

Macroscopic Dynamics of Ferronematics

E. Jarkova¹, H. Pleiner^{1*}, H.-W. Müller¹, and H.R. Brand²

¹ Max-Planck-Institute for Polymer Research, 55021 Mainz, Germany

² Theoret. Physik III, Universität Bayreuth, 95440 Bayreuth, Germany

J. Chem. Phys. 118, 2422 (2003)

Abstract

We derive the complete set of macroscopic dynamic equations for ferrofluids in the nematic liquid crystal phase under an external magnetic field. We include the magnetization as an independent dynamic degree of freedom. Special emphasis is laid on possible static and dynamic crosscouplings between magnetization, director reorientations and flow. As examples we discuss the field dependence of the sound spectrum and the rheology of shear flow.

PACS: 61.30.-v, 75.50.Mm, 05.70.Ln

1 Introduction

Ferronematics belong to a class of complex materials which stably incorporate nano-sized magnetic particles into a liquid crystalline matrix. The production of those or similar smart materials opens up the perspective for a variety of new technological developments. Indeed, nematic liquid crystals are interesting materials due to their optical birefringent and scattering properties, which are controllable by external electrical or magnetic fields or by shear stresses. In their seminal work Brochard and de Gennes¹ introduced the challenging idea to intensify the magnetic sensitivity of the orientational order parameter by doping the LC matrix with a small amount of ferromagnetic particles, i.e. with a ferrofluid.² Due to the strong surface anchoring of the magnetic grains on the

surrounding nematic matrix the susceptibility of the substance is raised by several orders of magnitude in comparison to pure nematics.³⁻⁵

Very soon after the first synthesis of ferrofluids² a variety of thermotropic, discotic and lyotropic ferronematics were reported.⁶⁻⁹ Particularly appropriate for experimental studies is a lyotropic micellar nematic system^{3,10,11} doped with an aqueous ferrofluid.^{12,13} The feature of liquid crystal transparency is an issue of considerable interest.¹⁴ Recently highly transparent lyotropic ferrofluid nematic liquid crystals have been reported.¹⁵

With the synthesis of the first thermotropic ferronematics⁴ it turned out that the orientation between the magnetization and the nematic ordering is not completely rigid. This led Burylov and Raikher¹⁶ to an approach which treats the direction of the (saturated) magne-

*email: pleiner@mpip-mainz.mpg.de

tization and the nematic director as independent variables (F-nematic, see below). In a recent thermodynamic consideration based on a Ginzburg-Landau description¹⁷ we worked out the various possible equilibrium structures of such a nematic ferrofluid system. They can undergo phase transitions from the isotropic phase (no order) either into the *superparamagnetic* (S) nematic phase (only nematic but no spontaneous magnetic order) or into the *ferromagnetic* (F) nematic phase (nematic and spontaneous magnetic order). In the latter case the magnetization is either parallel to an external magnetic field (with the nematic director parallel or perpendicular to it) or the three directions are all different but lie in the same plane. However, since there is as yet no experimental proof for the existence of an F-nematic phase, we will focus here on the usual S-case. We mention, however, that in an external magnetic field the dynamics of an F-nematic with the magnetization parallel to the field is quite similar to that of the S-type, since they differ only in the origin of the equilibrium magnetization. In the remainder of this paper we shall follow the usual convention and refer to the S-phase by the simple term *ferronematic*. If such a ferronematic is exposed to an external magnetic field the induced magnetization is always co-aligned, while the nematic director can be parallel¹⁸ or perpendicular³ to it. In the former case the equilibrium configuration the system is uniaxial. This case will be described in the main text below. The biaxial case, where the director is perpendicular to the external field, is only slightly more complicated and will be discussed briefly in Appendix A.

To derive the dynamic equations we use the hydrodynamic method. Hydrodynamics describes a system in the long wavelength limit and for long time scales. The hydrodynamic equations are derived by means of symmetry and thermodynamic arguments. The main advantage of the hydrodynamic method lies in its

generality, which allows its application to very different systems. However, the occurrence of phenomenological parameters in the static and dynamic expansions are the prize one has to pay for this generality. Therefore coefficients turning up in the equations below have to be determined by microscopic models or by experiments.

There are cases, where non-hydrodynamic, relaxing processes become so slow that their dynamics takes place on a macroscopic time scale as well. Then it is appropriate to also include non-hydrodynamic, but slowly relaxing variables in the dynamic description of such a system. However, there are no first principles to select those slowly relaxing variables and heuristic arguments have to be used instead. Such a phenomenological macroscopic description does not have the solid foundation of a truly hydrodynamic theory. On the other hand, by comparing the implications of this extended description with experiments, one can learn for which systems, and under what conditions, the inclusion of those non-hydrodynamic variables is required for a reasonable description.

In ferrofluids the magnetization (its orientation as well as its absolute value) relaxes to the equilibrium value set by the external field. The appropriate relaxation time is much larger than all microscopic time scales and can be relevant for the macroscopic dynamics, particularly in complicated lyotropic systems.¹⁹ In this case one should treat the magnetization as an additional dynamic variable with its own dynamical (relaxation) equation. For isotropic ferrofluids this has been done in²⁰ and within a hydrodynamic description quite recently in.²¹ In ferronematics the orientations of the director and the magnetization are linked in equilibrium, but the dynamics off equilibrium can be quite different: an external field reorients the magnetization quite easily due to the superparamagnetic effect, but is less effective regarding the director, which is only coupled to the

magnetic field by the diamagnetic anisotropy effect. Vice versa, an electric field strongly orients the director but not the orientation of the magnetization. Thus, it is easy to create situations, where the director and the magnetization are quite different and where they relax independently into their equilibrium orientation. We also keep the absolute value of the magnetization as a relevant variable, but not the degree of nematic order, since the latter is not specifically influenced by a magnetic field or by the magnetic degrees of freedom and, thus, behaves as in ordinary nematics (and is relevant near the phase transitions to the isotropic phase or to fluid smectic liquid crystalline phases, only).

For ferronematics hydrodynamic equations were derived²² in the limit that the magnetic degree of freedom has already relaxed to its equilibrium value. Concentrating on novel dynamic effects linear in the magnetic field, it was shown that flow alignment, heat conduction, diffusion, thermodiffusion, viscosity and director reorientation are all modified in the presence of an external field. The appropriate material tensors – relevant also for the present treatment – are listed in Appendix B. In²³ an extended description of electromagnetic effects in nematic liquid crystals has been given within the framework of generalized dissipative Maxwell equations. A non-hydrodynamic magnetic degree of freedom (a generalized dissipative magnetic field) is introduced, the dynamics of which can be compared to the dissipative part of our magnetization dynamics, while there is no counterpart in²³ to the reversible dynamics of an independent magnetization degree of freedom (see below).

Here we generalize the set of hydrodynamic equations for ferronematics (in a constant external magnetic field) by including the magnetization as an additional, slowly relaxing variable. Special emphasis is laid on the static (Sec.2) and dynamic (Sec.3) crosscouplings between the nematic and the magnetic degrees of

freedom. As examples for their relevance, and possible ways for measuring such effects, we discuss in more detail sound propagation and damping (Sec.4) as well as the rheology during shear flow (Sec.5).

2 Statics and thermodynamics

The macroscopic description of a system starts with the identification of the relevant variables. Apart from the quantities that are related to local conservation laws, like mass density (ρ), momentum density (\mathbf{g}), energy density (ε) and concentration (c) of the magnetic particles²⁴ or that are related to spontaneously broken continuous symmetries like reorientations of the director \mathbf{n} , we take, as discussed in the Introduction, the magnetization \mathbf{M} as slowly relaxing variable. To take into account Maxwell's equations the magnetic induction \mathbf{B} must be considered as well. According to the Eulerian description all variables are local fields, i.e. volume densities that depend on space and time. The director \mathbf{n} is a unit vector and due to the special nematic symmetry all equations have to be invariant under the replacement $\mathbf{n} \rightarrow -\mathbf{n}$. The magnetization $\mathbf{M} = M\mathbf{m}$ is associated with rotations $\delta\mathbf{m}$ as well as changes in the absolute value δM .

