element at position \mathbf{x} and time t . Therefore it is the fraction

$$f(\mathbf{m}, \mathbf{x}, t) = \frac{\rho(\mathbf{x}, \mathbf{m}, t)}{\rho(\mathbf{x}, t)}. \quad (4)$$

Now the fields as mass density, momentum density, etc. are defined on the mesoscopic space. For distinguishing these fields from the macroscopic ones we add the word ‘‘mesoscopic’’.

2.1 Mesoscopic Balance Equations

Let G denote a region in $\mathbb{R}^3 \times \mathcal{M}$ and \mathbf{X} the density of an extensive quantity. Then the global quantity in the region G changes due to a flux over the boundary of G and due to production and supply within G :

$$\frac{d}{dt} \int_G \mathbf{X} d^3x d\mathbf{m} = \int_{\partial G} \boldsymbol{\Phi}_X(\cdot) d\mathbf{a} + \int_G \Sigma_X(\cdot) d^3x d\mathbf{m}. \quad (5)$$

A generalized Reynolds transport theorem in the mesoscopic space, analogous to the one in Ehretraut (1996) is used to transform the time derivative, and Gauss theorem is applied to the boundary integral. The boundary ∂G of G consists of a boundary in position space and a boundary on the manifold \mathcal{M} . Then it results in regular points of the continuum the *local mesoscopic balance* (Blenk et al., 1991)

$$\frac{\partial}{\partial t} \mathbf{X} + \nabla_x \cdot [\mathbf{v}(\cdot) \mathbf{X} - \mathbf{S}(\cdot)] + \nabla_m \cdot [\mathbf{u}(\cdot) \mathbf{X} - \mathbf{R}(\cdot)] = \Sigma_X(\cdot). \quad (6)$$

Here the independent field $\mathbf{u}(\cdot)$, defined on the mesoscopic space, describes the change in time of the set of mesoscopic variables: With respect to \mathbf{m} the *mesoscopic change velocity* $\mathbf{u}(\cdot)$ is the analogue to the mesoscopic material velocity $\mathbf{v}(\cdot)$ referring to \mathbf{x} : If a particle is characterized by $(\mathbf{m}, \mathbf{x}, t)$, then for $\Delta t \rightarrow +0$ it is characterized by $(\mathbf{m} + \mathbf{u}(\cdot)\Delta t, \mathbf{x} + \mathbf{v}(\cdot)\Delta t, t + \Delta t)$ at later times $t + \Delta t$. Besides the usual gradient also the gradient with respect to the set of mesoscopic variables appears. This expresses the fact that there is a flux over the boundary on the manifold \mathcal{M} . \mathbf{S} is the spatial part of the non-convective flux of \mathbf{X} , and \mathbf{R} is the non-convective flux in mesoscopic space.

According to equation (4) we obtain from the mesoscopic mass balance (8) a balance of the MDF $f(\cdot)$ by inserting its definition (Papenfuss, 2000; Papenfuss and Muschik, 1998):

$$\begin{aligned} \frac{\partial}{\partial t} f(\cdot) + \nabla_x \cdot [\mathbf{v}(\cdot) f(\cdot)] &+ \nabla_m \cdot [\mathbf{u}(\cdot) f(\cdot)] + \\ &+ f(\cdot) \left[\frac{\partial}{\partial t} + \mathbf{v}(\cdot) \cdot \nabla_x \right] \ln \rho(\mathbf{x}, t) = 0. \end{aligned} \quad (7)$$

3 Application of the Mesoscopic Concept to Dipolar Media

Let us denote the orientation of a single dipole by a unit vector \mathbf{n} with $\mathbf{n} \cdot \mathbf{n} = 1$. The orientation of the dipole can take any value on the unit sphere S^2 . According to the concept of the mesoscopic theory we introduce mesoscopic fields, defined on the enlarged space, the mesoscopic space $\mathbb{R}_x^3 \times \mathbb{R}_t \times S^2$, where the last argument in the domain of the fields is the orientation of the dipole \mathbf{n} . This mesoscopic space is the same as for liquid crystals, and therefore also the mesoscopic balance equations look the same for a dipolar medium, as for liquid crystals. The difference between these two materials shows up in the constitutive theory. An important difference is that for liquid crystals a so called "head-tail-symmetry" is observed, meaning that there \mathbf{n} and $-\mathbf{n}$ have to be identified. A similar symmetry does *not* exist for dipoles, because the dipole, and the reversed one are distinguished.