Assuming local thermodynamic equilibrium, i.e. all other, fast relaxing quantities are already in equilibrium, the Gibbs relation

$$d\varepsilon = Td\sigma + \mu d\rho + \mu_c dc + v_i dg_i + H_i dB_i + h_i^M dM_i + h_i^{n'} dn_i + \Phi_{ij} d(\nabla_j n_i) \quad (1)$$

connects the macroscopic variables to the entropy density σ . In eq.(1) the thermodynamic quantities: chemical potential (μ), temperature (T), relative chemical potential (μ_c), nematic molecular fields (Φ_{ij} , $h_i^{n'}$), velocity (v_i), magnetic Maxwell field (H_i) and the magnetic molecular field (h_i^M) are defined as par-

tial derivatives of the energy density with the respect to the appropriate variables.²⁵

In equilibrium the magnetization is generally a nonlinear function of the external field. This function is known from experiments²⁶ or from simulations.²⁷ The equilibrium value of the magnetization, \mathbf{M}_0 at a given external field, which may have a field independent part in case of true ferromagnetism, is an input parameter into our dynamic theory. In addition, we assume the equilibrium orientation of the director to be parallel to the field (and the equilibrium magnetization). The case of a perpendicular director alignment is discussed in Appendix A.

The statics, i.e. the relation between the conjugate quantities and the variables, is conveniently set up by providing an energy density function, usually in harmonic approximation (bilinear in the variables). We will do here the same for the magnetic part of the energy density and refer to Appendix C for a more general description. Thus $\varepsilon = \varepsilon_n + \varepsilon_M$ with the magnetic energy density

$$\varepsilon_M(\mathbf{B}, \mathbf{M}) = \frac{B^2}{2} - \mathbf{M} \cdot \mathbf{B} + \frac{1}{2}A_1(\mathbf{n} \cdot \mathbf{M})^2 + \frac{1}{2}A_2\mathbf{M}^2 \quad (2)$$

and ε_n the energy density of a conventional nematic liquid crystal²⁵ (without magnetization and external field). The term $\frac{1}{2}A_1(\mathbf{n} \cdot \mathbf{M})^2$ describes the coupling between the nematic director and the magnetic particles. This strong interaction was observed in a series of experiments³⁻⁵ studying the Fredericks transition in ferronematics. For $\mathbf{n} \parallel \mathbf{M}$ being the energy minimum, A_1 has to be negative. Generally the coefficients $A_{1,2}$ are functions of the state variables, like temperature, pressure etc., but also of \mathbf{M}_0^2 (or the external field strength). Only for a linearized description, valid for small deviations from equilibrium, are the coefficients constant.

Using eq.(2), the magnetic Maxwell field H_i

is defined in the usual way

$$H_i = \left(\frac{\partial \varepsilon}{\partial B_i} \right)_{\mathbf{M}, \mathbf{n}, \dots} = B_i - M_i, \quad (3)$$

while the magnetic molecular field h_i^M reads

$$\begin{aligned} h_i^M &= \left(\frac{\partial \varepsilon}{\partial M_i} \right)_{\mathbf{B}, \mathbf{n}, \dots} \\ &= -B_i + A_1 n_i n_j M_j + A_2 M_i \end{aligned} \quad (4)$$

Note that because of the definition (3), it is not possible to have a direct coupling between the external field \mathbf{B} and the director; the field orientation of the director is mediated by the magnetization via the term $\sim A_1$.

In equilibrium h_i^M has to be zero and $B_i = A_1 n_i n_j M_j + A_2 M_i$ results. With that solution the magnetic energy density (2) can be expressed as $\varepsilon_M(\mathbf{B}) = \frac{1}{2} H_i(\mathbf{B}) B_i$. In the regime of linear magnetism or if linearized for small deviations from equilibrium, one can use instead the usual magnetic susceptibility tensors, defined by $B_i = \mu_{ij} H_j$ and $M_i = \chi_{ij} H_j$, which both have the uniaxial form $\mu_{ij} = \mu_{\perp} \delta_{ij}^{tr} + \mu_{\parallel} n_i n_j = \mu_{\perp} \delta_{ij} + \mu_a n_i n_j$ (where $\delta_{ij}^{tr} = \delta_{ij} - n_i n_j$ is the transverse Kronecker symbol). Comparison with the equilibrium solution of (4) gives $A_1 = \mu_{\parallel} / \chi_{\parallel} - \mu_{\perp} / \chi_{\perp}$ and $A_2 = \mu_{\perp} / \chi_{\perp}$. Written in this way the magnetic energy density (2) obtains the familiar form $\varepsilon_M(\mathbf{B}) = \frac{1}{2} \mu_{ij} H_i H_j$. In the general case, again, the magnetic susceptibilities are not constants, but depend on the magnetization or the external field.

In ordinary nematic liquid crystals instead of $\varepsilon_M(\mathbf{B})$ usually the Legendre-transformed energy $\varepsilon_M(\mathbf{H}) = \varepsilon_M(\mathbf{B}) - \mathbf{B} \cdot \mathbf{H}$ is used, for which we get the well-known form $2\varepsilon_M(\mathbf{H}) = -\chi_a(\mathbf{n} \cdot \mathbf{H})^2 - (\chi_{\perp} - 1)\mathbf{H}^2$ where very often the vacuum field energy (\mathbf{H}^2) is omitted.²⁹

The molecular field $h_i^{n'}$

$$h_i^{n'} = \delta_{ij}^{tr} \left(\frac{\partial \varepsilon}{\partial n_j} \right)_{\mathbf{M}, \mathbf{B}, \dots} = A_1 \delta_{ij}^{tr} M_j M_k n_k \quad (5)$$

and Φ_{ij} that follows from the gradient part of the energy density²⁸

$$\Phi_{ij} = \left(\frac{\partial \varepsilon}{\partial \nabla_j n_i} \right)_{\mathbf{M}, \mathbf{B}, \dots} = K_{ijkl} \nabla_k n_l, \quad (6)$$

can be combined to the field

$$h_i^n = \frac{\delta}{\delta n_i} \int \varepsilon dV = h_i^{n'} - \nabla_j \Phi_{ij}. \quad (7)$$

Since \mathbf{n} is a unit vector that can only rotate, $n_i h_i^{n'} = 0$, which is ensured in (5) by the transverse Kronecker symbol. In equilibrium, $\mathbf{n} = \text{const.}$ and $\delta_{ij}^{tr} M_j = 0$, hence $h_i^n = 0$.