In the following we will need only the balances of mass, and of internal angular momentum, which will be the only balance equations given explicitly here. The whole set of mesoscopic balance equations can be taken from the ones known from liquid crystal theory to be found in Blenk et al. (1993), Ehrebraut et al. (1997), Muschik et al. (1995), Blenk et al. (1991), Blenk and Muschik (1991). (\cdot) is the abbreviation for the mesoscopic set of variables $((\cdot) = (\mathbf{x}, \mathbf{n}, t))$.

Mesoscopic balance of mass:

$$\frac{\partial}{\partial t} \rho(\cdot) + \nabla_x \cdot \{\rho(\cdot) \mathbf{v}(\cdot)\} + \nabla_n \cdot \{\rho(\cdot) \mathbf{u}(\cdot)\} = 0. \quad (8)$$

The mesoscopic mass density $\rho(\mathbf{x}, \mathbf{n}, t)$ is the mass density of dipoles of the particular orientation \mathbf{n} , and analogously for the other mesoscopic fields. \mathbf{v} is the material velocity of particles of orientation \mathbf{n} . \mathbf{u} is the orientation change velocity, $\mathbf{u} = \dot{\mathbf{n}}$. It is tangential to the unit sphere, because \mathbf{n} is normalized ($\mathbf{n} \perp \mathbf{u}$). ∇_n denotes the covariant derivative with respect to the orientation variable. The last term on the left hand side of the mass balance is the flux in orientation space. It expresses the fact that the fraction of dipoles of orientation \mathbf{n} changes if the dipoles change their orientation.

Mesoscopic balance equations of momentum, angular momentum, and energy can be derived in addition. In the balance of angular momentum the internal angular momentum, the spin appears, due to rotations of the dipoles. As the balance of spin is the only equation needed here, we will show only this one:

Mesoscopic balance of spin:

$$\begin{aligned} & \frac{\partial \rho(\cdot) \mathbf{s}(\cdot)}{\partial t} + \nabla \cdot \left(\rho(\cdot) \mathbf{v}(\cdot) \mathbf{s}(\cdot) - (\mathbf{n} \times \mathbf{\Pi}(\cdot))^T \right) + \\ & + \nabla_n \cdot (\rho(\cdot) \mathbf{u}(\cdot) \mathbf{s}(\cdot)) = \mathbf{e} : \mathbf{t}(\cdot) + \rho(\cdot) \mathbf{n} \times \mathbf{g}(\cdot) \quad . \end{aligned} \quad (9)$$

The symbols are: $\mathbf{s}(\cdot)$: mesoscopic specific spin density (due to rotations of dipoles), $\mathbf{n} \times \mathbf{\Pi}(\cdot)$: couple stresses (short range momentum density), $\mathbf{t}(\cdot)$: stress tensor, $\mathbf{n} \times \mathbf{g}(\cdot)$: couple forces (long range momentum density). In our example the couple forces are the momentum exerted on a dipole by the external field. The antisymmetric part of the stress tensor causes a production of spin. The spin density is related to the orientation change velocity by

$$\mathbf{s}(\cdot) = \theta \mathbf{n} \times \mathbf{u} \quad . \quad (10)$$

θ is the moment of inertia of the dipoles. It is assumed here to be a scalar quantity, i.e. in this model only rotations perpendicular to the orientation of the dipole have a moment of inertia, and the same one for all rotation axes.

As always in continuum mechanics the balance equations are not a closed set of equations, but constitutive equations are needed. In order to make a constitutive ansatz the domain of the constitutive mappings, the state space, has to be chosen. For mesoscopic material properties there are two possibilities: The state space can include only mesoscopic quantities, or it can include mesoscopic and macroscopic quantities. Here the second possibility is appropriate, because the magnetic induction \mathbf{B} is for instance a macroscopic quantity.