3 Dynamics

The hydrodynamic equations for conserved, broken-symmetry and slowly relaxing variables are

$$\frac{\partial}{\partial t} \rho + \text{div } \rho \mathbf{v} = 0 \quad (8)$$

$$\frac{\partial}{\partial t} \sigma + \text{div } \sigma \mathbf{v} + \text{div } \mathbf{j}^\sigma = \frac{R}{T} \quad (9)$$

$$\frac{\partial}{\partial t} g_i + \nabla_j (v_j g_i + \delta_{ij} p + \sigma_{ij}^{th} + \sigma_{ij}) = 0 \quad (10)$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j \right) n_i + (\mathbf{n} \times \boldsymbol{\omega})_i + Y_i = 0 \quad (11)$$

$$\rho \left(\frac{\partial}{\partial t} + v_j \nabla_j \right) c + \text{div } \mathbf{j}^c = 0 \quad (12)$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j \right) M_i + (\mathbf{M} \times \boldsymbol{\omega})_i + X_i = 0 \quad (13)$$

with

$$\begin{aligned} \sigma_{ij}^{th} &= -B_j H_i - \frac{1}{2} (M_j h_i^M - M_i h_j^M) \\ &\quad - \frac{1}{2} (n_j h_i^n - n_i h_j^n) + \Phi_{kj} \nabla_i n_k \end{aligned} \quad (14)$$

where $\omega_i = \frac{1}{2} \epsilon_{ijk} \nabla_j v_k$ is the vorticity. The thermodynamic pressure p is given by

$$p = -\varepsilon + T\sigma + \mu\rho + \mathbf{g} \cdot \mathbf{v} + \mathbf{B} \cdot \mathbf{H}. \quad (15)$$

The parts of the currents shown explicitly in (8-14) are not material dependent, but are

given by general symmetry and thermodynamic principles,²⁵ like transformation behavior under translations (convective terms) or rotations (e.g. $\dots \times \boldsymbol{\omega}$) and by the requirement of zero entropy production ($R = 0$).

Using the relation²⁵ $\Omega_{ij}(-M_i B_j + h_i^M M_j + h_i^n n_j + \Phi_{ki} \nabla_j n_k + \nabla_k (n_j \Phi_{ik})) = 0$, which is valid for any constant antisymmetric matrix $\Omega_{ij} = -\Omega_{ji}$ and which follows from the fact that the energy density (1) has to be invariant under constant rotation, eq.(14) can be transformed into

$$\begin{aligned} \sigma_{ij}^{th} &= -\frac{1}{2} (B_j H_i + B_i H_j) + \frac{1}{2} (\Phi_{ki} \nabla_j n_k \\ &\quad + \Phi_{kj} \nabla_i n_k) + \frac{1}{2} \nabla_k (n_j \Phi_{ik} - n_i \Phi_{jk}). \end{aligned} \quad (16)$$

Here the antisymmetric part has the form of a divergence, which ensures angular momentum conservation. It can be brought into a manifestly symmetric form by some redefinitions.²⁸

The phenomenological parts of the entropy current j_i^σ , the stress tensor σ_{ij} , the concentration current j_i^c and the quasi-currents Y_i and X_i associated with the temporal changes of the director and the magnetization, respectively, are given below. The source term R/T in (9) is the entropy production, which is zero for reversible and positive for irreversible processes. The phenomenological part of the stress tensor σ_{ij} has to be symmetric guaranteeing angular momentum conservation.

Since we are not dealing with electromagnetic effects, we can use the static Maxwell equations to determine \mathbf{B}

$$\text{curl } \mathbf{H} = \text{curl } (\mathbf{B} - \mathbf{M}) = 0, \quad \text{div } \mathbf{B} = 0. \quad (17)$$

The dynamic equation for the energy density follows from (8-13) via (1) and is not shown here.

The phenomenological currents written down in eq.(8-13) can be split into dissipative (superscript D) and into reversible (superscript R) parts, depending on whether they

give rise to a finite amount of dissipation ($R > 0$) or to a zero entropy production ($R = 0$). Using general symmetry and invariance arguments and the fact, that a magnetic field changes sign under time reversal, we obtain

$$j_i^{\sigma R} = -\kappa_{ij}^R(M)\nabla_j T - D_{ij}^{TR}(M)\nabla_j \mu_c \quad (18)$$

$$j_i^{cR} = -D_{ij}^R(M)\nabla_j \mu_c - D_{ij}^{TR}(M)\nabla_j T \quad (19)$$

$$\sigma_{ij}^R = -\frac{1}{2}\lambda_{kji}h_k^n - c_{kij}^R(M)h_k^M - \nu_{ijkl}^R(M)A_{kl} \quad (20)$$

$$Y_i^R = -\frac{1}{2}\lambda_{ijk}A_{jk} + (\gamma^{-1})_{ij}^R(M)h_j^n + \chi^R(\mathbf{n} \times \mathbf{h}^M)_i \quad (21)$$

$$X_i^R = b_{ij}^R(M)h_j^M + \chi^R(\mathbf{n} \times \mathbf{h}^n)_i - c_{ijk}^R(M)A_{jk} \quad (22)$$

with $A_{ij} = \frac{1}{2}(\nabla_i v_j + \nabla_j v_i)$. The magnetization-dependent tensors $\kappa_{ij}^R(M)$, $D_{ij}^R(M)$, $D_{ij}^{TR}(M)$, $(\gamma^{-1})_{ij}^R(M)$, $b_{ij}^R(M)$, $\nu_{ijkl}^R(M)$, $c_{ijk}^R(M)$ are all odd functions of the magnetization (see Appendix B (B.1-B.3)). For the case of \mathbf{M} already relaxed to its equilibrium value, similar tensors exist with \mathbf{M} replaced by the external field (except for b_{ij}^R and c_{ijk}^R). Their physical meaning has been discussed in.²²

Due to the new degree of freedom (magnetization) there is an additional term in the nematic quasi-current eq.(21) $\chi^R(\mathbf{n} \times \mathbf{h}^M)_i$ and a counter term in X_i^R , which describes a dynamic cross-coupling between magnetization and nematic director. It does not exist in ordinary nematics nor in isotropic ferrofluids. Its physical meaning (together with b_{ij}^R and c_{ijk}^R) will be explored in the Secs.(4,5). The flow alignment tensor λ_{ijk} has the usual form²⁸ $\lambda_{ijk} = \lambda(\delta_{ij}^{tr}n_k + \delta_{ik}^{tr}n_j)$.

For the derivation of the dissipative parts of the phenomenological currents one usually expands the dissipation function R to second order in the thermodynamic forces and then obtains the dissipative currents by taking the variational derivatives with respect to

the forces. We find for the dissipation function

$$\begin{aligned} R = & \frac{1}{2}\kappa_{ij}(\nabla_i T)(\nabla_j T) + \frac{1}{2}\nu_{ijkl}A_{ij}A_{kl} \\ & + \lambda_{ijk}^D(M)h_i^n A_{jk} + \frac{1}{2}D_{ij}(\nabla_i \mu_c)(\nabla_j \mu_c) \\ & + \frac{1}{2}b_{ij}^D h_i^M h_j^M + c_{ijk}^D h_i^M A_{jk} + \chi_{ij}^D(M)h_j^M h_i^n \\ & + \frac{1}{2\gamma_1}h_i^n \delta_{ij}^{tr}h_j^n + D_{ij}^T(\nabla_j T)(\nabla_i \mu_c) \quad (23) \end{aligned}$$

Here ν_{ijkl} is the uniaxial viscosity tensor²⁸ and κ_{ij} , D_{ij} and D_{ij}^T describe heat conduction, diffusion and thermodiffusion, respectively. Director diffusion or relaxation is given by one coefficient γ_1 , while magnetization relaxation b_{ij}^D contains a transverse (rotational) and a longitudinal (absolute value) coefficient $b_{ij}^D = b_{\perp}^D \delta_{ij}^{tr} + b_{\parallel}^D n_i n_j$. All the 2nd rank tensors mentioned above have this form. The magnetization-dependent tensors are odd functions of M and listed in Appendix B (B.1-B.3). The meaning of λ_{ijk}^D has been discussed in,²² while χ_{ij}^D will show up in Sec.(5). The 3rd rank tensor c_{ijk}^D again is specific to nematic ferrofluids and contains one coefficient $c_{ijk}^D = c^D(\epsilon_{imk}n_m n_j + \epsilon_{imj}n_m n_k)$ describing a dissipative coupling between flow and orientation of the magnetization.

The range of possible values of the coefficients in eq.(23) is restricted by the positivity of the entropy production.

The dissipative parts of the currents then read

$$j_i^{\sigma D} = -\kappa_{ij}\nabla_j T - D_{ij}^T\nabla_j \mu_c \quad (24)$$

$$j_i^{cD} = -D_{ij}\nabla_j \mu_c - D_{ij}^T\nabla_j T \quad (25)$$

$$\sigma_{ij}^D = -\nu_{ijkl}^D A_{kl} - \lambda_{kij}^D(M)h_k^n - c_{kij}^D h_k^M \quad (26)$$

$$\begin{aligned} Y_i^D = & \frac{1}{\gamma_1}\delta_{ij}^{tr}h_j^n + \chi_{ij}^D(M)h_j^M \\ & + \lambda_{ijk}^D(M)A_{jk} \quad (27) \end{aligned}$$

$$X_i^D = b_{ij}^D h_j^M + \chi_{ji}^D(M)h_j^n + c_{ijk}^D A_{jk} \quad (28)$$

This set of equations can be compared with the dissipative dynamics given in²³ for

the magnetic and nematic degrees of freedom. Interpreting $-H_i^D$ as our current X_i^D and $c \text{curl} \mathbf{E}^0$ as our conjugate \mathbf{h}^M , Eq.(13) of²³ is compatible with eqs. (26-28) in the uniaxial case ($\mathbf{n} \parallel \mathbf{m}$ in equilibrium). There is however no counterpart in²³ to the reversible dynamics of eqs.(20-22).

4 Propagation of sound

In this section we derive the longitudinal eigenmodes (sound) of the system with the nematic director \mathbf{n}^0 parallel to the magnetic field in equilibrium. In ordinary nematics the sound velocity is isotropic and does not depend on the external field, since the nematic orientational fluctuations do not couple to sound. Only sound damping is anisotropic due to the anisotropy of viscosity and heat conduction. In nematic ferrofluids with the magnetization relaxed to its equilibrium value \mathbf{M}_0 , sound is accompanied by transverse forces,²² but the sound velocity is still isotropic and field-independent. Here we focus on the effect of the new variable (magnetization) on the sound spectrum. Thus, we can neglect all diffusional processes connected e.g. with viscosity and heat conduction as well as their reversible counterparts. Only the relaxations of the director and the magnetization in the field are kept. The relevant linearized static fields are then

$$h_i^{n'} = A_1 M_0 \delta M_i - A_1 M_0^2 \delta n_i \quad (29)$$

$$h_i^M = -\delta B_i + A_1 M_0 \delta n_i + A_2 \delta M_i + A_1 n_i^0 n_j^0 \delta M_j \quad (30)$$

with n_i^0 the unit vector along the field. Assuming a one-dimensional plane wave the space-time dependence is $\sim \exp i(-\omega t + \mathbf{k} \cdot \mathbf{r})$ for all deviations from equilibrium and the linearized set of dynamic equations becomes an algebraic one. Even then the full system of equations is rather complicated in the case of ferronemat-

ics. It can be simplified by looking at particular geometries.

Let us consider sound in the case where the external magnetic field, the equilibrium magnetization, and the director are perpendicular to the wave vector. Field fluctuations δB_i are fixed by the static Maxwell equations (17) to $\delta B_i = \delta M_j (\delta_{ij} - k_i k_j k^{-2})$. However, since $A_2 \gg 1$ (because of $\chi_{\perp} \ll 1$), this contribution can safely be neglected in (30). Then there are only three relevant variables left, the density ρ , the longitudinal velocity component ($\mathbf{v} \parallel \mathbf{k}$) and the (longitudinal) component $\delta \mathbf{M} \parallel \mathbf{B}$, which is transverse to \mathbf{k} , changing the sound dispersion relation into

$$\frac{\omega^2}{k^2} - c_0^2 = \frac{\mu_{\parallel}}{\rho} (c_3^R + c_4^R)^2 \chi_{\parallel} H^2 \frac{i\omega}{i\omega - \tau} \quad (31)$$

with c_0 the usual adiabatic sound velocity and $\tau = b_{\parallel}^D \mu_{\parallel} / \chi_{\parallel}$ the relaxation time of the longitudinal magnetization. Here B^2 has been replaced by $(\chi_{\parallel} H)^2$ and $A_1 + A_2$ by $\mu_{\parallel} / \chi_{\parallel}$. The coefficient c_3^R (B.2) also exists in isotropic ferrofluids. Thus, this change in the sound dispersion relation is possible in the isotropic phase as well.³⁰

If the sound period is much bigger than the inverse relaxation time of the magnetization $k c_0 \gg 1/\tau$, there is a field dependent addition to the sound velocity, while in the opposite, realistic case $k c_0 \ll 1/\tau$, there is a field-dependent sound damping due to magnetization relaxation

$$\Im \mathbf{m}(\omega) = -\frac{1}{2} \frac{(c_3^R + c_4^R)^2}{\rho b_{\parallel}^D} \chi_{\parallel}^2 H^2 k^2 \quad (32)$$

that comes in addition to the usual magnetic-field-independent sound damping (not shown here). This effect can be used to measure the combination $c_3^R + c_4^R$ of new coefficients by varying the external field. In the case of sound waves along the field direction ($\mathbf{k} \parallel \mathbf{B}$) the situation is quite similar, only a different combination of transport parameters is involved, i.e. in (31,32) $c_3^R + c_4^R$ has to be replaced by

$c_1^R + 2c_2^R + c_3^R + c_4^R + 2c_5^R + c_6^R$. A completely different situation arises, when the sound is oblique to the field \mathbf{B} (i.e. \mathbf{k} has an angle different from zero or π with \mathbf{B}). Then the longitudinal variables $div\mathbf{v}$, $\delta\rho$, and $\mathbf{B} \cdot \delta\mathbf{M}$ are coupled to the transverse ones ($curl\mathbf{v}$, $\delta_{ij}^{tr}\delta M_j$, δn_i) mainly through the existence of the tensor $c_{ijk}^R(M)$ (20, 22, B.2). This has the consequence that shear flow is connected to sound and vice versa, and if one excites one of them the other is excited as well. A similar phenomena for isotropic ferrofluids is discussed in.³⁰

The picture is qualitatively similar for the biaxial case ($\mathbf{n} \perp \mathbf{B}$). When the sound is along the preferred directions (either parallel to \mathbf{n} , to \mathbf{B} or perpendicular to both) the sound dispersion gets an extra field dependent damping similar to (32), while in the oblique case again sound is coupled to shear flow.

5 Rheology

To evaluate the influence of the static and dynamic coupling between the nematic degree of freedom and the magnetization we investigate the stress induced by applying a shear flow that varies periodically in time. To make the problem simpler we consider the particular geometry, where the equilibrium orientation of the nematic director and the magnetization are perpendicular to the flow direction and along the gradient direction. The linear response of the system that contains information on the material properties is described by the frequency dependent response function $G(\omega)$, which is defined as the ratio of the induced stress (say σ_{xz}) to the applied "strain" $e_{xz} \equiv iA_{xz}/\omega$

$$G(\omega) = \frac{\sigma_{xz}}{e_{xz}} = G'(\omega) + iG''(\omega), \quad (33)$$

which has poles at the frequencies of those eigenmodes that couple to shear flow. The real part G' (imaginary part G'') describes the reversible, in-phase (irreversible, out-of-phase)

response. When calculating $G(\omega)$ the flow is assumed to show the undisturbed, externally imposed linear profile.

The relaxation process of the magnetization to its equilibrium value is fast in comparison to the nematic director relaxation/diffusion time. This gives the possibility to investigate the influence of the nematic-magnetization couplings on the two modes separately. Since the slow nematic mode seems to be more convenient for experiments, we will concentrate on this mode in the following. We consider the situation without an external magnetic field first and then the much more complicated case with a field (along the gradient direction). This probes the coupled dynamics of the transverse variables ($curl\mathbf{v}$, $\delta_{ij}^{tr}\delta M_j$, δn_i).