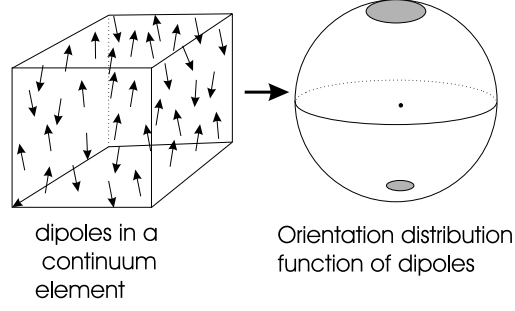


Figure 1: The dipole moments give rise to an orientation distribution function on the unit sphere.

3.1 Orientation Distribution Function and Alignment Tensors

Macroscopically the dipole moments show up as a magnetization only, if their orientations are not distributed isotropically, but they are oriented more or less parallel. This orientational order can be described introducing an orientation distribution function (ODF) (see figure 1).

The orientation distribution function gives the probability density of finding a dipole of orientation \mathbf{n} in the continuum element at position \mathbf{x} and time t . Therefore it is defined as the following mass fraction

$$f(\mathbf{x}, \mathbf{n}, t) = \frac{\rho(\mathbf{x}, \mathbf{n}, t)}{\rho(\mathbf{x}, t)} . \quad (11)$$

From the mesoscopic balance of mass and the macroscopic equation of continuity one can derive the following equation of motion for the ODF in the case of an incompressible fluid, where in addition dipoles of different orientations have the same translational velocity ($\mathbf{v}(\mathbf{x}, \mathbf{n}, t) = \mathbf{v}(\mathbf{x}, t)$):

$$\frac{\partial f(\cdot)}{\partial t} + \mathbf{v}(\mathbf{x}, t) \cdot \nabla f(\cdot) + \nabla_{\mathbf{n}} \cdot (\mathbf{u}(\cdot) f(\cdot)) = 0 . \quad (12)$$

As the orientation distribution function of the dipoles is not directly measurable, we are looking for macroscopic quantities, which distinguish between orientational order of the dipoles and an isotropic distribution. These are the anisotropic moments of the ODF and they are called alignment tensors. They are defined as

$$\mathbf{a}^{(k)}(\mathbf{x}, t) := \int_{S^2} f(\mathbf{x}, \mathbf{n}, t) \underbrace{\mathbf{n} \dots \mathbf{n}}_k d^2 \mathbf{n} , \quad (13)$$

where $\underbrace{\dots}_k$ denotes the symmetric irreducible (traceless) part of a tensor. These alignment tensors are introduced in such a way, that they all vanish if the distribution of dipole orientations is isotropic. Sometimes it is convenient to introduce also alignment tensors $\mathbf{A}^{(k)}$, which are not traceless:

$$\mathbf{A}^{(k)}(\mathbf{x}, t) := \int_{S^2} f(\mathbf{x}, \mathbf{n}, t) \underbrace{\mathbf{n} \dots \mathbf{n}}_k d^2 \mathbf{n} . \quad (14)$$

3.2 Exploitation of the Balance of Spin

The differential equation for the distribution function is not a closed equation, as long as no expression for the orientation change velocity \mathbf{u} is given. The equation to determine the orientation change velocity is the balance of spin. As it is very difficult to solve this partial differential equation on $\mathbb{R}_{\mathbf{x}}^3 \times \mathbb{R}_t \times S^2$, and also the initial conditions are not clear, we will make here the following simplifying assumptions:

1. The spin is stationary: $\frac{\partial \mathbf{S}(\cdot)}{\partial t} = 0$.
2. There are no spatial gradients, the system is homogenous.