The response function without magnetic field has the following form

$$\frac{G'}{\omega^2} = \frac{C\Gamma}{\omega^2 + \Gamma^2} \quad (34)$$

$$\frac{G''}{\omega} = \nu_3 - \frac{\xi_1}{q_1} + \frac{C\omega^2}{\omega^2 + \Gamma^2}, \quad (35)$$

with

$$\xi_1 = \frac{1 - \lambda^2}{4} + \frac{(c^D)^2}{b_{\perp}^D \gamma_1} - \frac{c^D \chi^R \lambda}{b_{\perp}^D} \quad (36)$$

$$q_1 = \frac{1}{\gamma_1} + \frac{(\chi^R)^2}{b_{\perp}^D} \quad (37)$$

$$C = \frac{\xi_1}{q_1} - \frac{(c^D)^2}{b_{\perp}^D} = \frac{1}{q_1} \left(\frac{1}{4} - \left[\frac{\lambda}{2} + \frac{c^D \chi^R}{b_{\perp}^D} \right]^2 \right) \quad (38)$$

$$\Gamma = q_1 K_3 k^2 \quad (39)$$

Eqs.(34,35) are, as discussed above, only applicable for low frequencies with $\omega \ll A_2 b_{\perp}^D$. In the loss modulus G'' the apparent Newtonian shear viscosity differs from the bare shear viscosity ν_3 by $-\xi_1/q_1$ where $1/q_1$ (37) is the renormalized nematic orientational viscosity that deviates from γ_1 due to the reversible

dynamic crosscoupling (χ^R) between director and magnetization (21,22), and the transverse magnetization relaxation b_{\perp}^D . The new contribution $\sim \xi_1$ is due to the relaxation of the magnetization (b_{\perp}^D) and of the director (γ_1), as well as due to the dissipative crosscoupling between flow and magnetization (c^D) and the reversible crosscouplings of the director with flow (λ) and magnetization (χ^R). This change in the apparent viscosity is absent in isotropic ferrofluids, but present in ordinary nematics (for $\lambda \neq \pm 1$). However, since ferronematics, ordinary nematics and isotropic ferrofluids are different phases with generally different (bare) viscosities, the additional change of the viscosity due the magnetization degree of freedom is only a quantitative effect and probably difficult to observe. There are, however, magnetic oscillations (around $M = 0$) induced by the periodic shear flow (directly via c^D) or mediated

by director oscillations (via χ^R and λ). The oscillating magnetization points out of the shear plane with a peak amplitude that has its maximum for $\omega \approx \Gamma$. Such magnetic oscillations can be measured by a Hall probe.

Qualitatively new, compared to isotropic ferrofluids, is the occurrence of a director diffusional mode in G' and G'' with amplitude C (38) and width Γ (39). The amplitude C shows that, even without a magnetic field, the magnetization as an independent variable with its own dynamics influences the relaxation process of the director. However this mode has its strongest influence at $\omega \approx \Gamma$, which is very low, if the wave vector k is small, and it may be hard to detect experimentally. Therefore we also study the case with an external field, where not only the relevant frequencies are shifted to higher values, but where also another, qualitatively new behavior is seen. Indeed we find

$$\frac{G'}{\omega} = \frac{1}{2Mq_2} \left(\frac{\Omega^2 \xi_1 - \Omega M q_2 \xi_2 + q_1^2 \xi_1}{(\Omega - q_2 M)^2 + (q_1 - M^2 q_3)^2} - \frac{\Omega^2 \xi_1 + \Omega M q_2 \xi_2 + q_1^2 \xi_1}{(\Omega + q_2 M)^2 + (q_1 - M^2 q_3)^2} \right) \quad (40)$$

$$\begin{aligned} \frac{G''}{\omega} = \nu_3 - \frac{1}{2Mq_2} & \left(\frac{\Omega^3 \xi_3 - \Omega^2 M \xi_3 q_2 + \Omega q_1^2 \xi_3 + M q_2 q_1 \xi_1}{(\Omega - q_2 M)^2 + (q_1 - M^2 q_3)^2} \right. \\ & \left. - \frac{\Omega^3 \xi_3 + \Omega^2 M \xi_3 q_2 + \Omega q_1^2 \xi_3 - M q_2 q_1 \xi_1}{(\Omega + q_2 M)^2 + (q_1 - M^2 q_3)^2} \right) \end{aligned} \quad (41)$$

where $\Omega \equiv \omega / (K_3 k^2 + \chi_a H^2)$ is the scaled frequency and $M = \chi_{\parallel} H$. The abbreviations $q_{2,3}$

$$\begin{aligned} q_2 &= \frac{1}{\gamma_1^R} + \frac{1}{\gamma_2^R} + \frac{2\chi_2^D \chi^R}{b_{\perp}^D} \\ &- \frac{(\chi^R)^2}{(b_{\perp}^D)^2} (b_1^R + b_2^R) \end{aligned} \quad (42)$$

$$q_3 = \frac{1}{(b_{\perp}^D)^3} (\chi_2^D b_{\perp}^D - \chi^R (b_1^R + b_2^R))^2 \quad (43)$$

are combinations of the various static susceptibilities and the transport parameters involved, while $\xi_2 = \xi_1 + q_1 \xi_3$, and $\xi_3 = (c^D)^2 / b_{\perp}^D$ with ξ_1 and q_1 given in (36,37), respectively. Since

we concentrate on the frequency dependence here, we have suppressed some additional M^2 -dependences of ξ_{1-3} and q_2 , which would render the results (40,41) extremely involved.

In the presence of an external field the relaxation mode is much more complicated than without a field. The most important, and experimentally most easily detectable feature is the shift of the nematic pole to finite frequencies $\Omega = \pm q_2 M$ (cf. Fig.1). The existence of q_2 is characteristic for nematic ferrofluids, since neither the reversible coupling between director and magnetization rotations (χ^R), nor the reversible counterparts to direc-

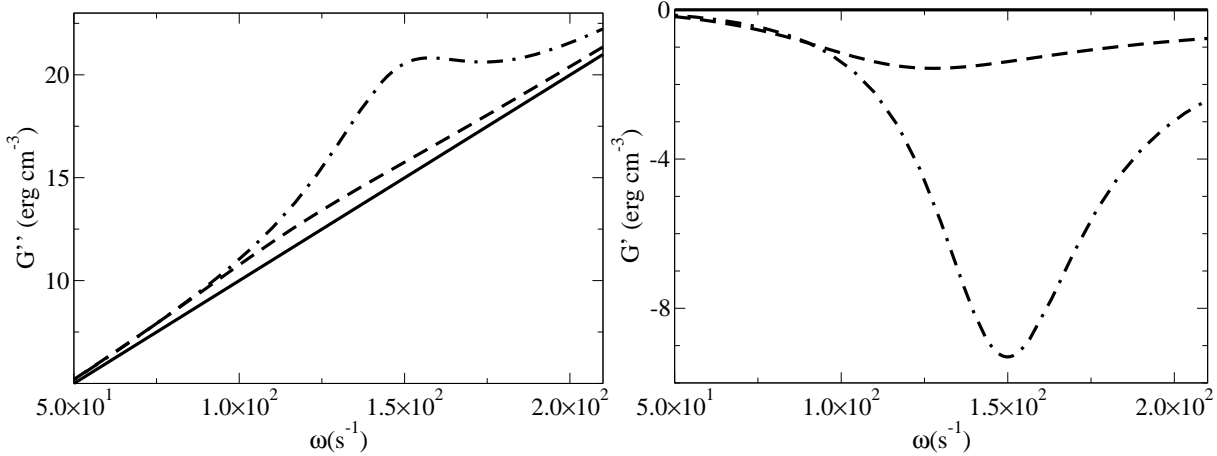


Figure 1: G'' (left) and G' (right) as functions of the frequency for increasing magnetic field strengths: $H = 0$ (solid), $H = 320G$ (dashed), and $H = 350G$ (dash-dotted); in our rationalized Gaussian unit system (cf. eq.(2)) $1G^2 = (1/4\pi)erg\ cm^{-3}$. The values of the parameters were taken to be $\gamma_1 = 10^{-1} g\ s^{-1}cm^{-1}$, $K_3 = 10^{-6} dyn$, $k = 100cm^{-1}$, $\nu_3 = 10^{-1} g\ s^{-1}cm^{-1}$, $\chi_a = 10^{-4}$, $\chi_{\parallel} = 10^{-4}$, $\chi^R = 3\ s^{-1}G^{-1}$, $\chi_2^D = 3\ cm\ s\ g^{-1}$, $b_{\perp}^D = 1\ s^{-1}$, $b_1^R + b_2^R = 40\ G^{-1}s^{-1}$, $c^D = 7 \times 10^{-3} G$, $(1/\gamma_1^R) + (1/\gamma_2^R) = 1\ s\ cm\ g^{-1}G^{-1}$, and $\lambda = 1$ (to eliminate ordinary nematic effects). On this scale the zero field effects (34,35) can hardly be seen.

tor diffusion/relaxation ($\gamma_{1,2}^R$) and magnetization relaxation ($b_{1,2}^R$) exist in ordinary nematics or isotropic ferrofluids. For the true frequency ω the shift increases $\sim H^3$ for fields larger than $\sqrt{K_3 k^2 / \chi_a}$. In an oscillating Couette shear experiment, under the approximation of a linear velocity profile, this behavior can be measured. There is also the possibility that the

nematic orientational mode becomes propagating for some intermediate fields strengths, if $q_2 M > q_1 - M^2 q_3$. As in the field-free case the oscillating shear flow induces also magnetization oscillations, but with a field there are oscillating components not only out of the shear plane, but also along the flow direction.

6 Conclusion

We derive the complete set of macroscopic dynamic equations for ferronematics introducing the magnetization as an independent slowly relaxing variable. We show that orientational changes of the magnetization are coupled to nematic director reorientations not only in the statics, but in the dynamics as well. In addition, there are reversible and dissipative dynamic crosscouplings between (compressional, shear and elongational) flow, (rotations and changes of the absolute value of the) magnetization and director reorientations. Some of

these couplings are only possible, when a finite magnetization is present due to spontaneous magnetic order and/or due to an external magnetic field. In order to measure some combinations of the parameters that describe these crosscouplings we study the sound wave spectrum and the rheology of shear flow. There is an additional field dependent contribution to sound damping due to such a crosscoupling. For the case of a sound wave propagating in a direction oblique to the preferred directions (set by equilibrium magnetization, nematic director, or perpendicular to them) compressional flow (and changes of the absolute value

of the magnetization) are coupled to shear flow (and rotations of the director and the magnetization). In addition we discussed the linear response of the system to oscillatory shear flow concentrating on frequencies below the transverse magnetization relaxation frequency. This allows to describe the influence of the magnetic dynamic degree of freedom on the nematic director diffusion/relaxation mode for ferronematic systems and the influence of the magnetic field on it. Even without a magnetic field the apparent viscosity is different from the bare one and the modified nematic director diffusion couples to the flow response. In the presence of an external field the director diffusion/relaxation is shifted to a finite frequency, which approximately increases with the third power of the field strength. Even in the field-free case there are shear-flow-induced oscillations of the magnetization out of the shear plane.

Acknowledgements

E.J. wants to thank V. Berejnov for fruitful email discussion. Partial support of this work through the Schwerpunktsprogramm 1104 'Colloidal Magnetic Fluids' of the Deutsche Forschungsgemeinschaft is gratefully acknowledged.

Appendix A Biaxiality

In this Appendix we deal with the case that in equilibrium the nematic director \mathbf{n}^0 is perpendicular to the external field \mathbf{B} (and the magnetization \mathbf{M}_0). This case is often realized in lyotropic systems.³ In the plane perpendicular to the field the direction of \mathbf{n} is not specified, i.e. the rotational in-plane symmetry is spontaneously broken and the system is biaxial. Thus, there is one rotation of \mathbf{n} (the in-plane rotation) that is hydrodynamic (diffusive), while the out-of-plane rotation is re-

laxing (non-hydrodynamic) due to the external field. This difference to the uniaxial case discussed in the main body of this manuscript (where both rotations of \mathbf{n} are relaxing) becomes apparent in the statics, i.e. in the magnetic energy of the system and hence in the molecular fields h_i^M and $h_i^{n'}$.

The static eqs.(2-7) still apply, however with $A_1 > 0$ ensuring $\mathbf{n}^0 \perp \mathbf{M}_0$. The linearized expressions (4,5) for $h_i^{n'}$ and h_i^M now only contain the out-of-plane components of \mathbf{n} ($M_k \delta n_k$), while the in-plane component appears in the gradient part Φ_{ij} only.

The form of the dynamic equations is not affected. Only the material tensors have a more complicated form (containing more coefficients generally) due to the lower orthorhombic symmetry. We will give the biaxial form of the field-free material tensors, while those that have to contain an odd number of factors \mathbf{M} are given in Appendix B (for the uniaxial as well as the biaxial case). In the biaxial case we have to discriminate between the directions of the director and the magnetization, which we denote by its unit vector $\mathbf{m} = \mathbf{M}/M$. The form of the material tensors of usual biaxial nematics is given in³¹ and is contained below for convenience. The symmetric 2nd rank tensors, like the heat conduction tensor κ_{ij} , the diffusion tensor D_{ij} , the thermodiffusion tensor D_{ij}^T , and magnetic relaxation tensor b_{ij}^D , have 3 independent coefficients

$$\kappa_{ij} = \kappa_{\parallel} n_i n_j + \kappa_{\perp} \delta_{ij}^3 + \kappa_3 m_i m_j \quad (\text{A.1})$$

where $\delta_{ij}^3 = \delta_{ij} - n_i n_j - m_i m_j$.

For the third order tensors

$$\begin{aligned} c_{ijk}^D &= c_1^D (\epsilon_{imk} n_m n_j + \epsilon_{imj} n_m n_k) \\ &+ c_2^D (\epsilon_{imk} m_m m_j + \epsilon_{imj} m_m m_k) \end{aligned} \quad (\text{A.2})$$

$$\begin{aligned} \lambda_{ijk} &= \lambda_1 (\delta_{ij}^3 n_k + \delta_{ik}^3 n_j) \\ &+ \lambda_2 (m_i m_j n_k + m_i m_k n_j) \end{aligned} \quad (\text{A.3})$$

there is one coefficient more than in the uniaxial case, each. The orientational diffusion

and relaxation of the director is governed by 1 coefficient each (cf. eq.(23))

$$R = \frac{1}{2} \left(\frac{1}{\gamma_1} \delta_{ij}^3 + \frac{1}{\gamma_3} m_i m_j \right) h_i^n h_j^n + \dots \quad (\text{A.4})$$

and the viscosity tensor has four additional coefficients

$$\begin{aligned} \nu_{ijkl} = & \nu_1 m_i m_j m_k m_l + \nu_2 n_i n_j n_k n_l + \nu_3 \delta_{ij}^3 \delta_{kl}^3 \\ & + \nu_4 (m_k m_l n_i n_j + m_i m_j n_k n_l) \\ & + \nu_5 (m_k m_l \delta_{ij}^3 + m_i m_j \delta_{kl}^3) \\ & + \nu_6 (n_k n_l \delta_{ij}^3 + n_i n_j \delta_{kl}^3) \\ & + \nu_7 (m_j m_l n_i n_k + m_j m_k n_i n_l \\ & \quad + m_i m_k n_j n_l + m_i m_l n_j n_k) \\ & + \nu_8 (m_j m_l \delta_{ik}^3 + m_j m_k \delta_{il}^3 \\ & \quad + m_i m_k \delta_{jl}^3 + m_i m_l \delta_{jk}^3) \\ & + \nu_9 (n_j n_l \delta_{ik}^3 + n_j n_k \delta_{il}^3 \\ & \quad + n_i n_k \delta_{jl}^3 + n_i n_l \delta_{jk}^3) \end{aligned} \quad (\text{A.5})$$

The reversible dynamic coupling between director and magnetization is still given by only one coefficient χ^R (21,22), but now contains only the out-of-plane components $Y_i^R m_i$ or $h_i^n m_i$ and $X_i^R \delta_{ij}^3$ or $h_i^M \delta_{ij}^3$.