Then the spin balance (9) simplifies to

$$\rho(\cdot)\mathbf{u}(\cdot) \cdot \nabla_{\mathbf{n}}\mathbf{s}(\cdot) = \boldsymbol{\epsilon} : \mathbf{t}(\cdot) + \rho(\cdot)\mathbf{n} \times \mathbf{g}(\cdot) \quad . \quad (15)$$

$\mathbf{t}(\cdot)$ and $\mathbf{g}(\cdot)$ are constitutive quantities. They depend on the variables in the state space (see below) in a material dependent manner. From eq. (15) we see that $\boldsymbol{\epsilon} : \mathbf{t}(\cdot) + \rho(\cdot)\mathbf{n} \times \mathbf{g}(\cdot)$ vanishes if $\nabla_{\mathbf{n}}\mathbf{s}(\cdot) = \mathbf{0}$, i.e. it is a homogenous function of $\nabla_{\mathbf{n}}\mathbf{s}(\cdot)$ with a constitutive function $\mathbf{G}(\cdot)$. Therefore we can write down a constitutive equation of the form

$$\boldsymbol{\epsilon} : \mathbf{t}(\cdot) + \rho(\cdot)\mathbf{n} \times \mathbf{g}(\cdot) = \mathbf{G}(\cdot) \cdot \nabla_{\mathbf{n}}\mathbf{s}(\cdot) \quad . \quad (16)$$

Additionally \mathbf{u} is orthogonal to \mathbf{n} . With the projector \mathbf{P} onto the space orthogonal to \mathbf{n} we have:

$$\mathbf{u} = \mathbf{P} \cdot \mathbf{G}(\cdot), \quad \mathbf{P} = \boldsymbol{\delta} - \mathbf{n}\mathbf{n} \quad . \quad (17)$$

For the mesoscopic quantity $\mathbf{G}(\cdot)$ we make a constitutive ansatz. Any vector field satisfying equation (19) is a solution of the stationary balance of spin for homogeneous systems.

3.3 State Space and Constitutive Equation

The set of variables, the constitutive quantities depend upon, i.e. the domain of the constitutive mappings is the state space \mathcal{Z} . Here we chose

$$\mathcal{Z} = \{\rho, \mathcal{T}, \mathbf{B}, \dot{\mathbf{B}}, \mathbf{a}^{(1)}, \mathbf{a}^{(2)}, \nabla_{\mathbf{n}}\rho(\cdot), \mathbf{n}\} \quad , \quad (18)$$

\mathcal{T} : temperature, \mathbf{B} : magnetic induction. It includes macroscopic and mesoscopic variables. The macroscopic variables are temperature, mass density, magnetic induction, its time derivative, and the first and second order alignment tensor. These alignment tensors in the state space account for the fact that the dipoles tend to align parallel, i.e. the surrounding dipoles exert an aligning "mean field". In a simpler model it would be enough to include only the first order alignment tensor. The first order tensor expresses the tendency of the dipoles to align parallel. The second order alignment tensor accounts for the influence of a quadrupolar ordering. We will discuss the case without the second order alignment tensor as a special case later.

The orientation change velocity \mathbf{u} depending on the orientation gradient of the mesoscopic mass density $\nabla_{\mathbf{n}}\rho(\cdot)$ means the inclusion of orientation diffusion.

The mass density ρ in the state space is the macroscopic one, because the dependence on the orientation \mathbf{n} is written out explicitly.

Then a representation theorem for \mathbf{G} linear in all quantities except for the mesoscopic variable \mathbf{n} gives:

$$\mathbf{u} = \mathbf{P} \cdot \mathbf{G}(\cdot) = (\boldsymbol{\delta} - \mathbf{n}\mathbf{n}) \cdot \left(\beta_1 \mathbf{B} + \beta_2 \dot{\mathbf{B}} + \beta_3 \nabla_{\mathbf{n}}\rho(\cdot) + \beta_4 \mathbf{a}^{(1)} + \beta_5 \mathbf{a}^{(2)} \cdot \mathbf{n} \right) . \quad (19)$$

The coefficients β_j are functions of the macroscopic mass density $\rho(\mathbf{x}, t)$ and the temperature $\mathcal{T}(\mathbf{x}, t)$. They do not depend on the mesoscopic mass density as the dependence on the mesoscopic variable \mathbf{n} is written explicitly in the constitutive ansatz.