Appendix B Tensors linear in \mathbf{M}

In this Appendix we list those tensors that contain an odd number of factors \mathbf{M} . For simplicity we restrict ourselves to the linear case of one \mathbf{M} factor. We do not specify the relative orientation of \mathbf{M} with \mathbf{n} , so these forms can be used for the uniaxial ($\mathbf{n} \parallel \mathbf{M}$ in equilibrium) as well as the biaxial case ($\mathbf{n} \perp \mathbf{M}$ in equilibrium). The reversible second rank magnetization-dependent material tensors, like the reversible analogues of heat conduction $\kappa_{ij}^R(M)$, diffusion $D_{ij}^R(M)$, thermodiffusion $D_{ij}^{TR}(M)$, director $(\gamma^{-1})_{ij}^R(M)$ and magnetization relaxation $b_{ij}^R(M)$, are all of the form

$$\begin{aligned} \kappa_{ij}^R(M) = & \kappa_1^R \epsilon_{ijk} M_k + \kappa_2^R \epsilon_{ijk} n_k n_p M_p \quad (\text{B.1}) \\ & + \kappa_3^R (\epsilon_{ipq} M_p n_q n_j - \epsilon_{jpq} M_p n_q n_i) \end{aligned}$$

They are antisymmetric $\kappa_{ij}^R(M) = -\kappa_{ji}^R(M)$ according to Onsager's relation $\kappa_{ij}^R(M) = \kappa_{ji}^R(-M)$ and give zero entropy production.²²

The third rank tensor c_{ijk}^R describing dynamic crosscoupling between flow and magnetization is symmetric in the two last indices and reads

$$\begin{aligned} c_{ijk}^R(M) = & c_1^R M_i n_j n_k + c_2^R (\delta_{ij} M_k \\ & + \delta_{ik} M_j) \\ & + c_3^R M_i \delta_{jk} + c_4^R n_i M_p n_p \delta_{jk} \\ & + c_5^R (n_i M_j n_k + n_i M_k n_j) \\ & + c_6^R n_i M_p n_p n_j n_k \end{aligned} \quad (\text{B.2})$$

The reversible analogue of the viscosity tensor has eight components²²

$$\begin{aligned} \nu_{ijkl}^R(M) = & \nu_1^R [\epsilon_{imp} n_j n_k n_l + \epsilon_{jmp} n_i n_k n_l \\ & - \epsilon_{kmp} n_j n_i n_l - \epsilon_{lmp} n_j n_k n_i] n_p M_m \\ & + \nu_2^R [\epsilon_{jmp} n_l \delta_{ik} - \epsilon_{lmp} n_j \delta_{ik} \\ & \quad + \epsilon_{jmp} n_k \delta_{il} - \epsilon_{kmp} n_j \delta_{il} \\ & \quad + \epsilon_{imp} n_k \delta_{jl} - \epsilon_{kmp} n_i \delta_{jl} \\ & \quad + \epsilon_{imp} n_l \delta_{jk} - \epsilon_{lmp} n_i \delta_{jk}] n_p M_m \\ & + \nu_3^R [\epsilon_{kmp} n_l \delta_{ij} + \epsilon_{lmp} n_k \delta_{ij} \\ & \quad - \epsilon_{imp} n_j \delta_{kl} - \epsilon_{jmp} n_i \delta_{kl}] n_p M_m \\ & + \nu_4^R [\epsilon_{ikp} n_j n_l + \epsilon_{ilp} n_j n_k + \epsilon_{jlp} n_i n_k \\ & \quad + \epsilon_{jkp} n_i n_l] n_p n_m M_m \\ & + \nu_5^R [\epsilon_{ikp} n_j n_l + \epsilon_{ilp} n_j n_k \\ & \quad + \epsilon_{jlp} n_i n_k + \epsilon_{jkp} n_i n_l] M_p \\ & + \nu_6^R [\epsilon_{ikp} \delta_{jl} + \epsilon_{ilp} \delta_{jk} \\ & \quad + \epsilon_{jlp} \delta_{ik} + \epsilon_{jkp} \delta_{il}] n_p n_m M_m \\ & + \nu_7^R [\epsilon_{ikp} \delta_{jl} + \epsilon_{ilp} \delta_{jk} \\ & \quad + \epsilon_{jlp} \delta_{ik} + \epsilon_{jkp} \delta_{il}] M_p \\ & + \nu_8^R [\epsilon_{ikp} (M_j n_l + M_l n_j) \\ & \quad + \epsilon_{ilp} (M_j n_k + M_k n_j) \\ & \quad + \epsilon_{jlp} (M_i n_k + M_k n_i) \\ & \quad + \epsilon_{jkp} (M_i n_l + M_l n_i)] n_p \end{aligned} \quad (\text{B.3})$$

This fourth order tensor is antisymmetric in the exchange of the first pair of indices with

the second one, thus guaranteeing zero entropy production.

The dissipative analogue of the flow alignment tensor λ_{ijk}^D is symmetric w.r.t. the 2 last indices and transverse to \mathbf{n} in the first index

$$\begin{aligned} \lambda_{ijk}^D(M) &= \lambda_1^D(\delta_{iq}^{tr}\epsilon_{pj}M_p n_k + \delta_{iq}^{tr}\epsilon_{pk}M_p n_j) \\ &+ \lambda_2^D(\delta_{ik}^{tr}M_p \epsilon_{pj}n_q + \delta_{ij}^{tr}M_p \epsilon_{pk}n_q) \\ &+ \lambda_3^D(M_j \epsilon_{ipk}n_p + M_k \epsilon_{ipj}n_p) \\ &+ \lambda_4^D(M_q n_q n_j \epsilon_{ipk}n_p + M_q n_q n_k \epsilon_{ipj}n_p) \\ &+ \lambda_5^D M_p \epsilon_{piq}n_q n_j n_k + \lambda_6^D M_p \epsilon_{piq}n_q \delta_{jk}^{tr} \end{aligned} \quad (\text{B.4})$$

The dissipative second rank tensor χ_{ij}^D is different from the reversible ones (B.1), since it has to contain an odd number of \mathbf{n} 's

$$\chi_{ij}^D(M) = \chi_1^D \delta_{ik}^{tr} M_k n_j + \chi_2^D \delta_{ij}^{tr} M_k n_k \quad (\text{B.5})$$

Such a coupling does not exist in isotropic ferrofluids, nor in conventional nematics.

In the biaxial case some new additional terms cubic in \mathbf{M} can be produced in (B.2-B.5) by replacing any pair $n_i n_j$ with $M_i M_j$, e.g. δ_{ij}^{tr} by $\delta_{ij}^3 = \delta_{ij} - n_i n_j - M_i M_j$ and $M_i M_j$ terms.

Appendix C Nonlinear magnetization

In the general case the equilibrium value of the magnetization \mathbf{M}^0 is a nonlinear function of an external magnetic induction \mathbf{B} , $M_i^0 = f_i(\mathbf{B})$. In isotropic ferrofluids simple theoretical models predict $\mathbf{f}(\mathbf{B}) = f(B)\mathbf{B}/B$ with $f(B)$ the Langevin function and $B = |\mathbf{B}|$. Actual experiments²⁶ and computer simulations²⁷ show a somewhat more complicated form for $f(B)$. We assume the function $\mathbf{f}(\mathbf{B})$ as known and given. To our hydrodynamic theory this is an input as are any other aspects of the equilibrium structure, e.g. the existence and the orientation of \mathbf{n} w.r.t the external field. Assuming $\mathbf{f}(\mathbf{B})$ to be monotonous we can invert it and call it $\mathbf{g}(\mathbf{M}) = \mathbf{f}^{-1}(\mathbf{B})$. Using the same arguments as in Sec.(2) one can construct the

magnetic part of the energy density in the following form

$$\begin{aligned} \varepsilon(\mathbf{B}, \mathbf{n}, \mathbf{M}) &= \frac{B^2}{2} - \mathbf{M} \cdot \mathbf{B} \\ &+ \int (n_i n_j g_j^{(1)} + \delta_{ij}^{tr} g_j^{(2)}) dM_i \end{aligned} \quad (\text{C.1})$$

where, in contrast to the isotropic case, g_i has been decomposed into a longitudinal and a transverse part. This leads to

$$H_i = \left(\frac{\partial \varepsilon}{\partial B_i} \right)_{\mathbf{M}, \mathbf{n}, \dots} = B_i - M_i \quad (\text{C.2})$$

$$\begin{aligned} h_i^M &= \left(\frac{\partial \varepsilon}{\partial M_i} \right)_{\mathbf{n}, \mathbf{B}, \dots} \\ &= -B_i + n_i n_j g_j^{(1)} + \delta_{ij}^{tr} g_j^{(2)}. \end{aligned} \quad (\text{C.3})$$

In equilibrium the magnetic molecular field h_i^M is zero and $\mathbf{B} = \mathbf{g} \parallel \mathbf{n}$ (requiring $n_i n_j g_j = g_i^{(1)}$ and $\delta_{ij}^{tr} g_j^{(2)} = 0$ in equilibrium). Outside equilibrium $g_i(\mathbf{M}) \neq B_i$ and \mathbf{n} is not parallel to \mathbf{B} , generally.

For the nematic molecular field

$$\begin{aligned} h_i^{n'} &= \delta_{ij}^{tr} \left(\frac{\partial \varepsilon}{\partial n_j} \right)_{\mathbf{M}, \mathbf{B}, \dots} \\ &= \delta_{ij}^{tr} n_k \int (dM_j (g_k^{(1)} - g_k^{(2)}) \\ &\quad + dM_k (g_j^{(1)} - g_j^{(2)})) \end{aligned} \quad (\text{C.4})$$

is found. Linearizing about equilibrium the choices $g_i^{(1)} = (A_1 + A_2)n_i n_j M_j$ and $g_i^{(2)} = A_2 \delta_{ij}^{tr} M_j$ lead back to eqs.(4,5).

References

- ¹ F. Brochard and P.G. de Gennes, *J. Phys.(France)* **31**, 691 (1970).
- ² R.E. Rosensweig, *Ferrohydrodynamics*, Cambridge University Press, Cambridge (1985).
- ³ V. Berejnov, J.-C. Bacri, V. Cabuil, R. Perzynski and Y.L. Raikher, *Europhys. Lett.* **41**, 507 (1998).

- ⁴ S.-H. Chen and N.M. Amer, *Phys. Rev. Lett.* **51**, 2298 (1983).
- ⁵ M. Koneracká, V. Závášová, P. Kopčanský, J. Jadżyn, G. Czechowski, and B. Żywuski, *J. Magn. Magn. Mater.* **157/158**, 589 (1996).
- ⁶ J. Rault, P.E. Cladis, and J.P. Burger, *Phys. Lett. A* **32**, 199 (1970).
- ⁷ S.S. Chen and S.H. Chiang, *Mol. Cryst. Liq. Cryst.* **144**, 359 (1987).
- ⁸ C.F. Hayes, *Mol. Cryst. Liq. Cryst.* **36**, 245 (1976).
- ⁹ L. Liébert and A. Martinet, *J. Physique (France) Lett.* **40**, 363 (1979).
- ¹⁰ L.J. Yu and A. Saupe, *Phys. Rev. Lett.* **45**, 1000 (1980).
- ¹¹ Y. Galerne and J.P. Marcerou, *Phys. Rev. Lett.* **51**, 2109 (1983).
- ¹² J.C. Bacri and A.M.F. Neto, *Phys. Rev.* **E50**, 3860 (1994).
- ¹³ S. Fontanini, A.L. Alexe-Ionescu, G. Barbero, and A.M.F. Neto, *J. Chem. Phys.* **106**, 6187 (1997).
- ¹⁴ J. Yamamoto and H. Tanaka, *Nature* **409**, 321 (2001).
- ¹⁵ V.V. Berejnov, Y. Raikher, V. Cabuil, *J. Colloid Interface Sci.* **199**, 215 (1998).
- ¹⁶ S.V. Burylov and Y.L. Raikher, *Mol. Cryst. Liq. Cryst.* **258**, 107 (1995).
- ¹⁷ H. Pleiner, E. Jarkova, H.-W. Müller, and H.R. Brand, *Magnetohydrodynamics* **37**, 254 (2001).
- ¹⁸ C.Y. Matuo and A.M.F. Neto, *Phys. Rev.* **E60**, 1815 (1999).
- ¹⁹ D. Spoliansky, J. Ferré, J.-P. Jamet, and V. Ponsinet, *J. Magn. Magn. Mater.* **201**, 200 (1999).
- ²⁰ M.I. Shliomis, *Sov. Phys. Usp.* **17**, 153 (1974).
- ²¹ H.-W. Müller, M. Liu, *Phys. Rev.* **E64**, 061405 (2001).
- ²² E. Jarkova, H. Pleiner, H.-W. Müller, A. Fink, and H.R. Brand, *Eur. Phys. J.* **E5**, 583 (2001).
- ²³ M. Liu, *Phys. Rev.* **E50**, 2925 (1994).
- ²⁴ In lyotropic systems one could also take into account additionally the concentration of the solvent c_S without changing the major results, since it has the same transformation behavior as c and thus makes the same type of coupling terms.
- ²⁵ H. Pleiner and H.R. Brand, Hydrodynamics and Electrohydrodynamics of Nematic Liquid Crystals, in *Pattern Formation in Liquid Crystals*, eds. A. Buka and L. Kramer, Springer, New York, p. 15 (1996).
- ²⁶ A.F. Pshenichnikov, *J. Magn. Magn. Mat.* **145**, 319 (1995).
- ²⁷ Z.W. Wang, C. Holm, H.-W. Müller, *Phys. Rev.* **E**, submitted, (2002).
- ²⁸ P.C. Martin, O. Parodi, and P.S. Pershan, *Phys. Rev.* **A6**, 2401 (1972).
- ²⁹ P.G. de Gennes and J. Prost, *The Physics of Liquid Crystals* (Clarendon Press, Oxford, 1993).
- ³⁰ H.-W. Müller and M. Liu, *Phys. Rev. Lett.* **89**, 67201, (2002); H.-W. Müller, Y. Jiang, and M. Liu, *Phys. Rev.* **E67**, 03101 (2003). (2002).
- ³¹ H.R. Brand and H. Pleiner, *Phys. Rev.* **A24**, 2777 (1981).