3.4 Equation of Motion for the Magnetization

The first moment of eq. (12) reads

$$\frac{\partial}{\partial t} \int_{S^2} \mathbf{f} \mathbf{n} d^2\mathbf{n} + \mathbf{v} \cdot \nabla \int_{S^2} \mathbf{f} \mathbf{n} d^2\mathbf{n} + \int_{S^2} \mathbf{n} \nabla_{\mathbf{n}} \cdot (\mathbf{f} \mathbf{u}) d^2\mathbf{n} = 0 \quad . \quad (20)$$

On the other hand the variable \mathbf{n} is proportional to the microscopic magnetization (magnetization per unit mass), i.e. it is the orientation of the microscopic dipole moment: $\mathbf{m} = \alpha \mathbf{n}$ with $\alpha = \text{const.}$. The first moment of the orientation distribution function is proportional to the average of the microscopic magnetization, i.e. the macroscopic magnetization

$$\mathbf{M}(\mathbf{x}, t) = \alpha \rho(\mathbf{x}, t) \int_{S^2} f \mathbf{n} d^2 \mathbf{n} = \alpha \rho(\mathbf{x}, t) \mathbf{a}^{(1)} \quad . \quad (21)$$

The first two terms in equation (20) are derivatives of the first order alignment tensor. The third term is integrated by parts using Gauss theorem on the unit sphere. The resulting equation reads:

$$\frac{\partial \mathbf{a}^{(1)}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{a}^{(1)} = \frac{d \mathbf{a}^{(1)}}{dt} = \int_{S^2} f \mathbf{u} \cdot \nabla_{\mathbf{n}}(\mathbf{n}) d^2 \mathbf{n} \quad . \quad (22)$$

Then inserting the equation for the orientation change velocity eq. (19) and taking into account $\nabla_{\mathbf{n}}(\mathbf{n}) = \mathbf{P} = \delta - \mathbf{nn}$ and $\mathbf{n} \cdot \nabla_{\mathbf{n}}(\dots) = 0$ (because $\nabla_{\mathbf{n}}$ is the covariant derivative on the unit sphere) we obtain:

$$\begin{aligned} \frac{d \mathbf{a}^{(1)}}{dt} = \int_{S^2} \left(\beta_1 \mathbf{B} + \beta_2 \dot{\mathbf{B}} + \beta_3 \nabla_{\mathbf{n}} \rho(\cdot) + \beta_4 \mathbf{a}^{(1)} + \beta_5 \mathbf{a}^{(2)} \cdot \mathbf{n} \right. \\ \left. - \beta_1 \mathbf{nn} \cdot \mathbf{B} - \beta_2 \mathbf{nn} \cdot \dot{\mathbf{B}} - \beta_4 \mathbf{nn} \cdot \mathbf{a}^{(1)} - \beta_5 \mathbf{a}^{(2)} : \mathbf{nnn} \right) f d^2 \mathbf{n} \quad , \end{aligned} \quad (23)$$

using that \mathbf{P} is a projector ($\mathbf{P} \cdot \mathbf{P} = \mathbf{P}$).

The third term is transformed as follows:

$$\begin{aligned} \int_{S^2} \beta_3 \nabla_{\mathbf{n}} \rho(\cdot) f d^2 \mathbf{n} &= \int_{S^2} \beta_3 (\nabla_{\mathbf{n}} \rho(\cdot)) \frac{\rho(\cdot)}{\rho(\mathbf{x}, t)} d^2 \mathbf{n} \\ &= \frac{1}{\rho(\mathbf{x}, t)} \int_{S^2} \beta_3 \nabla_{\mathbf{n}} \left(\frac{\rho(\cdot)^2}{2} \right) d^2 \mathbf{n} = 0 \end{aligned} \quad (24)$$

because the integration is over a closed surface, and no boundary term occurs.

The first moment of the dipole distribution function is proportional to the magnetization (see equation (21)). In the resulting equation there enter also the second and the third orientational moments $\mathbf{A}^{(2)}$ and $\mathbf{A}^{(3)}$ of the dipole distribution function:

$$\mathbf{A}^{(2)} = \int_{S^2} f(\cdot) \mathbf{nn} d^2 \mathbf{n} \quad (25)$$

$$\mathbf{A}^{(3)} = \int_{S^2} f(\cdot) \mathbf{nnn} d^2 \mathbf{n} \quad , \quad (26)$$

which are denoted by capital \mathbf{A} , because they are not traceless. From equation (23) it follows in terms of the magnetization:

$$\begin{aligned} \alpha^{-1} \frac{d \mathbf{M}(\mathbf{x}, t)}{\rho(\mathbf{x}, t) dt} &= \beta_1 \mathbf{B}(\mathbf{x}, t) + \beta_2 \dot{\mathbf{B}}(\mathbf{x}, t) \\ &+ \beta_4 \alpha^{-1} \frac{\mathbf{M}(\mathbf{x}, t)}{\rho(\mathbf{x}, t)} + \beta_5 \alpha^{-1} \mathbf{a}^{(2)}(\mathbf{x}, t) \cdot \frac{\mathbf{M}(\mathbf{x}, t)}{\rho(\mathbf{x}, t)} \\ &- \beta_1 \mathbf{A}^{(2)}(\mathbf{x}, t) \cdot \mathbf{B}(\mathbf{x}, t) - \beta_2 \mathbf{A}^{(2)}(\mathbf{x}, t) \cdot \dot{\mathbf{B}}(\mathbf{x}, t) \\ &- \beta_4 \alpha^{-1} \mathbf{A}^{(2)}(\mathbf{x}, t) \cdot \frac{\mathbf{M}(\mathbf{x}, t)}{\rho(\mathbf{x}, t)} - \beta_5 \mathbf{a}^{(2)}(\mathbf{x}, t) : \mathbf{A}^{(3)}(\mathbf{x}, t) \quad . \end{aligned} \quad (27)$$

This is a macroscopic equation for the field quantities depending on position and time. If the material is

incompressible the left hand side simplifies to

$$\frac{d}{dt} \frac{\mathbf{M}(\mathbf{x}, t)}{\rho(\mathbf{x}, t)} = \frac{d\mathbf{M}(\mathbf{x}, t)}{dt} \frac{1}{\rho(\mathbf{x}, t)} . \quad (28)$$

The resulting differential equation for the magnetization is of the type of a relaxation equation. Interestingly orientation diffusion does not show up in the macroscopic equation for the magnetization.

Let us consider the special case that the second order alignment tensor is not included in the state space, i.e. $\beta_5 = 0$ and the material is incompressible:

$$\begin{aligned} \frac{1}{\alpha\rho} \frac{d\mathbf{M}}{dt} &= \beta_1 \mathbf{B} + \beta_2 \dot{\mathbf{B}} + \beta_4 \frac{1}{\alpha\rho} \mathbf{M} \\ &- \beta_1 \mathbf{A}^{(2)} \cdot \mathbf{B} - \beta_2 \mathbf{A}^{(2)} \cdot \dot{\mathbf{B}} - \beta_4 \frac{1}{\alpha\rho} \mathbf{A}^{(2)} \cdot \mathbf{M} . \end{aligned} \quad (29)$$

In both cases there is a coupling to higher order moments, at least to the second order one, which cannot be avoided, even if the second order alignment tensor is not included in the state space ($\beta_5 = 0$). Therefore a closure relation is needed, expressing the higher order moments in terms of the second order one. Such a closure relation can be derived from the principle of maximum entropy (Papenfuss and Muschik, 1999), or it has to be postulated as a constitutive equation. The simplest assumption is, that the orientations of the dipoles are statistically independent (which is an approximation only). Then the closure relations are very simple:

$$\mathbf{A}^{(2)} = \int_{S^2} f(\mathbf{x}, \mathbf{n}, t) \mathbf{n} \mathbf{n} d^2 \mathbf{n} = \int_{S^2} f(\mathbf{x}, \mathbf{n}, t) \mathbf{n} d^2 \mathbf{n} \int_{S^2} f(\mathbf{x}, \mathbf{n}, t) \mathbf{n} d^2 \mathbf{n} = \mathbf{a}^{(1)} \mathbf{a}^{(1)} \quad (30)$$

$$\begin{aligned} \mathbf{A}^{(3)} &= \int_{S^2} f(\mathbf{x}, \mathbf{n}, t) \mathbf{n} \mathbf{n} \mathbf{n} d^2 \mathbf{n} = \\ &= \int_{S^2} f(\mathbf{x}, \mathbf{n}, t) \mathbf{n} d^2 \mathbf{n} \int_{S^2} f(\mathbf{x}, \mathbf{n}, t) \mathbf{n} d^2 \mathbf{n} \int_{S^2} f(\mathbf{x}, \mathbf{n}, t) \mathbf{n} d^2 \mathbf{n} = \mathbf{a}^{(1)} \mathbf{a}^{(1)} \mathbf{a}^{(1)} . \end{aligned} \quad (31)$$

With these assumptions on the higher order alignment tensors, there results a closed equation for the magnetization. For the irreducible alignment tensor we have to insert $\mathbf{a}^{(2)} = \mathbf{A}^{(2)} - \frac{1}{3} \boldsymbol{\delta}$ with the unit tensor $\boldsymbol{\delta}$. With the second order alignment tensor in the state space we have:

$$\begin{aligned} \frac{d\mathbf{M}}{dt} &= \beta_1 \alpha\rho \mathbf{B} + \beta_2 \alpha\rho \dot{\mathbf{B}} + \beta_4 \mathbf{M} + \beta_5 \left(\frac{1}{\alpha^2 \rho^2} \mathbf{M} \mathbf{M} - \frac{1}{3} \boldsymbol{\delta} \right) \cdot \mathbf{M} \\ &- \beta_1 \frac{1}{\alpha\rho} \mathbf{M} \mathbf{M} \cdot \mathbf{B} - \beta_2 \frac{1}{\alpha\rho} \mathbf{M} \mathbf{M} \cdot \dot{\mathbf{B}} - \beta_4 \frac{1}{\alpha^2 \rho^2} \mathbf{M} \mathbf{M} \cdot \mathbf{M} - \\ &\quad \beta_5 \frac{1}{\alpha^2 \rho^2} \left(\frac{1}{\alpha^2 \rho^2} \mathbf{M} \mathbf{M} - \frac{1}{3} \boldsymbol{\delta} \right) : \mathbf{M} \mathbf{M} \mathbf{M} , \end{aligned} \quad (32)$$

and without the second order alignment tensor in the state space:

$$\begin{aligned} \frac{d\mathbf{M}}{dt} &= \beta_1 \alpha\rho \mathbf{B} + \beta_2 \alpha\rho \dot{\mathbf{B}} + \beta_4 \mathbf{M} \\ &- \beta_1 \frac{1}{\alpha\rho} \mathbf{M} \mathbf{M} \cdot \mathbf{B} - \beta_2 \frac{1}{\alpha\rho} \mathbf{M} \mathbf{M} \cdot \dot{\mathbf{B}} - \beta_4 \frac{1}{\alpha^2 \rho^2} \mathbf{M} \mathbf{M} \cdot \mathbf{M} , \end{aligned} \quad (33)$$

Although the state space included the orientation gradient of the mesoscopic mass density, in order to account for orientation diffusion, the corresponding term does not show up in the final macroscopic equation (β_3 does not appear). The reason is that integrating the \mathbf{n} -divergence over the (closed) unit sphere gives zero (see equation (24)).

If the value of the magnetization is small enough, we can neglect quadratic and higher order terms of the magnetization. In this linear limit the equations (32) and (33) simplify to

$$\begin{aligned} \frac{d\mathbf{M}}{dt} &= \beta_1 \alpha \rho \mathbf{B} + \beta_2 \alpha \rho \dot{\mathbf{B}} + \beta_4 \mathbf{M} - \frac{1}{3} \beta_5 \mathbf{M} \\ \frac{d\mathbf{M}}{dt} &= \beta_1 \alpha \rho \mathbf{B} + \beta_2 \alpha \rho \dot{\mathbf{B}} + \beta_4 \mathbf{M} \quad , \end{aligned} \quad (34)$$

respectively.

Both are of the form of the well known DEBYE equation (Debye, 1945). This fact can be used to identify the coefficients α_1 , α_2 , β_4 , and β_5 .

4 Conclusions

Starting with the mesoscopic balance equations, especially the balance of mass, and the balance of internal angular momentum in the stationary case, we derived a differential equation for the orientation distribution function of the dipoles. The first moment of this distribution function is proportional to the magnetization. The resulting equation for the magnetization is of relaxation type, and it is nonlinear, even in the simplest case of a state space. Its linear limit is of the form of the well-known DEBYE-equation.

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